

Per- and Polyfluoroalkyl Substances Draft Chemical Action Plan

Draft for Public Comment

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Draft for Public Comment

Hazardous Waste and Toxics Reduction Program
Washington State Department of Ecology
Olympia, Washington



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Executive Summary

Per- and polyfluoroalkyl substances (PFAS) are a family of more than 4,700 synthetic organic chemicals. PFAS can withstand high temperatures and survive highly corrosive environments. They are used in the manufacture of coatings, surface treatments, and specialty chemicals in cookware, carpets, food packaging, clothing, cosmetics, and other common consumer products. PFAS also have many industrial applications and are an active ingredient in certain types of fire-fighting foams (aqueous film-forming foams, or AFFF). PFAS coatings resist oil, grease, and water.

A Chemical Action Plan (CAP) identifies, characterizes, and evaluates uses and releases of a specific Persistent Bioaccumulative Toxin (PBT), a group of PBTs, or metals of concern, and recommends actions to protect human health or the environment.

The Departments of Ecology (Ecology) and Health (Health) (jointly "we") developed this Draft PFAS CAP (Draft CAP) to recommend actions to address PFAS in the environment and resulting human impacts. This Draft CAP builds on work that started in 2016, when we convened an advisory committee to inform and guide our PFAS CAP development work. In April of 2018, we issued an Interim CAP for PFAS (Interim CAP), recommending actions to address problems with PFAS. The Interim CAP was updated in <u>January 2019</u>. Following additional input by the Advisory Committee, we issued <u>Preliminary CAP Recommendations</u> (Preliminary Recommendations) in May 2019. These recommendations also took into account the Washington State Legislature's 2018 adoption of laws that impact PFAS use in firefighting foam and food packaging in the state.

This Draft CAP was informed by Advisory Committee input received in July 2019 and updated based on new information available about PFAS since May 2019. We are issuing the Draft CAP for public comment; we will then prepare a Final CAP after considering the comments we receive.

Why are we concerned about PFAS?

PFAS use leads to persistent perfluorinated breakdown products in our environment. PFAS are used in many applications for consumer, commercial, and industrial products. For most products, little is known about the specific PFAS and amounts they contain, or the potential to expose humans or the environment during production, use, and disposal. Many PFAS—such as those used for firefighting foam—degrade in the environment to form perfluoroalkyl acids (PFAAs). No known natural mechanisms can break these PFAAs down. Some places PFAS have been detected in Washington include surface waters, groundwater, wastewater treatment

² https://fortress.wa.gov/ecy/publications/documents/1804005.pdf

³ https://www.ezview.wa.gov/Portals/_1962/Documents/PFAS/PrelimRecommendations-2019-PFAS-CAP.pdf

plant (WWTP) effluent, freshwater and marine sediments, freshwater fish tissue, and osprey eggs. Any toxic or other hazardous effects of these chemicals will be with us for many decades.

Nearly everyone in Washington is likely exposed to PFAS. National surveys show that most people tested have some PFAS in their blood. Many sources lead to exposure. Workers in jobs related to PFAS-containing products have the highest exposures. People consuming PFAS-contaminated drinking water or food can also be highly exposed. For most people, exposure occurs through food, drinking water, and contact with things like disposable packaging or treated textile products, to name a few.

Some PFAS are bioaccumulative. Bioaccumulation of PFAS has been confirmed in marine and terrestrial species, zooplankton and other invertebrates, and fish. Animals do not need to be near sources of PFAS to show bioaccumulation. PFAS have also been shown to be taken up by plants. Long-chain PFAS tend to be more bioaccumulative. Some PFAS are known to bioaccumulate in people because they are readily absorbed following ingestion, resist metabolic breakdown, and are poorly excreted from the human body.

Some PFAS show harmful effects to wildlife and to people. Animal studies show strong evidence of the association between exposure to several PFAAs and developmental, liver, and immune toxicity. Epidemiological studies suggest links between PFAA exposure and several negative health outcomes in human beings, including increases in cholesterol levels, immune suppression, and lower birthweights. Higher exposures have also shown associations with some cancers, such as testicular and kidney cancers.

Replacement products are still poorly understood. U.S. manufacturers have ceased manufacturing and using long-chain PFAS, such as perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA), with the exception of certain specialty uses. Certain short-chain PFAS used as replacements may be lower in toxicity and bioaccumulation, but their exposure and toxicity characteristics are still being studied. Short-chain replacements can be more mobile in the environment and just as persistent long-term, resulting in potentially expensive remediation should they be confirmed harmful to wildlife and humans.

Responding to PFAS contamination is expensive and requires cross-agency coordination.

When PFAS concentrations in drinking water supplies exceed health advisory levels, timely mitigation is needed to protect human health. Without identified funding, public water systems and their ratepayers must absorb expensive response costs. Multiple local, state, and federal agencies may be involved in investigating and responding to a drinking water contamination event.

Recommendations for action

We are recommending actions to address PFAS contamination of the environment and the resulting potential impacts to animal and human health. We base these recommendations on our assessment of scientific information available regarding the behavior of PFAS.

1.0 Ensure drinking water is safe

1.1 Identify funding for PFAS drinking water mitigation

When PFAS concentrations in drinking water supplies exceed health advisory levels, timely mitigation may be needed to protect human health. This can create immediate costs to water systems. Without funding, public water systems and their ratepayers must absorb what can be a costly response. Lower-income and overburdened communities are less able to absorb unplanned ratepayer cost increases when PFAS contamination of their water supply is identified. Funding supports water systems as they explore ways to mitigate the problem, both immediately and long-term, and must:

- Investigate contamination sources.
- Find an alternative water source.
- Design and install expensive filters on contaminated water sources.
- Maintain and monitor new filtration systems.

Recommendation

State agencies, the Washington State Legislature, and local water systems should work together to fund PFAS drinking water mitigation. These costs should be reimbursed by responsible parties under applicable laws. Once PFAS water contaminants are officially classified as hazardous substances by the federal government or by the state of Washington, they can be addressed under the state Model Toxics Control Act (MTCA) framework.

Drinking Water State Revolving Fund is a U.S. Environmental Protection Agency (EPA)-funded loan program administered by Health. The loans are used to:

- Improve drinking water infrastructure.
- Finance the cost of installing treatment or other infrastructure improvements over a number of years.

Drinking Water State Revolving Fund can provide emergency loans in the event a water system is issued a "Do Not Use" order as a result of PFAS contamination. The program recently funded a reservoir project for City of Spokane to allow Spokane to provide reliable water service to Airway Heights. Airway Heights has PFAS in their wells and is now relying on City of Spokane for its water.

EPA provides funding to Health Office of Drinking Water for set-aside activities and source water protections. Health can use these funds in limited circumstances to defray costs of additional water testing.

Other funding programs in the state could be tapped for loans or grants to help with costs of new infrastructure in response to PFAS contamination:

- Public Works Assistance Account overseen by Public Works Board.
- Community Development Block Grant overseen by Department of Commerce.
- Rural Development loans and grants overseen by U.S. Department of Agriculture.

Public water systems can pursue reimbursement from potentially liable parties under the state MTCA when PFAS are declared hazardous substances under MTCA. Even under MTCA, water systems may have to carry costs long-term or permanently because:

- The process of identifying responsible parties and being reimbursed can take years.
- Responsible parties may be difficult, if not impossible, to determine.
- The potentially liable party could be a local entity under the same public administration as the water utility (for example, a local fire station).

In each of these cases, the costs borne by the water system would be long-term or permanent.

Cost

Initial investigation and mitigation costs at PFAS-contaminated sites are reported in the millions of dollars. These costs have been borne by the U.S. Department of Defense (DOD), the water systems or local governments impacted, and the agency programs at Health and Ecology that support water systems and contaminated site cleanup.

For example, the Issaquah PFAS Pilot Project received \$400,000 through the State Building Construction Account for groundwater assessment work to be conducted during the 2019 – 2021 biennium. An additional \$300,000 was requested for this project from the 2020 program operating budget, but these funds are not yet available as a result of COVID-19—related impacts to Ecology's operating budget. In future years, 2021 – 2023, additional resources of \$750,000 are being requested for additional groundwater investigation.

With mitigation measures identified, implementation funds are sought from granting sources as described above. In addition to costs for investigating the source of the contamination, filter maintenance and monitoring also require ongoing expenditures. Such costs could also be covered under grants, but may require additional resources from water supply systems.

Each contaminated drinking water site has specific needs, which complicates cost estimation. Without knowing the number of impacted systems in the state, we are unable to estimate total costs to implement this recommendation.

1.2 Provide technical support for site characterization, source investigation, and mitigation at contaminated sites

Local water districts and governments often lack the expertise and resources to investigate sources of PFAS contamination. Technical assistance helps them understand the advantages and disadvantages of various options to reduce levels of PFAS in water and soil. Appropriate actions are informed by site-specific conditions and a knowledge of evolving drinking water treatments and cleanup methods. Research into the unusual properties of PFAS will inform mitigation as replacement PFAS products also make their way into the environment. To recover mitigation costs, Ecology must identify the party or parties responsible for the source of contamination.

Recommendation:

Ecology and Health will continue to develop expertise and provide technical assistance and guidance to drinking water purveyors, local jurisdictions, and responsible parties in order to address PFAS contamination and conduct cleanup actions.

Those actions include:

- Ecology will continue to collaborate with involved parties at PFAS contamination sites in the state. These efforts will help to better understand the sources, composition, and distribution of PFAS contamination in soil and water. Identification and evaluation of appropriate cleanup actions and their costs will be informed by this work. This work is being done within Ecology's existing resources.
- Health will continue to provide water systems with advice and assistance to
 understand the mitigation options and guide voluntary action on unregulated PFAS
 until the rulemaking for PFAS in drinking water is complete. To-date, technical
 assistance has focused on public water systems near military bases with PFAS
 detections in groundwater. Department of Health continues to include local health
 departments in outreach and guidance. This work is being done within Health's
 existing resources.
- Ecology will look at targeting Safe Drinking Water Action Grants (a category of Remedial Action Grants for Local Governments) to help address PFAS-contaminated drinking water once some of the uncertainties discussed above have been addressed.
- Ecology will investigate PFAS contamination in groundwater and surface water.
 These efforts would support local health departments, cities, counties, and other public entities in Washington when PFAS contamination is discovered. Initial investigation efforts could identify areas at high risk of contamination. This could include areas where trainings or firefighting activities used large quantities of PFAS-containing AFFF, or where spills released the foam. Ecology could prioritize funding for site-specific assessments and groundwater testing. Funding for this action is estimated below.
- Ecology will consider the number of people impacted, the concentration of the PFAAs in the drinking water, and vulnerable populations present when prioritizing mitigation and cleanup activities. Ecology will use mapping tools such as

- Environmental Justice (EJ) screen and Information by Location (IBL) in the Washington Tracking Network (WTN) portal to characterize the demographics of the population served by impacted drinking water.
- We will obtain chemical identities from products and at contaminated sites to find chemical "fingerprints" useful in identifying source locations. Analytical methods may not yet be developed to obtain all the required data.

Cost

To support PFAS investigations as needed, Ecology requested resources from the Legislature to:

- Provide monitoring assistance to local jurisdictions when PFAS contamination is discovered.
- Assist with investigations, including researching potential sources, collecting samples, conducting laboratory analysis, and installing monitoring wells.

This type of environmental monitoring work was funded in 2020 and 2021 through the approved state 2019 – 2021 supplemental budget.⁴

1.3 Support biomonitoring and other health studies to answer important health questions

Biomonitoring can help us understand the best way to reduce human exposure to PFAS. Biomonitoring helps people compare their PFAS exposure level to national averages, and could connect residents to health information as it becomes available.

Recommendation:

Health should continue to find opportunities for Washington residents to participate in exposure and health studies. These studies help answer important community and public health questions about PFAS exposure and health outcomes. For example, Health requested and supports inclusion of Airway Heights as one of eight sites in the Agency for Toxic Substances and Disease Registry's (ATSDR) PFAS Exposure Assessment study. Health also applied for but was not awarded a cooperative agreement to include a Washington site in the ATSDR Multisite PFAS Health Study.

State agencies should also support investigations into pathways of PFAS contamination in food, drinking water, and indoor environments. They should pursue policies to mitigate and reduce these sources of human exposure over time.

Cost

Biomonitoring studies are expensive and the state would need funding to support these types of investigations. Additional funding could be secured through competitive grants for such activities. Benchmark costs have been estimated based on reports from several sites in the U.S.

⁴ https://ofm.wa.gov/sites/default/files/public/budget/statebudget/20supp/Z-0776.2Operating.pdf

where biomonitoring testing has been conducted for residents near areas of PFAS contamination. Costs averaged up to \$1,000 per person tested.

2.0 Manage environmental PFAS contamination

Ecology establishes cleanup levels for hazardous substances in the environment. The cleanup level concentrations, under specific exposure conditions, are considered sufficiently "protective of human health and the environment." Currently, no enforceable federal or Washington state regulatory standards exist to determine whether a site with PFAS contamination requires cleanup or to regulate cleanup of PFAS at contaminated sites. Further, best practices for conducting such a cleanup are not established.

To support PFAS groundwater contamination investigation in the City of Issaquah, Ecology developed investigatory levels for PFOS and PFOA. These were advisory values, not regulatory cleanup levels.

Ecological receptors contribute to Washington state's health and economy overall. Collecting additional data and extending cleanup levels to other environmental media is also crucial to protecting them.

2.1 Establish PFAS cleanup levels for soil and groundwater

Recommendation

- Using existing authority under MTCA, Ecology will develop cleanup levels for PFOA and PFOS (and additional PFAS as appropriate). Ecology will use the State Board of Health's (SBOH) drinking water standards or other advisories adopted in rule to develop these cleanup levels.
- Ecology will explore methods for investigation and cleanup of PFAS contamination.
- Ecology will conduct monitoring for PFAS compounds in environmental media (soils, surface water, and sediment) and wildlife tissue to identify sources and assess exposure.
- Based on data collected, Ecology will consider developing cleanup levels for individual or mixtures of PFAS in soil, sediment, freshwater, and saltwater to protect ecological receptors.
- In this context, the following activities will be implemented to support activity under the recommendations above:
 - Trophic transfer and bioaccumulation of PFAS compounds should be further evaluated in aquatic and terrestrial food webs to further understand exposure.
 - Selected individual PFAS compounds, as well as common PFAS mixtures, should be evaluated for ecotoxicity in aquatic and terrestrial biota, using both laboratory and field methods.
 - Ecological risk assessment should be performed for PFAS compounds by detailing exposure and effects in order to estimate risks to nonhuman biota.

- An uncertainty analysis should accompany PFAS ecorisk assessment to promote transparency in the risk assessment and communication processes and to more clearly identify data gaps.
- Results of these risk assessments should support potential interventions (for example, species protections) and characterization of potential impacts on ecological services.
- Ecology will provide information to interested parties about cleanup efforts.

Cost

The cost to develop cleanup standards is being funded out of Ecology's Toxics Cleanup Program operating budget, and is expected to be approximately \$42,000 based on the cost of developing advisory levels. This estimate does not include work to collect additional exposure data, nor to develop cleanup levels for other environmental media (sediment and surface water).

Costs to develop and evaluate methods for addressing PFAS contamination are difficult to estimate due to significant uncertainties around:

- How (and in what concentrations) most PFAS affect people, animals, and plants.
- How best to measure the types and amounts of PFAS in the environment.
- How PFAS move through the environment and change over time.
- How to effectively clean up environmental PFAS contamination—including factors like protectiveness, feasibility, and cost.

Ecology is planning to conduct additional environmental monitoring in 2020 and 2021 funded through the approved state 2020 supplemental budget,⁵ however specific projects have not yet been selected.

2.2 Partner with local organizations in communities with contaminated water or contaminated sites

When testing identifies PFAS-contaminated drinking water in a new community, it can be challenging to communicate effectively with area residents.

Communities are unique, and there may be:

- Cultural and language barriers to effective communication.
- Economic, systemic, and social barriers to act on public health advice.

These barriers disproportionately affect low-income and other historically overburdened communities, including communities of color. During PFAS investigation and mitigation, state agencies should collaborate with local leadership and organizations to strengthen community awareness and engagement.

Community-based and community-led organizations (that are rooted in and directly serve these communities) can offer meaningful engagement support. For example:

⁵ https://ofm.wa.gov/sites/default/files/public/budget/statebudget/20supp/Z-0776.2Operating.pdf

- A recent \$120,000 two-year grant funded a local organization providing educational materials and conducting outreach in a community impacted by industrial activities.
- In one affected community, a local church group volunteered to distribute bottled water to elderly and disabled residents.

Recommendation

Department of Health will identify local health departments or community-based organizations to address health equity related to contaminated sites in public communications. Health will coordinate with Ecology to distribute funding to those organizations selected for assistance. Health's new Community Engagement Guide⁶ may support this effort.

Funded organizations would:

- Address potential health equity issues through culturally and linguistically informed engagement.
- Find trusted messengers or platforms to deliver audience-tested risk communication messages to engage historically overburdened and higher risk populations.
- Support impacted populations in finding their own solutions through collective action and decision-making.
- Engage the community throughout the course of the public health response, source investigation, and site cleanup.
- Invite area residents to actively participate on advisory committees, in site information meetings, and in public decision-making about remediation.
- Aim to remove participation barriers by providing child care, reducing transportation costs, and planning for convenient meetings times at familiar locations.
- When possible, appropriately compensate community advisors for participation—particularly in areas with low-income populations.

Cost

If PFAS are classified as hazardous substances under MTCA, community-led public engagement would be eligible for funding through Ecology's Public Participation Grant program (in the Contaminated Site Project category). Designated PFAS funds should be allocated specifically to PFAS-related impacts to communities.

Local outreach efforts depend on the extent and type of community outreach required for a specific contamination concern. As such, at this time, it is not possible to estimate the funding needed for these efforts.

2.3 Work to prevent PFAS releases from firefighting foam use and manufacturing

PFAS-containing Class B firefighting foam has been associated with drinking water contamination in Washington state. In their risk-based efforts to identify and mitigate PFAS in drinking water, both the military and Health focused on firefighting foam release sites.

⁶ https://www.doh.wa.gov/Portals/1/Documents/1000/CommEngageGuide.pdf

However, firefighting foam is not the only likely source of PFAS in state drinking water. Other states that are expanding testing for PFAS in drinking water have identified sources such as:

- Landfills.
- Industrial waste stream sludge.
- Manufacture of waterproof leather shoes.
- Manufacture of parchment paper.
- Taxidermy.
- Textile coating.
- Metal plating and finishing.
- Car washes.
- Pulp and paper mills.

More work is needed to understand PFAS use, sources, pathways of exposure, and effects on human health and the environment.

Recommendation

Ecology will continue to work proactively with industry, manufacturers, and businesses to eliminate releases to the environment from the use of PFAS-containing AFFF or other manufacturing processes using PFAS.

To address PFAS in AFFF, Ecology would continue implementing the Firefighting Agents and Equipment Toxic Chemical Use law (Chapter 70.75A Revised Code of Washington [RCW]⁷), as follows:

- Collaborate with firefighting foam users to develop and share outreach materials and best management practices that address the proper use, storage, and disposal of PFAS-containing AFFF.
- Ensure that industrial use of PFAS-containing AFFF provides for containment procedures along with collection of this foam and contaminated soil or sediment for proper designation and disposal. Costs to industrial users to collect and dispose of released PFAS-containing AFFF include plan development, employee training, methods for containment, and disposal of waste.
- Continue identifying organizations and industries that store and use AFFF in training and emergency firefighting, including the use of AFFF in tunnels.
- Assist state and local governments, airports, industry, and fire districts with prioritizing the quantification, disposal, and replacement of PFAS-containing AFFF, especially in communities with cumulative impacts, health disparities, and environmental justice considerations.
- Share information about PFAS-free Class B firefighting foam with firefighting foam users as information or research is available, including GreenScreen® certifications.
- Conduct compliance and enforcement actions to ensure the law is being followed.

⁷ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

Ecology will work proactively with industry, manufacturers, and businesses to eliminate releases to the environment from PFAS use in manufacturing or other processes.

- Ecology will review data from other states and countries to identify industrial or manufacturing uses of PFAS. Ecology will use this information to identify industries in Washington that have used or continue to use commercial quantities of PFAS. Ecology will also track future TRI reports (starting 2021) for industries.
- Ecology will evaluate PFAS release potential from those industries which may have, or continue to, use PFAS.
- Ecology will reach out to these industries to discuss their use of PFAS, identify opportunities to switch to safer alternatives, implement best practices, and ensure proper waste management.

Cost

Ecology identified additional foam stockpiles managed by commercial airports, manufacturing, and transportation facilities that represent a large pollution source, but do not currently qualify for the disposal program established under Chapter $70.75A^8$ RCW. Ecology estimates that it will cost between \$400,000 and \$650,000 to collect, transport, and dispose of such foam, including 0.25 full-time equivalent (FTE) to manage this program. Ecology has included this cost in its fiscal year (FY) 2021 – 2023 budget request.

Ecology has requested approximately \$36,000 for monitoring and compliance activities to be conducted under Chapter 70.75A RCW in FY 2021 – 2023.

Ecology estimates that support to industry to investigate and support reduction of non AFFF-related PFAS use would require the resources of 0.25 FTE for one year, at the cost of approximately \$50,000. This funding has not yet been budgeted or requested.

3.0 Reduce PFAS in products

People are exposed to PFAS in their homes when they use products, and via exposure to house dust that contains PFAS. Ingesting contaminated food and drinking water leads to the greatest portion of chronic exposure to PFAS (specifically to PFOS and PFOA) for the general population.

According to EPA, some of the most significant sources of human exposure to nine PFAS in the U.S. are carpets and commercial carpet-care liquids, which contribute to PFAS in residential and commercial indoor environments. Infants and children have higher exposure due to inhalation and ingestion of house dust. High PFAA levels were also identified in ski waxes, leather samples, outdoor textiles, and some baking papers.

Actions need to be implemented to remove or reduce levels of PFAS from products that contribute to human or environmental exposure. Removing chemicals from consumer products can reduce chemicals in indoor air and dust. These actions directly impact human and

⁸ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

environmental exposures. Research is needed to understand how these products contribute to human exposure.

3.1 Reduce PFAS exposure from carpets and rugs, water and stain resistance treatments, and leather and textile furnishings

Recommendation

Under Chapter 70.365⁹ RCW, Ecology identified carpets, water and stain resistance treatments, and leather and textile furnishings as significant sources and uses of PFAS. We recommend that Ecology determine whether safer alternatives are feasible and available. These determinations should be made by June 2022 and would be accompanied by proposed regulatory actions to reduce exposure.

Regulatory actions could include:

- Requesting that manufacturers:
 - Identify products that contain PFAS.
 - o Disclose their use of priority chemicals in product ingredients.
 - o Release information on exposure and chemical hazard.
 - Describe the amount and function of PFAS in products.
- Proposing restrictions on PFAS-containing carpet and carpet care products when a safer alternative is available and feasible, and the restriction will reduce a significant source of PFAS or protect sensitive populations or species.
- Proposing actions to reduce legacy PFAS-containing carpet and carpet care products remaining in homes, especially in low-income households, where items may be retained past the typical product lifespan.

In addition to the above, we recommend the following actions:

- Implement a purchasing preference policy for PFAS-free carpet. Work with vendors on the state flooring contract to offer PFAS-free carpet on all state master contracts and all agency contracts. Purchasing PFAS-free carpet could result in increased costs to the state.
- If safer alternatives are available, include them in Ecology's <u>Product Replacement Program</u>¹⁰ to replace PFAS-containing carpet in community centers, libraries, daycares, and other environments where children may be disproportionately exposed.

Cost

The Legislature funds these efforts under the Safer Products for Washington program. Ecology received \$807,000 for the 2019 – 2021 biennium, and a supplemental \$479,000 in FY 2020 to implement the program as a whole, for a total of \$1,286,000 through June 2021. As described in its July 2020 report to the legislature, Ecology identified eleven priority products, three of

⁹ https://app.leg.wa.gov/rcw/default.aspx?cite=70.365

¹⁰ https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Product-Replacement-Program

which were PFAS related (carpets, water and stain resistance treatments, and leather and textile furnishings).

Because Ecology conducts program activities as a whole, it is not possible to distinguish program costs attributed to only the PFAS-related priority products. However, one could approximate the PFAS-related costs as a proportion of entire program costs based on the number of priority products identified, i.e. three of eleven. Thus, the cost of activities associated with identifying PFAS-related priority products under Chapter 70.365 RCW would be approximately \$351,000.

A portion of the cost estimated above will also go toward work completed by the end of June 2021 related to proposing regulatory actions. Ecology will request additional funding to determine proposed regulatory actions by the end of June 2022, as described in Recommendations 3.2 and 3.3 below.

At this time, Ecology has not estimated the cost of additional actions (i.e., implementing a purchasing preference policy and replacing PFAS-containing carpet under the Product Replacement Program). Ecology is already funding a staff position to coordinate the identification of viable purchasing preference policies with the Washington State Department of Enterprise Services for a number of products, including PFAS-containing carpet.

Establishing the cost of replacing carpet in community centers, libraries, daycares, and other environments where children may be disproportionately exposed would require an estimate of the number of facilities targeted, and the square footage of carpet to be replaced. Funding could then be requested by Ecology's Product Replacement Program.

3.2 Identify additional sources and uses of PFAS to consider in the second Safer Products for Washington cycle

The priority products identified in 2020 under the Safer Products for Washington program do not account for all sources and uses of PFAS. We will continue research to better understand how other products contribute to PFAS concentrations in homes, workplaces, and the environment. These include PFAS in:

- Water resistant clothing and gear.
- Nonstick cookware and kitchen supplies (e.g. baking paper).
- Personal care products (including cosmetics and dental floss).
- Cleaning agents.
- Automotive products.
- Floor waxes and sealants.
- Ski waxes.
- Car waxes.

Recommendation

Ecology should engage with overburdened communities regarding consumer products that may contain PFAS. Communities use consumer products differently. It is important that the team identify consumer products which might be disproportionately exposing overburdened communities.

In addition to determining whether these products are significant sources or uses of PFAS, preliminary investigations into the availability and feasibility of safer alternatives should be conducted. This will help the Safer Products for Washington team identify opportunities to reduce additional sources and uses of PFAS. If safer alternatives are identified, outreach should be conducted to increase voluntary adoption. If these products are identified as significant sources or uses, they should be added as priority products in 2025 and regulatory actions should be determined by 2027.

Cost

Ecology will make budget requests to fund future cycles of the Safer Products for Washington Program, including consideration of the products listed above.

Ecology estimates that the costs of future Safer Products for Washington cycles would be similar to those incurred to-date (see <u>Recommendation 3.1</u> above), but could vary based on the complexity and the number of additional chemical-product combinations considered. The cost of identifying and implementing regulatory actions in future cycles is expected to be similar to those identified in Recommendation 3.3 below.

3.3 Implement other reduction actions for PFAS in products

Recommendation

Identify regulatory actions to reduce exposures and releases to the environment from the priority consumer products. Additional research, testing, and investigation would be required to determine appropriate reduction actions. Actions could include:

- Under Chapter 70.365¹¹ RCW, propose restrictions of PFAS in priority consumer products when a safer alternative is feasible and available, and the restriction will reduce a significant source of PFAS or protect sensitive populations or species. Ecology could:
 - Determine that no regulatory action is currently required.
 - o Require a manufacturer to provide notice of the use of PFAS.
 - Restrict or prohibit the manufacture, wholesale, distribution, sale, or use of PFAS
 in a priority consumer product.
 - Adopt rules to implement these regulatory actions.

¹¹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365

In addition to the above, we recommend the following actions:

- Gather input from low-income and other historically overburdened communities, including communities of color. Develop a list of ways to reduce exposure that include low cost and subsidized approaches. These may be particularly important measures to employ in communities with higher exposure from drinking water. No cost estimate is provided to conduct this evaluation or to develop exposure reduction recommendations.
- Establish a purchasing preference policy for PFAS-free products. Work with vendors
 to offer PFAS-free textiles, furniture, and paints. Apply this policy to all state master
 contracts and all agency contracts. Purchasing PFAS-free products could increase
 state costs.
- Propose a ban on the import or sale of all products in Washington containing phased-out long-chain PFAAs. Long-chain PFAAs include perfluorinated carboxylates (PFCAs) with eight or more fully fluorinated carbons (for example, PFOA) and perfluorinated sulfonates (PFSAs) with six or more fully fluorinated carbons (for example, perfluorohexane sulfonate (PFHxS) and PFOS), their salts, and precursor compounds capable of forming long-chain PFAAs.

Cost

The cost of selecting and implementing regulatory actions under Chapter <u>70.365</u>¹² RCW would be funded through the Safer Products for Washington program budget. This funding has not yet been received. As explained above, funding is provided for all priority products selected for regulation, included those that contain PFAS.

Ecology estimates costs of approximately \$1.5 million for this aspect of the program as a whole for the period July 2021 – June 2023. PFAS-related costs can be approximated as a proportion of entire program costs based on the number of individual priority products identified, i.e. three of eleven. On this basis, we estimate that regulatory actions for PFAS-related priority products identified in Recommendation 3.1 at approximately \$410,000.

4.0 Understand and manage PFAS in waste

Products people use in their homes and businesses can release PFAS. Waste streams generated in residential and commercial settings are treated in WWTPs or sent to disposal facilities such as landfills, which in turn can re-emit PFAS to the environment.

PFAS in municipal and industrial waste water entering WWTPs, may partition to different media (for example, solids and liquids) and transform into terminal PFAS compounds.

Decomposing domestic and industrial waste containing PFAS and rainfall can create leachate that contains PFAS released from disposed products. Older un-lined landfills can be a source of leachate released to groundwater. Leachate produced in lined landfills is typically transferred to

¹² https://app.leg.wa.gov/rcw/dispo.aspx?cite=70.365

WWTPs for further treatment. Both of these management methods have the potential to release PFAS to the environment.

Biosolids produced in WWTPs where PFAS are present can in turn be contaminated with PFAS. Fundamental PFAS concentration data to characterize Washington biosolids is lacking. Toxicity, concentration, and pathway of exposure determine the risks that PFAS in biosolids pose to human health and the environment.

4.1 Evaluate PFAS in wastewater treatment

Recommendation

Ecology should evaluate PFAS in WWTP influent and effluent to better understand PFAS discharges in Washington state.

- Ecology should develop a study design to sample PFAS in three different types of plants: WWTPs with secondary treatment, nutrient removal, and advanced solids removal. Sampling should include products of selected WWTP unit processes (for example primary and secondary clarifiers or dechlorination) to help differentiate removal efficiencies of the different treatment types.
- The study design should ensure that the sampled WWTPs either receive industrial discharges that are likely to contain PFAS or have drinking water sources with known PFAS contamination.
- Ecology should identify industries that are likely to generate wastewater containing PFAS.
- Based on the information from the study, Ecology should consider additional
 monitoring requirements for WWTP dischargers. This should include consideration
 of whether EPA has developed approved analytical methods for PFAS suitable for
 WWTP effluent and a regulatory target (a nationally recommended water quality
 criterion for PFAS) for waters of the state.
- Based on this evaluation Ecology should require possible PFAS monitoring for some or all domestic and industrial WWTPs.

Cost

Ecology received \$235,000 to conduct a WWTP sampling study by June 30, 2021. This includes costs for sample analysis, which can range from \$1,000 to \$1,500 per sample as well as project staff salaries.

The cost of establishing additional monitoring requirements based on the sampling study has not been determined. More funding sources may be needed to complete this work.

4.2 Evaluate landfill PFAS emissions

Recommendation

Ecology will develop a sampling program at selected landfills across the state. The sampling will test for PFAS in leachate, groundwater, and air emissions.

Leachate

The Solid Waste Management program (SWM) developed Phase I of the program, involving leachate sampling. With funding and approval, this phase is undergoing final planning. Sampling will begin when COVID-19 restrictions are withdrawn and normal field operations resume.

Ecology developed the study to better characterize landfill leachate. The study will:

- Sample leachate at selected landfills in the state.
- Determine the range of values for 33 PFAS substances in leachate, and compare to landfills throughout the country.
- Arrive at an estimate of the total PFAS materials in the landfill leachate through Total Oxidized Precursor (TOP) analyses.
- Determine if differences in amount of PFAS occurs in landfill cells of different ages.
- Determine if specific types of waste streams lead to higher PFAS values.
- Identify disposed wastes that are likely to generate PFAS releases to leachate.
- Perform a one-time testing of leachate from approximately 23 landfills.
- Consider additional sampling of leachate for landfills not yet sampled after the initial Phase I is completed. This second step of Phase I may include landfills that are undergoing MTCA cleanups, or landfills that contain specific refuse streams that have shown to have high PFAS values from the Phase I sampling.

If warranted, Ecology would manage PFAS in landfill leachate long-term by:

- Considering additional monitoring requirements for landfills to test leachate for PFAS using information from the study above.
- Potentially updating the rules (Chapters <u>173-350</u>¹³ and <u>351</u>¹⁴ Washington Administrative Code [WAC]) to require PFAS testing of leachate during landfill monitoring.

Groundwater and Gaseous Emissions

Phase II of the program will sample groundwater and gas emissions at landfills for PFAS. This phase of the program is in the conceptual stage. Landfills to be sampled will be based on the results of the Phase I leachate study. Groundwater will be sampled from existing monitoring wells.

The Solid Waste Management program (SWM), in conjunction with the Air Quality Program (AQ), will develop the gas emissions sampling portion of the program. Ecology will also consider landfill gas emissions monitoring being conducted by North Carolina State University and Oregon State University.

Landfill waste makeup

In parallel to landfill gas emission sampling above, Ecology will continue to research the makeup of PFAS waste entering and potentially currently stored in landfills.

¹³ https://apps.leg.wa.gov/wac/default.aspx?cite=173-350

¹⁴ https://apps.leg.wa.gov/wac/default.aspx?cite=173-351

Cost

The Phase I testing of leachate from 23 landfills has received \$34,500 of funding. Phase II has also received funding for \$55,500. However, given delays incurred as a result of COVID-19 restrictions in 2020, it is not clear whether Phase II can be conducted within the originally anticipated timeframe, and it is not known if the funding for this work will remain available.

Adding PFAS monitoring requirements to Chapter 173-350 WAC could take two and a half years and cost up to \$1.1 million. Less complex rulemaking could take two years and cost up to \$260,000. These cost estimates include employee time and expenses, but will vary based on the degree of consultation with Ecology's Assistant Attorneys General.

4.3 Evaluate Washington biosolids management

The information gaps regarding biosolids are significant and currently prevent assessment of risk from PFAS in biosolids that are land applied in Washington. Any regulatory changes should be founded on defensible data and science-based risk assessments. If Ecology uses scientific modeling to assess potential PFAS transfer from biosolids to soil or groundwater, realistic model parameters must be used.

Washington biosolids regulation in the near term should ensure sound agronomic land application practices on permitted sites where human exposure is limited. It is premature to add or change regulatory limits given the absence of data from Washington biosolids and problems identified with models and their input parameters.

Recommendation

We recommend the following key steps to address the current data gaps:

- Establish biosolids and soil sample collection and handling methods for PFAS analysis.
- Accredit Washington labs for EPA-validated analysis methods.
- Use EPA-validated analysis methods for biosolids and soils.
- Conduct credentialed third-party review of raw mass spectrometer PFAS data.
- Investigate land application sites where procedures mimic rates and practices under current state rule (Chapter <u>173-308</u> WAC¹⁵).
- Evaluate realistic exposure pathways.
- Evaluate risk modeling using realistic input values.
- Collaborate with stakeholders to get accurate and precise biosolids data. Initial results should remain anonymous.
- Compile analysis data with statistical review.

To conduct this work, Ecology will collaborate with municipalities managing WWTPs.

¹⁵ https://apps.leg.wa.gov/wac/default.aspx?cite=173-308

Cost

As of the date of this Draft CAP, it is not possible to estimate costs for implementing this recommendation. Ecology will recruit a senior employee to lead the biosolids data gathering process. Ecology will also submit program funding requests for both sampling and analysis to help with expenses.

What else are we doing about PFAS?

This section outlines other activities Ecology and Health are conducting in addition to developing this Draft CAP. For the most part, the activities described below implement legislation which has been adopted. Some of these activities also respond to earlier CAP recommendations, for example reducing AFFF releases to the environment, and further assessing certain products which contain PFAS (e.g. carpeting) as priority PFAS sources.

Rulemaking

State drinking water rulemaking

The SBOH initiated two <u>rulemaking activities</u>¹⁶ to address PFAS in drinking water. The SBOH is considering establishing a state action level for PFAS in drinking water. The proposed revisions to Chapter <u>246-290</u> WAC¹⁷ intend to improve public health protection by setting a PFAS regulatory standard for Group A public water systems in Washington. A state action level would be set for PFOS, PFOA, perfluorononoic acid (PFNA), PFHxS and perfluorobutane sulfonic (PFBS). The revisions also propose monitoring, recordkeeping and reporting, follow-up actions for PFAS. The SBOH is also considering amendments to the drinking water laboratory certification and data reporting rules (Chapter <u>246-390</u> WAC¹⁸) to align laboratory data reporting requirements with the anticipated changes to Chapter 246-290 WAC outlined above. Health's <u>overall timeline</u>¹⁹ and <u>lab rule timeline</u>²⁰ anticipates draft rules will be issued for comment in fall 2020, and the rulemaking completed in 2021.

Law implementation

Firefighting Agents and Equipment

Chapter 70.75A RCW²¹ establishes restrictions on Class B firefighting foam, which contain intentionally added PFAS chemicals:

• As of July 1, 2018, prohibits use of Class B firefighting foam for training.

¹⁶ https://www.doh.wa.gov/CommunityandEnvironment/DrinkingWater/RegulationandCompliance/RuleMaking

¹⁷ https://apps.leg.wa.gov/wac/default.aspx?cite=246-290

¹⁸ https://apps.leg.wa.gov/wac/default.aspx?cite=246-390

¹⁹ https://www.doh.wa.gov/Portals/1/Documents/4200/PFAS-Timeline.pdf

²⁰ https://www.doh.wa.gov/Portals/1/Documents/4200/LabRuleTimeline.pdf

²¹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

- As of July 1, 2020, prohibits the manufacture, sale, and distribution of Class B firefighting foam.
 - Interim exemptions include federally required users, petroleum storage and distribution facilities, or certain chemical plants.
- Requires manufacturers to notify Washington purchasers about the presence and purpose of PFAS in firefighting personal protective equipment.
- Two years after amendment of federal regulations (prohibiting the use of PFAS-containing foam), requires federal facilities to use non-PFAS foams that comply with the new federal regulation.
 - Airports must inform Ecology about their ability to switch to non-PFAS foams within 18 months of the change in federal regulations.
- Beginning 2024, restricts the purchase of PFAS-containing foams by oil terminals, oil refineries, and chemical plants.

Update: Ecology has conducted the following activities to implement the law.

- Developed an agency website to provide more information and outreach materials regarding the requirements of the law.²²
- Conducted outreach to manufacturers to explain the requirements and ensure compliance with the restrictions.
- Collaborated with firefighting foam users on the restriction of PFAS-containing firefighting foam use in training, and on the purchase restriction taking effect in 2020.
- Provided technical assistance to state and local governments and other jurisdictions to help them purchase PFAS-free Class B firefighting foam.
- Consulted with the Department of Enterprise Services to develop procurement preferences for state and local governments and other jurisdictions to purchase PFAS-free Class B firefighting foam alternatives.
- Informed firefighting personal protective equipment manufacturers of the requirement to notify purchasers about the presence of PFAS—and requested copies of the notification.
- Surveyed state and local governments and other jurisdictions about stocks of Class B firefighting foam through Ecology's <u>Product Replacement Program</u>.²³

Packages Containing Metals and Toxic Chemicals

The Packages Containing Metals and Toxic Chemicals law (Chapter 70.95G RCW ²⁴) includes the following restrictions:

• Effective January 2022, prohibits PFAS in plant fiber-based food packaging.

²² https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Addressing-priority-toxic-chemicals/PFAS/Toxics-in-firefighting

²³ https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Product-Replacement-Program

²⁴ https://app.leg.wa.gov/RCW/default.aspx?cite=70.95G&full=true

- Requires Ecology to conduct an AA to identify safer alternative products. This
 assessment must consider chemical hazard, performance, cost and availability, and
 exposure.
 - Ecology must submit the findings for external peer review and publish the results in the Washington State Register.
- Requires Ecology to report results to the Legislature before a ban on PFAS in food packaging can take effect.

Update: Ecology has completed—or is conducting—the following work to implement the law.

- Following a competitive bid process, Ecology selected SRC, Inc. to conduct the AA.
- A statement of work, deliverables, and timeline are available on the <u>PFAS in food</u> packaging AA website.²⁵
- SRC, Inc., and Ecology are working with a broad range of interested parties to gather the information needed for the assessment. Ecology and SRC, Inc. regularly conduct webinars to update interested stakeholders and the public on the project.
- Our analysis and research focuses on food wrappers, liners, and other packaging categories such as plates, bowls, trays, and take-out containers. Ecology and SRC, Inc. are reaching out to end-users of these products for input on the use of alternatives in the market.
- Ecology requested an external peer review by the Washington State Academy of Sciences and will publish the results in the Washington State Register.

The Pollution Prevention for Healthy People and Puget Sound Act

The Pollution Prevention for Healthy People and Puget Sound Act (Chapter 70.365 RCW²⁶) creates a process for Ecology, in consultation with Health, to regulate classes of chemicals in consumer products. Ecology is implementing the law through the Safer Products for Washington²⁷ program. It identifies PFAS as priority chemicals and requires Ecology to:

- Designate priority chemicals and identify products that contain these chemicals.
- Consider feasible and available chemical alternatives.
- Determine regulatory actions and adopt rules to implement regulatory actions.
- Conduct stakeholder consultation, legislative reporting, and rulemaking.

Update: Under the implementation program, Ecology completed the following:

• Published a <u>draft report</u>²⁸ in January 2020 recommending pretreated carpets and rugs and aftermarket treatments for carpets and rugs as priority consumer products for further research in January 2021.

²⁵ https://www.ezview.wa.gov/site/alias 1962/37610/pfas in food packaging alternatives assessment.aspx

²⁶ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365&full=true

²⁷ https://ecology.wa.gov/safer-products-wa

²⁸ https://fortress.wa.gov/ecy/publications/summarypages/2004004.html

- Reviewed comments received on the draft.
- Expanded the categories considered to also include water and stain resistance treatments as well as leather and textile furnishings as priority consumer products.
- Submitted the <u>final version of the report</u>²⁹ to the Legislature on July 6, 2020.

Children's Safe Products Act

The Children's Safe Products Act (CSPA), Chapter <u>70.240</u> RCW,³⁰ requires manufacturers to annually report the presence of certain chemicals (including PFOS and PFOA) in children's products sold in Washington state.

Update: Ecology implements the law as follows:

- Ecology receives manufacturer reports and conducts compliance activities.
- Manufacturer reports are <u>published online</u>.³¹

Other activities

Landfill leachate sampling

One of the 2019 Preliminary Recommendations addressed gathering more information about PFAS in landfill leachate. Ecology has begun this work. Ecology's Solid Waste Management Program (SWM) developed Phase I of a landfill leachate sampling program. This phase is funded and approved, and is undergoing final planning. (Sampling will begin when COVID-19 restrictions are withdrawn and normal agency field operations resume.) The study will sample leachate at selected landfills in the state to determine the range of values for 33 PFAS substances. Values will be compared to landfills across the country, and the data will be used to:

- Evaluate potential differences in amount of PFAS across landfill cells of different ages.
- Determine whether specific waste streams lead to higher PFAS values. This will identify disposed waste that is likely to release PFAS to leachate.

WWTP sampling

One of the 2019 Preliminary Recommendations addressed gathering more information about PFAS in WWTP influent and effluent. Ecology received funding to develop and conduct sampling of PFAS in influent, effluent, and biosolids at three municipal WWTPs receiving industrial discharges. This data would help inform which treatment processes are more effective at transforming and removing PFAS. Ecology is working on staffing this project in the context of COVID-19 hiring restrictions so that the work can be completed by June 30, 2021 when the funding expires.

²⁹ https://fortress.wa.gov/ecy/publications/summarypages/2004019.html

³⁰ https://app.leg.wa.gov/RCW/default.aspx?cite=70.240

³¹ https://hpcds.theic2.org/Search

Per- and Polyfluoroalkyl Substances Draft Chemical Action Plan

Fish Consumption Advisory

Health is developing fish consumption advisories for PFOS in freshwater fish based on Ecology fish sampling data. Health received additional data from Ecology in 2019 to provide an adequate basis for a fish consumption advisory. Health is reviewing these data and reevaluating screening levels in consideration of recent changes in recommended oral intake.

Introduction

A Chemical Action Plan (CAP) "identifies, characterizes, and evaluates uses and releases" for individual, or groups of, PBTs, and "recommends actions to protect human health or the environment" WAC <u>173-333-400(1)</u>. Chapter 173-333 WAC identifies perfluorooctane sulfonates (PFOS), a type of per- and polyfluoroalkyl substances (PFAS), as a chemical group that requires further action because they persist in the environment for long periods of time where they can bioaccumulate to levels that pose threats to human health and environment in Washington.

The Washington State Department of Ecology (Ecology) and the Washington State Department of Health (Health), jointly "we," prepared this Draft PFAS Chemical Action Plan (Draft CAP) to identify, characterize, and evaluate PFAS uses, releases, and current PFAS management approaches in Washington state. Based on these considerations, we recommend actions to reduce PFAS exposure, use, and release in Washington. The recommendations address urgent public health and environmental concerns while considering feasibility, social impacts, and economic costs. As described in detail in the PFAS CAP Requirements section below, the Draft CAP considers the family of PFAS as a whole.

The Draft CAP includes the following sections:

- A <u>PFAS Assessment Summary</u> section reviewing the findings of our analysis.
- The <u>Draft CAP Recommendations</u> stemming from these assessments and a discussion of PFAS-related activities Ecology and Health are conducting in addition to preparing this CAP.
- A description of the <u>PFAS CAP Requirements</u> guiding the preparation of the Draft CAP and next steps in the CAP process.

A series of ten appendices then assess current scientific knowledge and impacts of PFAS in Washington, each identifying the recommendations it informed. The appendices are organized as follows:

- Appendix 1: Chemistry—reviews the chemical characteristics of PFAS.
- Appendix 2: Analytical Methods—outlines analytical standards available to identify PFAS in environmental media.
- Appendix 3: Sources and Uses—summarizes commercial use of PFAS and how this
 use results in environmental releases.
- Appendix 4: Fate and Transport—describes how PFAS enter and behave in the environment.
- Appendix 5: Environmental Occurrence—describes how PFAS are distributed throughout Washington state's environment.
- Appendix 6: Ecological Toxicology—reviews toxicological impacts of PFAS to environmental media and their inhabitants.

³² https://app.leg.wa.gov/wac/default.aspx?cite=173-333-400

- Appendix 7: Health—reviews the potential for human exposure and health impacts for several PFAS and summarizes PFAS occurrence in state drinking water.
- Appendix 8: Biosolids—reviews the impacts of PFAS in biosolids generated from wastewater treatment.
- Appendix 9: Regulations—reviews state and federal regulations that apply to PFAS in Washington state.
- Appendix 10: Economic Analysis—presents qualitative and quantitative estimates of costs to implement recommendations, as well as costs and benefits from reducing PFAS in Washington's environment.

PFAS Assessment Summary

This section summarizes the major findings from our assessment of PFAS, their presence in our environment, and their potential impacts. In order to make this summary accessible, we did not include the citations that support each statement. Each appendix includes detailed assessment findings and associated references to scientific and other sources. Readers should consider the information below in the context of and in combination with the full analysis presented in each appendix.

Chemistry (Appendix 1)

More than 4,730 PFAS are registered in the Chemical Abstract Service. As of November 2019, U.S. Environmental Protection Agency's (EPA) master list of PFAS includes 7,866 chemical compounds. EPA identified 602 PFAS which are actively used in U.S. commerce. The large chemical family of PFAS is subdivided into non-polymer and polymer classes.

In their manufactured form, PFAS can be gases, liquids, and high-molecular weight polymer solids. Individual PFAS can be raw materials, compounds used in products, or environmental transformation products. One of the important chemical characteristics of PFAS is their resistance to extreme environments. This characteristic makes PFAS completely resistant to natural degradation.

Due to their well-established properties, perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) dominate the literature on PFAS. Much of the regulatory interest on PFAS in the environment focuses on PFOS and PFOA. Both of these chemicals are long-chain fluorine and carbon atoms. The number of carbon atoms distinguishes long-chain PFAS from short-chain PFAS. Since 2002, voluntary stewardship efforts eliminated PFOS and PFOA production in the U.S., Japan, and Europe.

Below are examples of how PFAS chemical characteristics are used in products:

- Carpet and textile treatments to impart stain and water resistance.
- Paper and packaging treatment to provide water, oil, and grease resistance as well as non-stick performance.
- Surfactants to impart water, oil, and dirt resistance to painted walls, sealed grout, or polished floors.
- Hydraulic fluids resistant to high temperature or reactive environments.
- PFAS-based aqueous film forming foams (AFFF) to extinguish Class B (flammable liquid fuel) fires.
- Durable and heat-stable fluoropolymer surfaces that creates non-stick cookware.
- Durable water-resistant layer for outdoor clothing that creates breathable but waterproof fabric.

Analytical Methods (Appendix 2)

A variety of analytical methods are available to analyze PFAS in consumer products and the environment, and the methods are still evolving. Currently, few methods are formally validated and published. <u>Appendix 2: Analytical Methods</u> reviews the standard and non-standard analytical methods for analyzing PFAS.

In November 2018, EPA published a multi-laboratory validated method, EPA Method 537.1 version 1.0, for analyzing 18 PFAS analytes in drinking water. EPA later updated this method with Revision 2.0. In December 2019, EPA announced a new validated method for testing additional PFAS in drinking water, EPA Method 533. This method focuses on PFAS with carbon chain lengths of four to twelve, and complements EPA Method 537.1 version 1.0.

Methods 537.1 Revision 2.0 and 533 are intended to analyze PFAS in drinking water. As a result, they are not effective for additional PFAS compounds or other sample types without modifying the method.

EPA is tentatively scheduled to issue Draft Method 8328 in 2020. The draft Method 8328 will make use of solid-phase extraction to sample water not used for drinking. Additional methods EPA is developing and validating to detect and quantify PFAS in air, water, and soil include:

- Clean Water Act Method 1600.
- OTM Method 45.
- Standard Operating Procedures for Total Organic Fluorine.
- Analytical Model to Identify Novel PFAS Using Non-Targeted Analysis Data.

Most of the available standard methods for PFAS analysis do not account for all known PFAS. Human exposures to PFAS are generally not from individual PFAS, but from complex mixtures. Analytical techniques are limited in determining which PFAS are in those mixtures. Non-specific methods for PFAS analysis assessed in Appendix 2 include:

- Combustion ion chromatography.
- Particle-induced gamma ray emission.
- Total oxidizable precursors assay.

Analysis of PFAS is progressing, but challenges remain because the complete list of PFAS relevant to environmental and human exposure is unknown. As more studies identify novel PFAS, an effective, comprehensive technique that is capable of quantitative, non-target analysis remains elusive.

Ecology's analysis of analytical methods does not include specific recommendations. Ecology supports the use of approved validated methods as recommended by EPA for specific targeted PFAS analysis. Implementation of several CAP recommendations requires PFAS sampling in environmental media, which would benefit from improved analytical approaches that can characterize unknown PFAS.

Sources and Uses (Appendix 3)

PFAS contamination is widespread. As of April 2020, in the U.S., 300 sites and 390 water systems in more than 40 states have known PFAS contamination. AFFF use during emergency response, equipment testing, and training exercises contributes to PFAS groundwater contamination. AFFF has been stored and used throughout Washington. Ecology estimates that 1.4 million liters may have been stored statewide in 2004, and 606,000 liters in 2011.

PFAS releases from manufacturing are linked to approximately 60 contaminated sites across the U.S. We do not know of any PFAS manufacturing which has occurred, or continues to occur, in Washington state. We estimate that the range of industry sectors (mining, paper products, resins, surfactants, etc.) that potentially use PFAS—as raw material or as a product component—represent approximately 1,200 manufacturing businesses. However, we do not yet know whether any of these operations have used, or currently use PFAS.

People are exposed to PFAS in their homes when they use contaminated products and through contact with house dust that contains PFAS. For example, textile-related products that use fluorinated chemicals include carpeting and upholstery, outer garments, tents, car seat covers, leather articles, etc. Studies have also identified a variety of PFAS in a range of cosmetics. Between 2014 and 2019, PFOS was reported in 112 children's products sold in Washington. High levels of PFAS have been reported in occupational settings (such as carpet shops and industries that use products containing PFAS), where we estimate that 255,515 Washington workers could be exposed.

PFAS may be entering the state's ambient environment as a result of waste disposal, landfill leachate (liquid that drains from a landfill), land application of industrial sludge, and discharges of municipal and industrial wastewater. Numerous products that contribute to waste streams contain PFAS. Some wastewater treatment plant (WWTP) effluent sampling in Washington found PFAS levels similar to WWTPs in other areas of the U.S.

Landfill leachate can be a potential source of PFAS to the environment. Privately and publicly operated landfills, which receive and store wastes, are likely to receive products containing PFAS. Uncontrolled leachate can migrate into groundwater, resulting in contamination. Controlled leachate, which may contain PFAS, is typically sent to WWTPs, potentially increasing PFAS in WWTP influent.

Data suggest that 51.66 to 17,043 metric tons of PFAS are landfilled in consumer products in Washington each year. From 1960 – 2002, we estimate Washington state's average annual contribution of six PFAS emissions (the sum of perfluoro-carboxylic acid [PFCA], fluorotelemer alcohol [FTOH], perfluorooctane sulfonyl fluoride [POSF], PFOS, perfluorohexane sulfonic acid [PFHxS], and perfluorodecane sulfonate [PDSF]), resulting from product use and waste streams, is approximately 29.5 metric tons per year.

Fate and Transport (Appendix 4)

Manufacturing processes can use and emit PFAS directly into the environment. Another source of environmental PFAS concentrations is the degradation of long-chain substances, usually called "precursors." Many PFAS degrade to perfluoroalkyl acids (PFAAs), which are very stable in the environment and are referred to as terminal substances. The timeframe for the transformation from precursor to terminal substance depends on the compounds present and the surrounding environmental conditions. Transformation processes include:

- Abiotic (without living organisms).
- Biotic aerobic (by organisms with access to oxygen).
- Biotic anaerobic (by organisms without oxygen).

Even though U.S. production of PFOS and PFOA was phased out by 2002 and 2015 respectively, levels of certain PFAAs have continued to increase in wildlife because of these transformation processes. Manufacturers continue to make other precursor compounds, which transform into PFAAs in the environment. Surface waters and wildlife have measurable levels of both precursors and PFAAs. This shows that exposure to precursors can be significant.

During direct or secondary manufacturing, PFAS can be released to the air through stack emissions. Once in the air, certain PFAS can travel large distances before deposition, as shown by their occurrence across the globe, far from all manufacturing sites.

Environmental release to bodies of water results from secondary manufacturing activities. Neither the state nor federal Clean Water Act (CWA) establish numeric standards for discharge of PFAS in industrial wastewater discharges. PFAS-containing product use and disposal in domestic wastewater can result in PFAS presence in sewage. Similarly, PFAS can also be present in domestic wastewater effluents, which are released to on-site wastewater systems, and typically discharge to groundwater.

Due to their high solubility, many PFAS compounds may be susceptible to leaching from landfills or contaminated biosolids, compost, and soils when exposed to water. PFAS will often localize at phase interfaces, such as soil and water or water and air boundaries.

Individual PFAS will adsorb to organic carbon in soil to varying degrees. How long PFAS remain in soil depends on site-specific factors. However, evidence shows that desorption is often incomplete. As such, soil contaminated with PFAS may remain as a low-volume source of contamination for ground and surface water for a long time.

Environmental Occurrence (Appendix 5)

In Washington, PFAAs have been detected in fresh and marine surface waters, stormwater in urban industrial catchments, WWTP effluent (treated water leaving the treatment plant), freshwater and marine sediments, catch basin sediments, freshwater and marine fish, mussels,

and osprey eggs. Environmental concentrations of PFAAs in Washington state surface waters, WWTP effluent, and freshwater fish tissue sampled in 2016 were consistent with PFAS levels in other parts of the U.S. not impacted by PFAS manufacturing facilities.

Beyond Washington, PFAS have been detected in other wildlife, with PFOS generally detected at the highest frequency and in the greatest amount.

Monitoring suggests that stormwater, WWTP effluent, and uncontrolled releases of AFFF are primary ways that PFAAs are delivered to water bodies. PFOS (and to a lesser extent, perfluorodecanoic acid [PFDA], perfluorododecanoic acid [PFDoA], perfluoroundecanoate [PFUnA], and perfluorooctane sulfonamide PFOSA) are widespread in freshwater fish tissue found in Washington state's water bodies. Samples of urban lake fish tissue had PFOS levels that are above Department of Health's initial screening levels and may trigger consumption advisories to protect human health. Sampling in 2018 confirmed that PFAS concentrations in freshwater fish collected from Washington urban lakes are consistent with other urban water bodies in North America.

Environmental monitoring in 2016 suggested that PFAA levels in surface waters and WWTP effluent had decreased since the last round of sampling in 2008. A shift was evident in WWTP effluent samples: short-chain PFAAs were replacing PFOA as the most dominant compounds.

At sites affected by urban sources and WWTP inputs, PFOS and other long-chain PFAAs are detected in osprey eggs at concentrations that are high enough to reduce hatchability. PFAS concentrations (primarily PFOS) in osprey eggs remained unchanged between 2008 and 2016.

Ecological Toxicology (Appendix 6)

PFOA and PFOS are the major PFAS contaminants found in oceanic waters. A variety of wildlife across the globe have measurable PFCA (perfluorooctanoate [PFO], perfluorononanoate [PFN], or perfluorodecanoate [PFD]) concentrations. Stability and water solubility allow PFAS to transport through marine environments, concentrate in marine organisms, and easily accumulate throughout all trophic levels.

Both short- and long-chain PFAS are environmentally persistent. Long-chain PFAS tend to be more bioaccumulative and produce adverse toxicological effects, even at relatively low contaminant levels. While resistant to degradation, short-chain PFAS appear to be less bioaccumulative and to have fewer significant toxicological effects. Though short-chain PFAS are less bioaccumulative, high mobility and bioavailability lead to relatively high levels in fish tissues.

Bioaccumulation or biomagnification has been confirmed in marine and terrestrial species, zooplankton and other invertebrates, and fish. PFAS have been shown to be taken up by plants from soil, with different PFAS presenting in different portions of the plant.

Biomagnification results in greater levels of PFAS in animals higher on the food chain (e.g., seals, polar bears), relative to animals at lower trophic levels. PFAS are persistent and able to transport long distances, and bioaccumulation is not required for sustained internal exposure. Therefore, animals do not need to be near sources of PFAS to show bioaccumulation, and exposure will continue regardless of accumulation.

Scientific literature supports the association between PFOA exposure and reduced antibody response in animals. Animal studies with both PFOS and PFOA show that they are well-absorbed orally, but poorly eliminated. PFAAs bind to proteins (rather than fats, like other bioaccumulating compounds), and are found mostly in the liver and blood. Documented toxicological effects of PFAS include:

- Inhibited growth and detrimental effects on photosynthesis on green algae and floating macrophyte, L. gibba (P. subcapitata, S. capricornutum, and C. vulgaris).
- Slight to moderate toxicity to aquatic invertebrates.
- Impacted fertility in adult fish.
- Risks for impacted development in Arctic marine food webs.
- Reduced plant root elongation.
- Induction of liver tumors in Wistar rats.
- Significantly stunted mammary epithelial growth and ultrastructural liver changes in mice
- Reduced hatchability and pathological liver changes in chickens.

Health (Appendix 7)

We are still learning about potential human health risks of PFAS. Much of what we know is from toxicity testing in laboratory animals. The evidence base is strongest for PFOA and PFOS, but is expanding for other PFAAs.

Animal studies provide strong evidence that some PFAAs produce liver and kidney toxicity, immune toxicity, reproductive and developmental toxicity, endocrine disruption (altered hormones), and certain tumors. Epidemiological studies link higher exposures to PFAAs with reduced antibody response to childhood vaccines, increased cholesterol and liver enzymes, and slightly reduced birth weights, among others.

It takes years for human bodies to excrete PFOS, PFOA, PFNA, PFHxS, and other long-chain PFAS—some are strongly bioaccumulative in people. Other PFAAs such as perfluorobutanoic acid (PFBA), PFBS, and perfluorohexanoic acid (PFHxA) are more rapidly cleared. For most PFAS, absorption, distribution, and clearance in humans have not been studied.

Since 1999, Centers for Disease Control and Prevention (CDC) surveys of the U.S. population detected PFOS, PFOA, PFHxS, and PFNA in the blood of nearly every participant. Levels have declined since phase-outs of these PFAS from domestic production and use, but current analysis methods cannot identify all PFAS in human blood, underestimating occurrence.

People can be exposed to PFAS from contaminated drinking water and other dietary sources, from indoor dust and air containing PFAS released from consumer products, and from use of PFAS-containing consumer products. Although difficult to assess, studies identify food and drinking water as the likely main routes of non-occupational exposure for people.

Several Washington drinking water sources have been contaminated near sites of AFFF release:

- City of Issaquah.
- Naval Air Station, Whidbey Island—with off-base impacts to the Town of Coupeville, plus adjacent public and private drinking water supplies.
- Naval Base Kitsap Bangor—with off-base impacts to private wells.
- Joint Base Lewis McChord, and the Fort Lewis and McChord field water systems with off-base impacts to city drinking water systems in Dupont, Lakewood, Tacoma, and Parkland.
- Fairchild Air Force Base—with off-base impacts to the City of Airway Heights and private well owners.

In each area, the sum of PFOA and PFOS in at least one drinking water well exceeded EPA's lifetime health advisory level (70 parts per trillion [ppt]). AFFF is the suspected contamination source in all of these areas. Ongoing site investigations may identify other sources.

Biosolids (Appendix 8)

Chapter $\underline{173-308}^{33}$ WAC, Biosolids Management, divides wastewater solids into two classes. Those that meet the regulatory standards to allow land application are classified as biosolids, whereas those that do not meet the regulatory standards are classified as sewage sludge. Washington law requires that biosolids are land applied (i.e., applied to agricultural fields as fertilizer) to the greatest extent possible, but sewage sludge is disposed in landfills. Currently, about 85-90% of biosolids generated in Washington are land-applied.

Some U.S. labs are analyzing biosolids using modified procedures based on EPA's Method 537. However, guidelines are inconsistent and results are not validated. For PFAS analysis using modified 537 methods, Ecology's lab accreditation unit at Manchester Environmental Laboratory recognizes the National Environmental Laboratory Accreditation Program (NELAP) for a few other Washington labs. EPA is in the process of validating a different procedure for analyzing PFAS in biosolids and soil—SW-846.

Most studies assessing contamination impacts from biosolids application sample WWTPs receiving influent directly from industries using fluorinated compounds. Although some industrial discharge in Washington is possible, we anticipate that the majority of perfluorinated compounds in Washington municipal wastewater originate from domestic sources.

³³ https://apps.leg.wa.gov/wac/default.aspx?cite=173-308

Worldwide monitoring data show that PFOA and PFOS concentrations in biosolids are trending downward, likely due to less production of the compounds. Studies with high loading rates to agricultural soils showed bioaccumulative effects in some vegetables or diminished growth of spring wheat. However, the PFOS and PFOA application rates in these studies were likely far higher than the PFOS and PFOA present in Washington biosolids. The lack of potential industrial contamination in Washington means these negative impacts on crop growth are not likely to be representative of biosolids applications in Washington state.

Adoption of extremely low regulatory limits for soil PFAS could have adverse consequences for organics and residual recycling, and may not provide demonstrated risk-reduction for human health and the environment.

Regulations (Appendix 9)

Regulatory action to restrict the production and use of PFAS has been enacted at both state and federal levels. The best understood long-chain PFAS (such as PFOS and PFOA) were voluntarily withdrawn from commercial use in the U.S. However, specialized uses are still permitted.

Washington state is considering developing drinking water standards for several PFAS. Regulatory activity in Washington includes, for example:

- Manufacturer reporting requirements under for children's products (Chapter 70.240³⁴ RCW).
- Restrictions for use, and eventual bans of AFFF (Chapter 70.75A³⁵ RCW).
- Assessments of safer alternatives for PFAS used in carpets and rugs, textile and leather furnishings, aftermarket textile treatment products (Chapter 70.365³⁶ RCW), and food contact packaging (Chapter 70.95G³⁷ RCW).

At the federal level, the Food and Drug Administration oversees PFAS use in food packaging. The ATSDR advises local, state, federal, and tribal agencies regarding human health effects. DOD enacted requirements to decrease PFAS use—such as in AFFF in food packing for military rations, for example. DOD will also continue with initiatives to address PFAS contamination resulting from its activities.

Under the Toxic Substances Control Act (TSCA), EPA has minimized and regulated the manufacture and use of certain long-chain PFAS. Data on PFAS use nationally will soon be collected via EPA's Toxics Release Inventory. EPA is coordinating these and other activities under its 2019 PFAS Action Plan.

³⁴ https://app.leg.wa.gov/RCW/default.aspx?cite=70.240

³⁵ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

³⁶ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365

³⁷ https://app.leg.wa.gov/RCW/default.aspx?cite=70.95G&full=true

Economic Analysis (Appendix 10)

Statewide costs for PFAS mitigation in drinking water will not be fully understood until further testing to characterize the occurrence in drinking water is complete. Currently, a few examples are available to consider the costs of drinking water mitigation for PFAS. Some examples do not separate investigation costs.

- The City of Issaquah spent more than \$600,000 (plus ongoing maintenance costs) to install a filter on one PFAS-contaminated city well.
- The Naval Air Station Whidbey Island spent \$1.37 million, to date, for investigation and mitigation efforts. Estimated cost to add an activated carbon treatment to the Town of Coupeville's water system is \$1.5 million.
- In 2017, Airway Heights public water system shut down PFAA-contaminated wells.
 Using the City of Spokane water system, Airway Heights used an emergency intertie
 (to flush their system with clean water) and added another connection (to supply
 water while they pursue treatment options). The water purchase (439 million
 gallons) could cost \$687,000 in the first year. The Air Force has agreed to pay the
 city.
- Activated carbon filtration treatment of drinking water in Lakewood is estimated to cost \$5.2 million for initial capital investments, and \$96,000 per year for ongoing maintenance.
- At Joint Base Lewis McChord, McChord Field System, activated carbon filtration treatment of water from three wells is estimated to cost \$10.3 million for initial capital investments, and \$830,000 per year for ongoing maintenance.

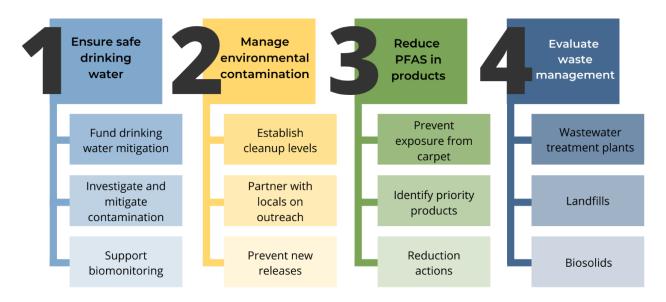
These costs are in line with similar drinking water remediation activities in other states. The total cost of remediation for PFAS-contaminated groundwater remains unknown, because there are no completed cleanups of PFAS contamination in the U.S.

Based on Washington state Model Toxics Control Act (MTCA) remediation ratios, we have estimated that overall remediation costs could range between \$5.3 million and \$62.8 million for a site where AFFF release results in groundwater contamination. Interim solutions such as filtering or alternative sources of drinking water could result in ten-year costs of \$6.5 million to \$10 million.

Draft CAP Recommendations

Our CAP recommendations address a broad range of concerns. Figure 1 provides an overview of the four main categories of recommendations proposed.

Figure 1. Recommendations found in this Draft Chemical Action Plan and associated sub-efforts to support implementation.



We based these recommendations on our assessment of scientific information—presented in the appendices—and Advisory Committee input (see section PFAS CAP Requirements below). These broad categories were first identified in the 2018 Interim Chemical Action Plan for Perand Polyfluorinated Alkyl Substances³⁸ (Interim CAP). Over time, as we improved our knowledge of PFAS, and as several pieces of legislation were passed and implemented, some of our earlier recommendations were acted upon and further refined in our May 2019 Preliminary CAP Recommendations³⁹ (Preliminary Recommendations). Our activity to address PFAS has continued since May 2019, resulting in additional evolution of our recommendations, described below.

For each of the four main recommendations, this section provides a summary of how the recommendations have evolved since they were first considered in 2018, and identifies implementation activities that have already begun. Most of the activities described below implement legislation that has been adopted. Some of these activities also respond to Interim CAP or Preliminary Recommendations, for example reducing AFFF releases to the environment, and further assessing certain PFAS-containing products.

³⁸ https://fortress.wa.gov/ecy/publications/SummaryPages/1804005.html

³⁹ https://www.ezview.wa.gov/Portals/_1962/Documents/PFAS/PrelimRecommendations-2019-PFAS-CAP.pdf

Implementing the Draft CAP recommended actions would require additional resources and funding. We include agency cost estimates for some actions. <u>Appendix 10: Economic Analysis</u> addresses economic impacts to other entities for some actions.

1.0 Ensure drinking water is safe

Protecting public health by ensuring safe drinking water is a fundamental responsibility of the Health Office of Drinking Water (ODW).

There are three types of drinking water systems in Washington:

- Group A water systems serve <u>85% of state residents</u>.⁴⁰ They service more than 15 connections or more than 25 people. ODW primarily regulates these public water systems.
- Group B water systems are smaller and serve 1.5% of state residents. The local health department usually oversees these systems. Group B systems have few testing requirements for continued operation.
- Private wells serve 13.5% of state residents. Private wells are only regulated in their design and installation. Chemical testing is not usually required.

Less than 1% of all Group A systems have been tested for PFAS. However, those that have been tested serve most water customers in the state. The percentage of Group B and private wells tested for PFAS is even lower. A water test is required to determine whether PFAS are in drinking water, because PFAS are tasteless and odorless at levels of public health concern.

Because testing and treating for PFAS in drinking water is expensive, exposures to PFAS-contaminated water may be disproportionately borne by populations who do not have the financial means to test for and remove these contaminants.

Previous CAP Recommendations

Interim CAP

The Interim CAP⁴¹ proposed three areas of action pertaining to drinking water protection.

First, it proposed supporting rulemaking for state drinking water standards. This recommendation is being implemented. The SBOH initiated two <u>rulemaking activities</u>⁴² to address PFAS in drinking water. The SBOH is considering establishing a state action level for PFAS in drinking water. The proposed revisions intend to improve public health protection by setting a PFAS regulatory standard for Group A public water systems in Washington. Proposed revisions to Group A Public Water Supplies (Chapter 246-290 WAC⁴³) include:

⁴⁰ https://www.doh.wa.gov/DataandStatisticalReports/EnvironmentalHealth/DrinkingWaterSystemData/DataDownload

⁴¹ https://fortress.wa.gov/ecy/publications/SummaryPages/1804005.html

⁴² https://www.doh.wa.gov/CommunityandEnvironment/DrinkingWater/RegulationandCompliance/RuleMaking

⁴³ https://apps.leg.wa.gov/wac/default.aspx?cite=246-290

- Establishing levels in drinking water expected to have no known adverse impact to health over a lifetime of exposure for PFOS (15 ppt), PFOA (10 ppt), PFNA (14 ppt), PFHxS (70 ppt), and PFBS (860 ppt).
- Monitoring, recordkeeping and reporting, follow-up actions, and other requirements for PFAS and other unregulated contaminants with established state advisory levels.
- Technical and editorial changes as needed.

The SBOH is also considering amendments to the drinking water laboratory certification and data reporting rules, Chapter 246-390 WAC.⁴⁴ These amendments would align laboratory data reporting requirements with the anticipated changes to Chapter 246-290 WAC outlined above.

Health's <u>overall timeline</u>⁴⁵ and <u>lab rule timeline</u>⁴⁶ anticipate draft rules will be issued for comment in fall 2020, and the rulemaking completed in 2021. Health is absorbing costs to conduct PFAS rulemaking with existing resources. As part of the rulemaking process, Health will assess costs for water systems to comply with testing requirements and to act when drinking water exceeds state action levels.

Second, the Interim CAP proposed to expand voluntary testing for PFAS to include drinking water sources and PFAS chemicals that have not yet been evaluated. It sought to prioritize water systems at high risk for contamination for early testing, such as those near airports or firefighter training centers. Health estimated a range of \$235,000 to \$8 million in costs to implement this recommendation based on the scope of water systems (Group A, Group B, or both) included. Health was unable to secure commercial laboratory services or sufficient funding for this initiative in 2018 – 2019. The drinking water rulemaking activity described above proposes a statewide requirement for public water systems to test for PFAS at least once. This would essentially implement the testing recommendation.

Third, the Interim CAP proposed to implement methods to reduce PFAS in drinking water. This recommendation sought to encourage water systems to implement options to meet EPA's health advisory level for PFOA and PFOS of 70 ng/L until state rulemaking is complete. It also proposed providing technical assistance to public water systems for talking to the public about contamination, mitigation options, and monitoring. As described in Appendix 7, Health is working closely with public drinking water systems known to be affected by PFAS releases.

The remainder of this recommendation was further developed in the Preliminary Recommendations, described below.

Preliminary Recommendations

The Preliminary Recommendations identified three activities aimed at providing support to public water systems adversely affected by PFAS releases into the environment:

⁴⁴ https://apps.leg.wa.gov/wac/default.aspx?cite=246-390

⁴⁵ https://www.doh.wa.gov/Portals/1/Documents/4200/PFAS-Timeline.pdf

⁴⁶ https://www.doh.wa.gov/Portals/1/Documents/4200/LabRuleTimeline.pdf

- Identify funding for PFAS drinking water mitigation.
- Provide technical support for site characterization, source investigation, and mitigation at contaminated sites.
- Seek funding for biomonitoring to support impacted residents and help answer important health questions.

These three recommendations are included in this Draft CAP, as described below. For the third recommendation, we refined our emphasis on finding opportunities for Washington residents to participate in exposure and health studies.

1.1 Identify funding for PFAS drinking water mitigation

Recommendation

State agencies, the Washington State Legislature, and local water systems should work together to fund PFAS drinking water mitigation. These costs should be reimbursed by responsible parties under applicable laws. Once PFAS water contaminants are officially classified as hazardous substances by the federal government or by the state of Washington, they can be addressed under the state Model Toxics Control Act (MTCA) framework.

Existing resources

Drinking Water State Revolving Fund is an EPA-funded loan program administered by Health. The loans are used to:

- Improve drinking water infrastructure.
- Finance the cost of installing treatment or other infrastructure improvements over a number of years.

Drinking Water State Revolving Fund can provide emergency loans in the event a water system is issued a "Do Not Use" order as a result of PFAS contamination. The program recently funded a reservoir project for City of Spokane to allow Spokane to provide reliable water service to Airway Heights. Airway Heights has PFAS in their wells and is now relying on City of Spokane for its water.

EPA provides funding to Health Office of Drinking Water for set-aside activities and source water protections. Health can use these funds in limited circumstances to defray costs of additional water testing.

Other funding programs in the state could be tapped for loans or grants to help with costs of new infrastructure in response to PFAS contamination:

- Public Works Assistance Account overseen by Public Works Board.
- Community Development Block Grant overseen by Department of Commerce.
- Rural Development loans and grants overseen by U.S. Department of Agriculture.

Public water systems can pursue reimbursement from potentially liable parties under the state MTCA when PFAS are declared hazardous substances under MTCA. Even under MTCA, water systems may have to carry costs long-term or permanently because:

- The process of identifying responsible parties and being reimbursed can take years.
- Responsible parties may be difficult, if not impossible, to determine.
- The potentially liable party could be a local entity under the same public administration as the water utility (for example, a local fire station).

In each of these cases, the costs borne by the water system would be long-term or permanent.

Why?

Without funding, public water systems and their ratepayers must absorb what can be a costly response. Funding would support water systems when they must:

- Investigate contamination sources.
- Find an alternative water source.
- Design and install expensive filters on contaminated water sources.
- Maintain and monitor new filtration systems.

When PFAS concentrations in drinking water supplies exceed health advisory levels, timely mitigation may be needed to protect human health. This can create immediate costs to water systems.

The water system must explore ways to mitigate the problem, both immediately and long-term. Mitigation planning should aim to minimize cost burdens for lower-income and overburdened communities who are less able to absorb ratepayer cost increases.

Cost

Initial investigation and mitigation costs at PFAS-contaminated sites are reported in the millions of dollars. These costs have been borne by DOD, the water systems or local governments impacted, and the agency programs at Health and Ecology that support water systems and contaminated site cleanup.

For example, the Issaquah PFAS Pilot Project received \$400,000 through the State Building Construction Account for ground water assessment work to be conducted during the 2019 – 2021 biennium. An additional \$300,000 was requested for this project from the 2020 program operating budget, but these funds are not yet available as a result of COVID-19 related impacts to Ecology's operating budget. In future years, 2021 – 2023, additional resources of \$750,000 are being requested for additional groundwater investigation.

With mitigation measures identified, implementation funds are sought from granting sources as described above. In addition to costs for investigating the source of the contamination, filter maintenance and monitoring also require ongoing expenditures. Such costs could also be covered under grants, but may require additional resources from water supply systems.

Each contaminated drinking water site has specific needs, which complicates cost estimation. Without knowing the number of impacted systems in the state, we are unable to estimate total costs to implement this recommendation.

1.2 Provide technical support for site characterization, source investigation, and mitigation at contaminated sites

Recommendation:

Ecology and Health will continue to develop expertise and provide technical assistance and guidance to drinking water purveyors, local jurisdictions, and responsible parties in order to address PFAS contamination and conduct cleanup actions.

Those actions include:

- Ecology will continue to collaborate with involved parties at PFAS contamination sites in the state. These efforts will help to better understand the sources, composition, and distribution of PFAS contamination in soil and water. Identification and evaluation of appropriate cleanup actions and their costs will be informed by this work. This work is being done within Ecology's existing resources.
- Health will continue to provide water systems with advice and assistance to
 understand the mitigation options and guide voluntary action on unregulated PFAS
 until the rulemaking for PFAS in drinking water is complete. To-date, technical
 assistance has focused on public water systems near military bases with PFAS
 detections in groundwater. Department of Health continues to include local health
 departments in outreach and guidance. This work is being done within Health's
 existing resources.
- Ecology will look at targeting Safe Drinking Water Action Grants (a category of Remedial Action Grants for Local Governments) to help address PFAS-contaminated drinking water, once some of the uncertainties discussed above have been addressed.
- Ecology will investigate PFAS contamination in groundwater and surface water.
 These efforts would support local health departments, cities, counties, and other public entities in Washington when PFAS contamination is discovered. Initial investigation efforts could identify areas at high risk of contamination. This could include areas where trainings or firefighting activities used large quantities of PFAS-containing AFFF, or where spills released the foam. Ecology could prioritize funding for site-specific assessments and groundwater testing. Funding for this action is estimated below.
- Ecology will consider the number of people impacted, the concentration of the PFAAs in the drinking water, and vulnerable populations present when prioritizing mitigation and cleanup activities. Ecology will use mapping tools such as Environmental Justice (EJ) screen and Information by Location (IBL) in the Washington Tracking Network (WTN) portal to characterize the demographics of the population served by impacted drinking water.

 Obtain chemical identities from products and at contaminated sites to find chemical "fingerprints" useful in identifying source locations. Analytical methods may not yet be developed to obtain all the required data.

Why?

Technical assistance helps people understand the advantages and disadvantages of various options to reduce levels of PFAS in water and soil.

Variation in environmental conditions and contamination sources makes site characterization difficult. Site-specific conditions should inform the selection of appropriate actions. Evolving cleanup methods, plus the differences unique to a site, lead to difficulty estimating costs.

PFAS have unusual properties and research into their movement through soils and aquifers is ongoing. Further, PFAS contamination sources need to be investigated. To recover mitigation costs, Ecology must identify the party or parties responsible for the source of contamination. Local water districts and governments often lack the expertise and resources to investigate sources of PFAS contamination.

Cost

To support PFAS investigations as needed, Ecology requested resources from the Legislature to:

- Provide monitoring assistance to local jurisdictions when PFAS contamination is discovered.
- Assist with investigations, including researching potential sources, collecting samples, conducting laboratory analysis, and installing monitoring wells.

This type of environmental monitoring work was funded in 2020 and 2021 through the approved state 2020 supplemental budget.⁴⁷

1.3 Support biomonitoring and other health studies to answer important health questions

Recommendation:

Health should continue to find opportunities for Washington residents to participate in exposure and health studies. These studies help answer important community and public health questions about PFAS exposure and health outcomes. For example, Health requested and supports inclusion of Airway Heights as one of eight sites in the ATSDR's PFAS Exposure Assessment study. Health also applied for but was not awarded a cooperative agreement to include a Washington site in the ATSDR Multisite PFAS Health Study.

⁴⁷ https://ofm.wa.gov/sites/default/files/public/budget/statebudget/20supp/Z-0776.2Operating.pdf

State agencies should also support investigations into pathways of PFAS contamination in food, drinking water, and indoor environments. They should pursue policies to mitigate and reduce these sources of human exposure over time.

Why?

Biomonitoring can help us understand how best to reduce human exposure to PFAS. Biomonitoring helps people compare their PFAS exposure level to national averages, and could connect residents to health information as it becomes available. Further health studies are needed to better understand the impacts of PFAS on human health.

Cost

Biomonitoring studies are expensive and the state would need funding to support these types of investigations. Additional funding could be secured through competitive grants for such activities. Benchmark costs have been estimated based on reports from several sites in the U.S. where biomonitoring testing has been conducted for residents near areas of PFAS contamination. Costs averaged up to \$1,000 per person tested.

2.0 Manage environmental PFAS contamination

PFAS have contaminated soil, groundwater, and surface water in certain Washington locations. To reduce exposure and protect human health, these contaminated areas may require a variety of responses. In Washington, PFAS-containing Class B firefighting foam used in firefighter training seems to be the primary source of drinking water supply contamination. Nationally, in addition to firefighting foam use, numerous sources—including manufacturing, industrial processes, and improper waste disposal—result in PFAS contamination.

Previous CAP Recommendations

Interim CAP

The Interim CAP proposed several areas of action pertaining to managing environmental PFAS contamination.

First, the Interim CAP recommended developing PFAS cleanup levels for soil and groundwater. This recommendation was reiterated in the Preliminary Recommendations, and is presented again in this Draft CAP, as Recommendation 2.1 below.

Second, the Interim CAP recommended identifying methods to reduce exposure to contamination. The recommendation focused on developing expertise, providing technical assistance and guidance to parties that address PFAS contamination, and collaborating with parties to better understand the sources, composition, and distribution of PFAS contamination in soil and water. This recommendation was also reiterated in the Preliminary Recommendations, and is included as Recommendation 2.1 below.

The Interim CAP recommended reducing risks to drinking water from firefighting foam. This included implementing AFFF notification and restrictions, surveying firefighting foam users to identify high-risk sites, developing outreach on responsible AFFF use, and replacing PFAS containing AFFF in non-exempt uses. It also recommended providing assistance for firefighting personal equipment notifications.

These recommendations focused on the future implementation of the Firefighting Agents and Equipment Toxic Chemical Use law (Chapter 70.75A RCW⁴⁸), passed in 2018. This law establishes restrictions on Class B firefighting foam, which contain intentionally added PFAS chemicals:

- As of July 1, 2018, prohibits use of Class B firefighting foam for training.
- As of July 1, 2020, prohibits the manufacture, sale, and distribution of Class B firefighting foam.
 - Interim exemptions include federally required users, petroleum storage and distribution facilities, or certain chemical plants.
- Requires manufacturers to notify Washington purchasers about the presence and purpose of PFAS in firefighting personal protective equipment.
- Two years after amendment of federal regulations (prohibiting the use of PFAS-containing foam), requires federal facilities to use non-PFAS foams that comply with the new federal regulation.
 - Airports must inform Ecology about their ability to switch to non-PFAS foams within 18 months of the change in federal regulations.
- Beginning 2024, restricts the purchase of PFAS-containing foams by oil terminals, oil refineries, and chemical plants.

Finally, the Interim CAP identified use of PFAS in industry or manufacturing as a potential source of release or environmental exposure. It recommended identifying potential industrial point sources of PFAS in the state and considering outreach on best management practices for handling and disposing of PFAS-containing wastes. This recommendation was also included in the Preliminary Recommendations.

Preliminary Recommendations

In addition to establishing cleanup levels and identifying methods to reduce exposure to contamination noted above, the Preliminary Recommendations proposed partnering with local organizations in communities with contamination, and providing them funding to lead messaging, consultation, and solution identification for PFAS contamination issues. This represents Recommendation 2.2 below.

The Preliminary Recommendations continued to focus on implementation of Chapter <u>70.75A</u> RCW, ⁴⁹ and future work is identified in Recommendation 2.3 below.

⁴⁸ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

⁴⁹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

Upon enactment of Chapter 70.75A RCW, Ecology <u>received funding</u>⁵⁰ of approximately \$215,000 for fiscal years 2017 through 2019 to implement the Law. Since the summer of 2019, Ecology has conducted the following activities to implement the law.

- Developed an agency website to provide more information and outreach materials regarding the requirements of the law.⁵¹
- Conducted outreach to manufacturers to explain the requirements and ensure compliance with the restrictions.
- Collaborated with firefighting foam users on the restriction of PFAS-containing firefighting foam use in training, and on the purchase restriction taking effect in 2020.
- Provided technical assistance to state and local governments and other jurisdictions to help them purchase PFAS-free Class B firefighting foam.
- Consulted with the Department of Enterprise Services to develop procurement preferences for state and local governments and other jurisdictions to purchase PFAS-free Class B firefighting foam alternatives.
- Informed firefighting personal protective equipment manufacturers of the requirement to notify purchasers about the presence of PFAS—and requested copies of the notification.
- Surveyed state and local governments and other jurisdictions about stocks of Class B firefighting foam through Ecology's <u>Product Replacement Program</u>.⁵²

Ecology also received funding through its Product Replacement Program to collect, transport, and dispose of PFAS-containing firefighting foam owned by the state's municipal fire departments by June 2021. This program intends to dispose of 30,000 to 40,000 gallons of foam. Ecology expects this activity to cost between \$300,000 and \$500,000.

Ecology conducted a review of this collection and disposal activity under the State Environmental Policy Act (SEPA). In September 2020, Ecology issued a <u>Determination of Non-Significance</u>⁵³ and associated <u>checklist</u>⁵⁴ documenting this <u>review</u>. ⁵⁵

The Preliminary Recommendations also addressed identifying industry sectors in Washington that also carry AFFF stocks or use commercial quantities of PFAS and finding opportunities to reduce such usage. These recommendations are also represented in Recommendation 2.3 below. This recommendation acknowledges new sources of information, namely EPA's Toxics Release Inventory, which can supplement our knowledge of PFAS used in industry in the state.

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⁵⁰ https://fnspublic.ofm.wa.gov/FNSPublicSearch/GetPDF?packageID=53000

⁵¹ https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Addressing-priority-toxic-chemicals/PFAS/Toxics-in-firefighting

⁵² https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Product-Replacement-Program

⁵³ https://apps.ecology.wa.gov/separ/Main/SEPA/Document/DocumentOpenHandler.ashx?DocumentId=97538

⁵⁴ https://apps.ecology.wa.gov/separ/Main/SEPA/Document/DocumentOpenHandler.ashx?DocumentId=97539

⁵⁵ https://apps.ecology.wa.gov/separ/Main/SEPA/Record.aspx?SEPANumber=202004521

Health is pursuing activities to protect human health from adverse impacts of environmental PFAS contamination based on data collected since PFAS CAP development began. Health is currently developing fish consumption advisories for PFOS in freshwater fish based on Ecology fish sampling data. Health received additional data from Ecology in 2019 to provide an adequate basis for a fish consumption advisory. Health is reviewing these data and reevaluating screening levels and considering recent changes in recommended oral intake.

2.1 Establish PFAS cleanup levels for soil and groundwater

Recommendation

- Using existing authority under MTCA, Ecology will develop cleanup levels for PFOA and PFOS (and additional PFAS as appropriate). Ecology will use the SBOH's drinking water standards or other advisories adopted in rule to develop these cleanup levels.
- Ecology will explore methods for investigation and cleanup of PFAS contamination.
- Ecology will conduct monitoring for PFAS compounds in environmental media (soils, surface water, and sediment) and wildlife tissue to identify sources and assess exposure.
- Based on data collected, Ecology will consider developing cleanup levels for individual or mixtures of PFAS in soil, sediment, freshwater, and saltwater to protect ecological receptors.
- In this context, the following activities will be implemented to support activity under the recommendations above:
 - Trophic transfer and bioaccumulation of PFAS compounds should be further evaluated in aquatic and terrestrial food webs to further understand exposure.
 - Selected individual PFAS compounds, as well as common PFAS mixtures, should be evaluated for ecotoxicity in aquatic and terrestrial biota, using both laboratory and field methods.
 - Ecological risk assessment should be performed for PFAS compounds by detailing exposure and effects in order to estimate risks to nonhuman biota.
 - An uncertainty analysis should accompany PFAS ecorisk assessment to promote transparency in the risk assessment and communication processes and to more clearly identify data gaps.
 - Results of these risk assessments should support potential interventions (for example, species protections) and characterization of potential impacts on ecological services.
- Ecology will provide information to interested parties about cleanup efforts.

Why?

Ecology establishes cleanup levels for hazardous substances in the environment. The cleanup level concentrations, under specific exposure conditions, are considered sufficiently "protective of human health and the environment." Cleanup levels are expected to protect people, overburdened populations, animals, and plants from potentially harmful exposures to

chemicals in the environment. They determine which geographic areas and environmental media have enough contamination to need further evaluation and potential cleanup actions.

Currently, no enforceable federal or Washington state regulatory standards exist to determine whether a site with PFAS contamination requires cleanup or to regulate cleanup of PFAS at contaminated sites. Further, best practices for conducting such a cleanup are not established. Ecological receptors contribute to Washington state's health and economy overall. Collecting additional data and extending cleanup levels to other environmental media is crucial to protecting them.

To support PFAS groundwater contamination investigation in the City of Issaquah, Ecology developed investigatory levels for PFOS and PFOA. These were advisory values, not regulatory cleanup levels. Based on information available at the time, the values were expected to be protective of human health and the environment. Formulation of these advisory levels cost Ecology approximately \$42,000. In order to develop regulatory cleanup levels, Ecology will need to continue evaluating the rapidly expanding body of scientific information related to PFAS.

Cost

The cost to develop cleanup standards is being funded out of Ecology's Toxics Cleanup Program operating budget, and is expected to be similar to the cost of developing the advisory levels described above. This estimate does not include work to collect additional exposure data, nor to develop cleanup levels for other environmental media (sediment and surface water).

Costs to develop and evaluate methods for addressing PFAS contamination are difficult to estimate due to significant uncertainties around:

- How (and in what concentrations) most PFAS affect people, animals, and plants.
- How best to measure the types and amounts of PFAS in the environment.
- How PFAS move through the environment and change over time.
- How to effectively clean up environmental PFAS contamination—including factors like protectiveness, feasibility, and cost.

Ecology is planning to conduct additional environmental monitoring in 2020 and 2021 funded through the approved state $\underline{2019 - 2021}$ supplemental budget, ⁵⁶ however specific projects have not yet been selected.

⁵⁶ https://ofm.wa.gov/sites/default/files/public/budget/statebudget/20supp/Z-0776.20perating.pdf

2.2 Partner with local organizations in communities with contaminated water or contaminated sites

Recommendation

Department of Health will identify local health departments or community-based organizations to address health equity related to contaminated sites in public communications. Health will coordinate with Ecology to distribute funding to those organizations selected for assistance. Health's new Community Engagement Guide⁵⁷ may support this effort.

Funded organizations would:

- Address potential health equity issues through culturally and linguistically informed engagement.
- Find trusted messengers or platforms to deliver audience-tested risk communication messages to engage historically overburdened and higher risk populations.
- Support impacted populations in finding their own solutions through collective action and decision-making.
- Engage the community throughout the course of the public health response, source investigation, and site cleanup.
- Invite area residents to actively participate on advisory committees, in site information meetings, and in public decision-making about remediation.
- Aim to remove participation barriers by providing child care, reducing transportation costs, and planning for convenient meetings times at familiar locations.
- When possible, appropriately compensate community advisors for participation—particularly in areas with low-income populations.

Why?

When testing identifies PFAS in drinking water in a new community, it can be challenging to communicate effectively with area residents.

Communities are unique, and there may be:

- Cultural and language barriers to effective communication.
- Economic, systemic, and social barriers to acting on public health advice.

These barriers disproportionately affect low-income and other historically overburdened communities, including communities of color. During PFAS investigation and mitigation, state agencies should collaborate with local leadership and organizations to strengthen community awareness and engagement.

Community-based and community-led organizations (that are rooted in and that directly serve these communities) can offer meaningful engagement support. For example:

 A recent \$120,000 two-year grant funded a local organization providing educational materials and conducting outreach in a community impacted by industrial activities.

⁵⁷ https://www.doh.wa.gov/Portals/1/Documents/1000/CommEngageGuide.pdf

• In one affected community, a local church group volunteered to distribute bottled water to elderly and disabled residents.

Cost

If PFAS are classified as hazardous substances under MTCA, community-led public engagement would be eligible for funding through Ecology's Public Participation Grant program (in the Contaminated Site Project category). Designated PFAS funds should be allocated specifically to PFAS-related impacts to communities.

Local outreach efforts depend on the extent and type of community outreach required for a specific contamination concern. As such, at this time, it is not possible to estimate the funding needed for these efforts.

2.3 Work to prevent PFAS releases from firefighting foam use and manufacturing

Recommendation

Ecology will work proactively with industry, manufacturers, and businesses to eliminate releases to the environment from the use of PFAS-containing AFFF or other manufacturing processes using PFAS.

To address PFAS in AFFF, Ecology would:

- Collaborate with users of firefighting foam to develop and share outreach materials and best management practices that address the proper use, storage, and disposal of PFAS-containing AFFF.
- Ensure that industrial use of PFAS-containing AFFF provides for containment procedures along with collection of this foam and contaminated soil or sediment for proper designation and disposal. Costs to industrial users to collect and dispose of released PFAS-containing AFFF include plan development, employee training, methods for containment, and disposal of waste.
- Continue identification of organizations and industries which store and use AFFF in training and emergency firefighting, including use of AFFF in tunnels.
- Assist state, and local governments, airport, industry, and fire districts with prioritizing the quantification, disposal and replacement of PFAS-containing AFFF, especially in communities with cumulative impacts, health disparities, and environmental justice considerations.
- Share information about PFAS-free Class B firefighting foam with users of firefighting foam as information or research is available, including GreenScreen® certifications.
- Conduct compliance and enforcement actions to ensure the law is being followed.

Ecology will work proactively with industry, manufacturers, and businesses to eliminate releases to the environment from the use of PFAS-in manufacturing or other processes.

- Ecology will review data from other states and countries to identify industrial or manufacturing uses of PFAS. Ecology will use this information to identify industries in Washington that have used or continue to use commercial quantities of PFAS. Ecology will also track future TRI reports (starting 2021) for industries.
- Ecology will evaluate PFAS release potential from those industries which may have, or continue to, use PFAS.
- Ecology will reach out to these industries to discuss their use of PFAS, identify opportunities to switch to safer alternatives, implement best practices, and ensure proper waste management.

Why?

PFAS-containing Class B firefighting foam has been associated with drinking water contamination in Washington state. In their risk-based efforts to identify and mitigate PFAS in drinking water, both the military and Health focus on firefighting foam release sites. However, firefighting foam is not the only likely source of PFAS in state drinking water. Other states that are expanding testing for PFAS in drinking water have identified sources such as:

- Landfills.
- Industrial waste stream sludges.
- Manufacture of waterproof leather shoes.
- Manufacture of parchment paper.
- Taxidermy.
- Textile coating.
- Metal plating and finishing.
- Car washes.
- Pulp and paper mills.

More work is needed to understand PFAS use, sources, pathways of exposure, and effects on human health and the environment.

Cost

Ecology identified additional foam stockpiles managed by commercial airports, manufacturing, and transportation facilities that represent a large pollution source, but do not currently qualify for the disposal program established under Chapter 70.75A RCW.⁵⁸ Ecology estimates that it will cost between \$400,000 and \$650,000 to collect, transport, and dispose of such foam, including 0.25 FTE to manage this program. Ecology has included this cost in its FY 2021 – 2023 budget request.

Ecology has requested approximately \$36,000 for monitoring and compliance activities to be conducted under Chapter 70.75A RCW in FY 2021 - 2023.

⁵⁸ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

Ecology estimates that supporting industry with investigation and reduction of non AFFF-related PFAS use would require the resources of 0.25 FTE for one year, at the cost of approximately \$50,000. This funding has not yet been budgeted or requested.

3.0 Reduce PFAS in products

A wide variety of industrial, commercial, and consumer products use PFAS. People can be exposed to the PFAS in consumer products when they use products, or as PFAS accumulate in indoor air and dust. Although PFOA and PFOS are not readily absorbed through skin, residues on hands can be absorbed if swallowed.

Previous CAP Recommendations

Interim CAP

The Interim CAP proposed several areas of action pertaining to reducing exposures from products (other than AFFF and fire-fighting turnout gear already discussed above):

- Identifying sources of PFAS exposure in the home resulting from PFAS present in carpets, textiles, cosmetics, waxes, and cleaning agents.
- Conducting alternatives assessments for uses of PFAS with the highest potential for human exposure.
- Completing an alternatives assessment of PFAS-containing food contact materials.

Within the timeframe of the issuance and revision of the Interim CAP, the Washington State Legislature adopted the Pollution Prevention for Healthy People and Puget Sound Act (Chapter 70.365 RCW⁵⁹), creating a process for Ecology, in consultation with Health, to regulate classes of chemicals in consumer products. The Legislature also adopted the Packages Containing Metals and Toxic Chemicals law (Chapter 70.95G RCW ⁶⁰) restricting PFAS in food packaging materials.

Ecology is implementing Chapter 70.365 RCW through the Safer Products for Washington program. The law directs us to work with stakeholders, report to the Legislature, and do four things on a repeating, five-year cycle:

- Identify at least five priority chemicals, based on hazard, exposure, and impacts.
- The first priority chemicals identified are bisphenols, organohalogen flame retardants, phthalates, polychlorinated biphenyls, and PFAS.
 - Identify consumer products that are significant sources of exposure to the priority chemicals for people and sensitive species.
- Determine regulatory actions to reduce exposure to people and sensitive species.
- Adopt rules to implement regulatory actions, which could include reporting requirements or restrictions on the use of a chemical in a product.

⁵⁹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365&full=true

⁶⁰ https://app.leg.wa.gov/RCW/default.aspx?cite=70.95G&full=true

Under the law, we will identify products that are significant sources of PFAS exposure and determine whether regulatory actions are needed to reduce exposures.

Chapter 70.95G RCW includes the following restrictions:

- Effective January 2022, prohibits PFAS in plant fiber-based food packaging.
- Requires Ecology to conduct an assessment to identify safer alternative products.
 This assessment must consider chemical hazard, performance, cost and availability, and exposure.
 - Ecology must submit the findings for external peer review and publish the results in the Washington State Register.
- Requires Ecology to report results to the Legislature before a ban on PFAS in food packaging can take effect.

Ecology has completed—or is conducting—the following work to implement the law:

- Following a competitive bid process, Ecology selected SRC, Inc. to conduct the alternatives assessment (AA).
- A statement of work, deliverables, and timeline are available on the <u>PFAS in food</u> packaging AA website.⁶¹
- SRC, Inc. and Ecology are working with a broad range of interested parties to gather the information needed for the assessment. Ecology and SRC, Inc. regularly conduct webinars to update interested stakeholders and the public on the project.
- Our analysis and research focuses on food wrappers, liners, and other packaging categories such as plates, bowls, trays, and take-out containers. Ecology and SRC, Inc. are reaching out to end-users of these products for input on the use of alternatives in the market.
- Ecology requested an external peer review by the Washington State Academy of Sciences and will publish the results in the Washington State Register.

Preliminary Recommendations

The Preliminary Recommendations moved the consideration of PFAS in products into its own main recommendation, focused on implementation of Chapter <u>70.365</u> RCW.⁶² This reorganization represents Recommendations 3.1 and 3.2 below.

Since issuing the Preliminary Recommendations in 2019, we continued implementing these product laws as follows:

- Under the Safer Products for Washington program, Ecology completed the following:
 - Published a <u>draft report</u>⁶³ in January 2020 recommending pretreated carpets and rugs and aftermarket treatments for carpets and rugs as priority consumer products for further research.

⁶¹ https://www.ezview.wa.gov/site/alias__1962/37610/pfas_in_food_packaging_alternatives_assessment.aspx

⁶² https://app.leg.wa.gov/RCW/default.aspx?cite=70.365&full=true

⁶³ https://fortress.wa.gov/ecy/publications/summarypages/2004004.html

- Reviewed comments received on the draft.
- Expanded the categories considered to also include water and stain resistance treatments as well as leather and textile furnishings as priority consumer products.
- Submitted the final <u>version of the report</u>⁶⁴ to the Legislature on July 6, 2020.
- The Children's Safe Products Act (CSPA), Chapter 70.240 RCW,⁶⁵ requires manufacturers to annually report the presence of certain chemicals (including PFOS and PFOA) in children's products sold in Washington state. Ecology implements the law as follows:
 - o Ecology receives manufacturer reports and conducts compliance activities.
 - Manufacturer reports are <u>published online</u>.⁶⁶

3.1 Reduce PFAS exposure from carpets and rugs, water and stain resistance treatments, and leather and textile furnishings

Recommendation

Under Chapter 70.365 RCW, Ecology identified carpets, water and stain resistance treatments, and leather and textile furnishings as significant sources and uses of PFAS. We recommend that Ecology determine whether safer alternatives are feasible and available. These determinations should be made by June 2022 and would be accompanied by proposed regulatory actions to reduce exposure.

Regulatory actions could include:

- Requesting that manufacturers:
 - Identify products that contain PFAS.
 - o Disclose their use of priority chemicals in product ingredients.
 - o Release information on exposure and chemical hazard.
 - Describe the amount and function of PFAS in products.
- Proposing restrictions on PFAS-containing carpet and carpet care products when a safer alternative is available and feasible, and the restriction will reduce a significant source of PFAS or protect sensitive populations or species.
- Proposing actions to reduce legacy PFAS-containing carpet and carpet care products remaining in homes, especially in low-income households, where items may be retained past the typical product lifespan.

In addition to the above, we recommend the following actions:

• Implement a purchasing preference policy for PFAS-free carpet. Work with vendors on the state flooring contract to offer PFAS-free carpet on all state master contracts

⁶⁴ https://fortress.wa.gov/ecy/publications/summarypages/2004019.html

⁶⁵ https://app.leg.wa.gov/RCW/default.aspx?cite=70.240

⁶⁶ https://hpcds.theic2.org/Search

- and all agency contracts. Purchasing PFAS-free carpet could result in increased costs to the state.
- If safer alternatives are available, include them in Ecology's <u>Product Replacement Program</u>⁶⁷ to replace PFAS-containing carpet in community centers, libraries, daycares, and other environments where children may be disproportionately exposed.

Why?

According to EPA, some of the most significant sources of human exposure to nine PFAS in the U.S. are carpets and commercial carpet-care liquids. Treated carpet in homes and offices can contribute to PFAS in indoor environments. Infants and children have higher exposure due to inhalation and ingestion of house dust. California DTSC identified PFAS in carpet as a <u>priority product</u>⁶⁸ under the Safer Consumer Products program. San Francisco adopted a <u>comprehensive carpet regulation</u>⁶⁹ prohibiting the use of PFAS.

Cost

The Legislature funds these efforts under the Safer Products for Washington program. Ecology received \$807,000 for the 2019 – 2021 biennium, and a supplemental \$479,000 in FY 2020 to implement the program as a whole, for a total of \$1,286,000 through June 2021. As described in its July 2020 report to the Legislature (see above), Ecology identified eleven priority products, of which three were PFAS-related (carpets, water and stain resistance treatments, and leather and textile furnishings).

Because program activities are conducted as a whole, it is not possible to distinguish program costs attributed to only the PFAS-related priority products. However, one could approximate the PFAS-related costs as a proportion of entire program costs based on the number of priority products identified (i.e., three of eleven). Thus, the cost of activities associated with identifying PFAS-related priority products under Chapter 70.365 RCW would be approximately \$351,000.

A portion of the cost estimated above will also go toward work completed by the end of June 2021 related to proposing regulatory actions. Ecology will request additional funding to determine regulatory actions by the end of June 2022, as described in Recommendations 3.2 and 3.3 below.

At this time, Ecology has not estimated the cost of additional actions (i.e., implementing a purchasing preference policy and replacing PFAS-containing carpet under the Product Replacement Program). Ecology is already funding a staff position to coordinate the identification of viable purchasing preference policies with the Washington State Department of Enterprise Services for a number of products, including PFAS-containing carpet.

⁶⁷ https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Product-Replacement-Program

⁶⁸ https://www.dtsc.ca.gov/SCP/carpets and rugs containing pfas.cfm

⁶⁹ https://sfenvironment.org/sites/default/files/policy/regulation_sfe-2018-01-ppo_gbrcbo.pdf

Establishing the cost of replacing carpet in community centers, libraries, daycares, and other environments where children may be disproportionately exposed would require an estimate of the number of facilities, and the square footage of carpet to be replaced. Funding could then be requested by Ecology's Product Replacement Program.

3.2 Identify additional sources and uses of PFAS to consider in the second Safer Products for Washington cycle

The priority products identified in 2020 under the Safer Products for Washington program do not account for all sources and uses of PFAS. We will continue research to better understand how other products contribute to PFAS concentrations in homes, workplaces, and the environment. These include PFAS in:

- Water resistant clothing and gear.
- Nonstick cookware and kitchen supplies (e.g., baking paper).
- Personal care products (e.g., cosmetics and dental floss).
- Cleaning agents.
- Automotive products.
- Floor waxes and sealants.
- Ski waxes.
- Car waxes.

Recommendation

Ecology should engage with overburdened communities regarding consumer products that may contain PFAS. Communities use consumer products differently. It is important that the team identify consumer products which might be disproportionately exposing overburdened communities.

In addition to determining whether these products are significant sources or uses of PFAS, preliminary investigations into the availability and feasibility of safer alternatives should be conducted. This will help the Safer Products for Washington team identify opportunities to reduce additional sources and uses of PFAS. If safer alternatives are identified, outreach should be conducted to increase voluntary adoption. If these products are identified as significant sources or uses, they should be added as priority products in 2025 and regulatory actions should be determined by 2027.

Why?

People are exposed to PFAS in their homes when they use products, and via exposure to house dust that contains PFAS. Ingesting contaminated food and drinking water leads to the greatest portion of chronic exposure to PFAS (specifically to PFOS and PFOA) for the general population.

PFAS-containing products in the home and in some occupations can be additional sources of exposure. High PFAA levels were identified in ski waxes, leather samples, outdoor textiles, and

some baking papers. Studies of indoor air and house dust indicate that PFAS exposure occurs from products in the home, such as carpet care liquids, nonstick cookware, food packaging, and waterproof clothing. Many other consumer products may contain PFAS ingredients (see the list above). Research is needed to understand how these products contribute to human exposure.

Cost

Ecology will make budget requests to fund future cycles of the Safer Products for Washington Program, including consideration of the products listed above.

Ecology estimates that the costs of future cycles of product consideration under Safer Products for Washington would be similar to those incurred to-date (see <u>Recommendation 3.1</u> above), but could vary based on the complexity and the number of chemical-product combinations considered. Costs to identify and implement regulatory actions in future cycles are expected to be similar to those identified in Recommendation 3.3 below.

3.3 Implement other reduction actions for PFAS in products

Recommendation

Identify regulatory actions to reduce exposures and releases to the environment from the priority consumer products containing PFAS identified in Recommendation 3.1 above. Additional research, testing, and investigation would be required to determine appropriate reduction actions. These activities will be conducted in the 2022 – 2023 biennium. Actions could include:

- Under Chapter 70.365 RCW, 70 propose restrictions of PFAS in priority consumer products when a safer alternative is feasible and available, and the restriction will reduce a significant source of PFAS or protect sensitive populations or species. Ecology could:
 - Determine that no regulatory action is currently required.
 - o Require a manufacturer to provide notice of the use of PFAS.
 - Restrict or prohibit the manufacture, wholesale, distribution, sale, or use of PFAS
 in a priority consumer product.
 - Adopt rules to implement these regulatory actions.

In addition to the above, we recommend the following actions:

 Gather input from low-income and other historically overburdened communities, including communities of color. Develop a list of ways to reduce exposure that include low cost and subsidized approaches. These may be particularly important measures to employ in communities with higher exposure from drinking water. No cost estimate is provided to conduct this evaluation or to develop exposure reduction recommendations.

⁷⁰ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365

- Establish a purchasing preference policy for PFAS-free products. Work with vendors
 to offer PFAS-free textiles, furniture, and paints. Apply this policy to all state master
 contracts and all agency contracts. Purchasing PFAS-free products could increase
 state costs.
- Propose a ban on the import or sale of all products in Washington containing
 phased-out long-chain PFAAs. Long-chain PFAAs include perfluorinated carboxylates
 (PFCAs) with eight or more fully fluorinated carbons (for example, PFOA) and
 perfluorinated sulfonates (PFSAs) with six or more fully fluorinated carbons (for
 example, PFHxS and PFOS), their salts, and precursor compounds capable of forming
 long-chain PFAAs.

Why?

Actions need to be implemented to remove or reduce levels of PFAS from products that contribute to human or environmental exposure. Removing chemicals from consumer products can reduce chemicals in indoor air and dust. These actions directly impact human and environmental exposures.

PFOS, PFOA, and related long-chain PFAS compounds are mostly phased-out of U.S. production, but are still produced in other countries. Rather than bans, EPA used voluntary phase-outs and Significant New Use Rules (40 CFR 721.9582) under TSCA to reduce their use. It appears to be legal to import long-chain substances into Washington state for commercial uses, and to distribute and sell products containing them.

Cost

The cost of selecting and implementing regulatory actions under Chapter 70.365 RCW would be funded through the Safer Products for Washington program budget. This funding has not yet been received. As explained above, funding is provided for all priority products selected for regulation, included those that contain PFAS. Ecology estimates costs of approximately \$1.5 million for this aspect of the program as a whole for the period July 2021 – June 2023. PFAS-related costs can be approximated as a proportion of entire program costs based on the number of priority products identified (i.e., three of eleven). On this basis, we estimate that regulatory action for PFAS-related priority products identified in Recommendation 3.1 at approximately \$410,000.

4.0 Understand and manage PFAS in waste

PFAS are released from products people use in their homes and businesses. These releases travel to wastewater treatment plants (WWTPs) and disposal facilities. PFAS entering and passing through these facilities could impact the environment. Investigating PFAS in

Washington's wastewater, landfills, and biosolids is needed to determine PFAS concentrations and inform development of appropriate control actions.

Previous CAP Recommendations

Interim CAP

The Interim CAP identified that handling and disposal of PFAS-containing wastes (including landfilling) required outreach on best management practices.

Preliminary Recommendations

The Preliminary Recommendations explored further evaluation of waste streams that could contain PFAS, creating a dedicated recommendation for evaluating wastewater treatment plant, landfill, and biosolids streams for PFAS contamination. These recommendations are included in Recommendations 4.1 to 4.3 below.

Preliminary Recommendation 4.1 addressed gathering more information about PFAS in WWTP influent and effluent. Ecology received funding to develop and conduct sampling of PFAS in influent and effluent at three municipal WWTPs receiving industrial discharges. This data would help inform which treatment processes are more effective at transforming and removing PFAS. Ecology is working on staffing this project in the context of COVID-19 hiring restrictions so that the work can be completed by June 30, 2021, when the funding expires.

Preliminary Recommendation 4.2 addressed gathering more information about PFAS in landfill leachate. Ecology began this work in 2020. Ecology's Solid Waste Management program developed Phase I of a landfill leachate sampling program. This phase is funded and approved, and is undergoing final planning. (Sampling will begin when COVID-19 restrictions are withdrawn and normal agency field operations resume.) The study will sample leachate at selected landfills in the state to determine the range of values for 33 PFAS substances. Values will be compared to landfills across the country, and the data will be used to:

- Evaluate potential differences in amount of PFAS across landfill cells of different ages.
- Determine whether specific waste streams lead to higher PFAS values. This will identify disposed waste that is likely to release PFAS to leachate.

4.1 Evaluate PFAS in wastewater treatment

Recommendation

Ecology should evaluate PFAS in WWTP influent and effluent to better understand PFAS discharges in Washington state.

 Ecology should develop a study design to sample PFAS in three different types of WWTPs: those with secondary treatment, nutrient removal, and advanced solids removal. Sampling should include products of selected WWTP unit processes (for

- example, primary and secondary clarifiers or dechlorination) to help differentiate removal efficiencies of the different treatment types.
- The study design should ensure that the WWTPs that are sampled receive industrial discharges that are likely to contain PFAS, or that have drinking water sources with known PFAS contamination.
- Ecology should identify industries that are likely to generate wastewater containing PFAS.
- Based on the information from the study, Ecology should consider additional
 monitoring requirements for WWTP dischargers. This should include consideration
 of whether EPA has developed approved analytical methods for PFAS suitable for
 WWTP effluent and a regulatory target (a nationally recommended water quality
 criterion for PFAS) for waters of the state.
- Based on this evaluation, Ecology should require possible PFAS monitoring for some or all domestic and industrial WWTPs.

Why?

PFAS travel from homes, businesses, and industry sources to WWTPs. Once they enter the WWTP, PFAS may partition to different media (for example, solids and liquids). PFAS are subject to aerobic and anaerobic biological processes, and transform into terminal PFAS compounds that resist further natural breakdown. Future WWTP design and operation would benefit from a greater understanding of how different wastewater treatment technologies transform PFAS or remove them from the effluent stream.

Cost

Ecology received \$235,000 to conduct a WWTP sampling study by June 30, 2021. Influent, effluent, and biosolids at three municipal WWTPs receiving industrial discharges will be sampled and analyzed. This includes costs for sample analysis, which can range from \$1,000 to \$1,500 per sample, as well as project staff salaries.

The cost of establishing additional monitoring requirements based on the sampling study has not been determined. More funding sources may be needed to complete this work.

4.2 Evaluate landfill PFAS emissions

Recommendation

Ecology will develop a sampling program at selected landfills across the state. The sampling will test for PFAS in leachate, groundwater, and air emissions.

Leachate

The Solid Waste Management program developed Phase I of the program, involving leachate sampling. With funding and approval, this phase is undergoing final planning. (Sampling will begin when COVID-19 restrictions are withdrawn and normal field operations resume.)

Ecology developed the study to better characterize landfill leachate. The study will:

- Sample leachate at selected landfills in the state.
- Determine the range of values for 33 PFAS substances in leachate, and compare to landfills throughout the country.
- Arrive at an estimate of the total PFAS materials in the landfill leachate through Total Oxidized Precursor (TOP) analyses.
- Determine if differences in amount of PFAS occurs in landfill cells of different ages.
- Determine if specific types of waste streams lead to higher PFAS values.
- Identify disposed wastes that are likely to generate PFAS releases to leachate.
- Perform a one-time testing of leachate from approximately 23 landfills.
- Consider additional sampling of leachate for landfills not yet sampled after the initial Phase I is completed. This second step of Phase I may include landfills that are undergoing MTCA cleanups, or landfills that contain specific refuse streams that have shown to have high PFAS values from the Phase I sampling.

If warranted, Ecology would manage PFAS in landfill leachate long-term by:

- Considering additional monitoring requirements for landfills to test leachate for PFAS using information from the study above.
- Potentially updating the rules (Chapters <u>173-350</u>⁷¹ and <u>351</u>⁷² WAC) to require PFAS testing of leachate during landfill monitoring.

Groundwater and Gaseous Emissions

Phase II of the program will sample groundwater and gas emissions at landfills for PFAS. This phase of the program is in the conceptual stage. Landfills to be sampled will be based on the results of the Phase I leachate study. Groundwater will be sampled from existing monitoring wells.

The Solid Waste Management program, in conjunction with the Air Quality program, will develop the gas emissions sampling portion of the program. Ecology will also monitor landfill gas emissions monitoring being conducted by North Carolina State University and Oregon State University.

Landfill waste makeup

In parallel to landfill gas emission sampling above, Ecology will continue to research the makeup of PFAS waste entering and potentially currently stored in landfills.

Why?

Landfills contain a variety of waste including inert materials (like wood or ash), disposed consumer products, and various organic wastes and solvents. Decomposing waste and rainfall can create leachate that contains water, metallic ions, acids, and other contaminants including

⁷¹ https://apps.leg.wa.gov/wac/default.aspx?cite=173-350

⁷² https://apps.leg.wa.gov/wac/default.aspx?cite=173-351

PFAS. Landfills manage these liquids differently, but they can be a source of contamination—particularly if sent to wastewater treatment.

Cost

The Phase I testing of leachate from 23 landfills has received \$34,500 of funding. Phase II has also received funding for \$55,500, however given delays incurred as a result of COVID-19 restrictions in 2020, it is not clear whether Phase II can be conducted within the originally anticipated timeframe, and it is not known if the funding for this work will remain available.

Adding PFAS monitoring requirements to Chapter 173-350 WAC could take two and a half years and cost up to \$1.1 million. Less complex rulemaking could take two years and cost up to \$260,000. These cost estimates include employee time and expenses, but will vary based on the degree of consultation with Ecology's Assistant Attorneys General.

4.3 Evaluate Washington biosolids management

The information gaps regarding biosolids are significant and currently prevent assessment of risk from PFAS in biosolids land applied in Washington. Any regulatory changes should be founded on defensible data and science-based risk assessments. If scientific modeling is used by Ecology to assess potential PFAS transfer from biosolids to soil or groundwater, realistic model parameters must be used.

Washington biosolids regulation in the near term should ensure sound agronomic land application practices on permitted sites where human exposure is limited. It is premature to add or change regulatory limits given the absence of data from Washington biosolids and problems identified with models and their input parameters.

Recommendation

We recommend the following key steps to address the current data gaps:

- Establish biosolids and soil sample collection and handling methods for PFAS analysis.
- Accredit Washington labs for EPA-validated analysis methods.
- Use EPA-validated analysis methods for biosolids and soils.
- Conduct credentialed third-party review of raw mass spectrometer PFAS data.
- Investigate land application sites where procedures mimic rates and practices under current state rule (Chapter 173-308 WAC⁷³).
- Evaluate realistic exposure pathways.
- Evaluate risk modeling using realistic input values.
- Collaborate with stakeholders to get accurate and precise biosolids data. Initial results should remain anonymous.
- Compile analysis data with statistical review.

⁷³ https://apps.leg.wa.gov/wac/default.aspx?cite=173-308

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To conduct this work, Ecology will collaborate with municipalities managing WWTPs.

Why?

Toxicity, concentration, and pathway of exposure determine the risks contaminants pose to human health and the environment. Fundamental PFAS concentration data to characterize Washington biosolids is lacking. This prevents accurate assessment of PFAS risk resulting from land application under the state biosolids program.

The recommendations work toward securing representative PFAS concentration data that is specific to Washington biosolids. Such data supports models that evaluate human health and environmental risks from contamination.

Cost

As of the date of this CAP, it is not possible to estimate costs for fully implementing this recommendation. Ecology will recruit a senior employee to lead the biosolids data gathering process. Ecology will also submit program funding requests for both sampling and analysis to help with expenses. As indicated under Recommendation 4.1, a limited biosolids sampling and analysis effort will occur as part of funding approved to sample WWTP influent and effluent by 2021.

PFAS CAP Requirements

We prepared this Draft CAP to meet the requirements of Chapter <u>173-333</u>⁷⁴ Washington Administrative Code (WAC): Persistent Bioaccumulative Toxins (PBTs). An advisory process and the requirements of Chapter 173-333 WAC informed our CAP.

Advisory committee

Ecology created an external advisory committee to provide stakeholder input and expertise (<u>WAC 173-333-430(3)</u>).⁷⁵ Beginning in 2016, we convened committee members from large and small business sectors, federally recognized tribal governments, community organizations, environmental and public health advocacy groups, local governments, and public health agencies. The following organizations, government agencies, and tribal governments are currently represented on the advisory committee:

- American Chemistry Council*
- Agency for Toxic Substances and Disease Registry
- Association of Washington Business
- Carpet and Rug Institute**
- City of Issaguah
- Clean Production Action
- Green Science Policy Institute
- Institute of Neurotoxicology & Neurological Disorders
- Island County Public Health
- King County Department of Natural Resources and Parks
- Washington State Patrol, Fire Training Academy
- Naval Facilities Engineering Command Northwest
- Outdoor Industry Association
- Port Gamble S'Klallam Tribe
- Port of Seattle Aviation
- Port of Seattle Fire Department
- Toxic-Free Future
- University of Washington
- Whidbey Island Water Systems Association
- Whitman College***
- Zero Waste Washington

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^{*}The American Chemistry Council took over representation for FluoroCouncil in August 2020.

^{**}The Carpet and Rug Institute is no longer participating on the committee as of March 2020.

^{***}A professor from Whitman College is representing academia.

⁷⁴ https://app.leg.wa.gov/wac/default.aspx?cite=173-333

⁷⁵ https://app.leg.wa.gov/wac/default.aspx?cite=173-333-430

Beginning in January 2016, we convened meetings of the advisory committee and hosted several webinars. These meetings were open to the public. An Interim CAP was published in April 2018, and was <u>revised in January 2019</u>. We issued <u>Preliminary CAP Recommendations</u> with supporting documentation for advisory committee review in July 2019. Their comments, <u>available on Ecology's PFAS CAP website</u>, 78 were considered while preparing this Draft CAP.

We continue to conduct public outreach via Ecology's PFAS CAP website (where CAP documents are available) and <u>Health's PFAS website</u>, ⁷⁹ and by maintaining a <u>CAP email list</u>, ⁸⁰ through which we distribute information to more than 330 subscribers.

CAP requirements

In addition to the advisory process, the requirements of WAC <u>173-333-420</u>⁸¹ informed the Draft CAP scope. A CAP considers chemical information, production, uses, releases, human health and environmental impacts, and current management approaches. We evaluated the necessary steps and costs of implementing Draft CAP recommendations.

A CAP is advisory in nature. Chapter 173-333-120(1)⁸² WAC "does not impose new requirements on persons using or releasing PBTs, and it does not create new authorities." A CAP does not implement new requirements or mandates on production or use of PFAS. We identify which requirements the Washington State Legislature has enacted and signed into law regarding management of certain PFAS since our process began (see the section Draft CAP Recommendations or Appendix 9: Regulations).

A CAP considers "other chemicals or products that are known or suspected to degrade to the chemical included on the PBT list," such as PFAS precursors (WAC <u>173-333-420(1)(b)</u>). ⁸³ Expanding knowledge of PFAS as a class shapes the current regulatory environment in Washington, which views PFAS as "a class of fluorinated organic chemicals containing at least one fully fluorinated carbon atom" (Chapters <u>70.95G</u>, ⁸⁴ <u>70.75A</u>, ⁸⁵ and <u>70.365</u> ⁸⁶ Revised Code of Washington (RCW)).

A CAP must consider "the use of available substitutes" (WAC 173-333-420(1)(d)). Our assessment reviews the rapid development of short-chain PFAS to substitute for certain long-

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⁷⁶ https://fortress.wa.gov/ecy/publications/documents/1804005.pdf

⁷⁷ https://www.ezview.wa.gov/Portals/_1962/Documents/PFAS/PrelimRecommendations-2019-PFAS-CAP.pdf

⁷⁸ https://www.ezview.wa.gov/site/alias 1962/37105/pfas chemical action plan.aspx

⁷⁹ https://www.doh.wa.gov/CommunityandEnvironment/Contaminants/PFAS

⁸⁰ http://listserv.ecology.wa.gov/scripts/wa-ECOLOGY.exe?A0=CHEMICAL-ACTION-PLAN

⁸¹ https://app.leg.wa.gov/wac/default.aspx?cite=173-333-420

⁸² https://app.leg.wa.gov/wac/default.aspx?cite=173-333-120

⁸³ https://apps.leg.wa.gov/wac/default.aspx?cite=173-333-420

⁸⁴ https://app.leg.wa.gov/RCW/default.aspx?cite=70.95G

⁸⁵ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A

⁸⁶ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365

chain PFAS. To meet regulatory requirements, we must assess both the opportunities and constraints that substitutes pose. Therefore, we evaluated the body of research on short-chain PFAS as well as long-chain PFAS.

A CAP must consider recommendations for "switching to (iv)" and "encouraging the development of (v)" safer alternatives (WAC 173-333-420(f)). Evaluation of the "availability and effectiveness of safer substitutes (v)(D)" for PFAS uses must form the basis for recommendations. As such, the Draft CAP considers whether substitutes for long-chain PFAS—primarily short-chain PFAS—are safer.

Finally, WAC 173-333-420(i) allows us to include "other information that Ecology determines is necessary to support the decision-making process." Commercially available PFAS—even those intended to be long-chain—often contain a mix of PFAS, including short-chain. Therefore, evaluating how only long-chain PFAS behave would result in a partial understanding of the impacts of commercial products and how PFAS degrade.

Research on the safety of short-chain PFAS is ongoing. Human and environmental health implications of short-chain PFAS are uncertain. EPA has acknowledged the need to finalize draft toxicity assessments and develop additional toxicity values for several PFAS.

Short-chain PFAS tend to be more water soluble and more mobile than long-chain PFAS. This means they can move more easily through soil to contaminate groundwater or surface water, and are harder to remove. For example, due to the persistence of even short-chain perfluoroalkyl acids (PFAAs), exposure to these substances will continue regardless of accumulation because bioaccumulation is not required for sustained internal exposure (see Appendix 6: Ecotoxicology, Section 6.2 Bioaccumulation). If research finds that exposure to short-chain PFAS poses health risks to people or the environment, mitigation may be more difficult or expensive.

We need to understand the combinations of PFAS in waste streams and how they degrade. Studies note the importance of evaluating exposure to precursors and PFAAs separately when considering toxicological risk. Over time, PFAS released to the environment from manufacturing operations transform into a variety of chemical products. The lifetimes and toxicity of transformation and degradation products contribute to uncertain environmental impacts.

Next steps

In accordance with WAC <u>173-333-430(6)</u>,⁸⁷ we are issuing this Draft CAP for review by the advisory committee and the public. We will accept comments through December 7, 2020.

⁸⁷ https://apps.leg.wa.gov/wac/default.aspx?cite=173-333-430

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During the public review period, we will conduct a series of webinar meetings to inform stakeholders about the updated version of the CAP. Stakeholders can register to attend the public comment meetings on the <u>PFAS CAP website</u>.⁸⁸ For more details on the overall CAP development process, see the resource <u>Focus On: PFAS Chemical Action Plan</u>.⁸⁹

After the public review period, we will consider comments from the advisory committee and the public, and, if necessary, conduct additional research. We will issue a Final PFAS CAP in accordance with WAC 173-333-430(7).

⁸⁸ https://www.ezview.wa.gov/?alias=1962&pageid=37105

⁸⁹ https://fortress.wa.gov/ecy/publications/documents/2004048.html

Acronyms

List of acronyms

General acronyms

Table 1. Acronyms found in the CAP summary.

Acronym	Definition
AA	Alternatives assessment
AWB	Association of Washington Business
AFFF	Aqueous film forming foam
ATSDR	Agency for Toxic Substances and Disease Registry
CAA	Clean Air Act
CAP	Chemical Action Plan
CDC	Centers for Disease Control and Prevention
CSPA	Children's Safe Products Act
CWA	Clean Water Act
DEPA	Danish Environmental Protection Agency
DOD	United States Department of Defense
DNRP	Department of Natural Resources and Parks
DON	United States Department of the Navy
DTSC	Department of Toxics Substances Control, California
ECHO	United States Environmental Protection Agency Enforcement and
	Compliance History
Ecology	Washington State Department of Ecology
EFSA	European Food Safety Authority
EJ	Environmental justice
EPA	United States Environmental Protection Agency
EWG	Environmental Working Group
FTE	Full-time equivalent
FY	Fiscal year
Health	Washington State Department of Health
HEPA	Heads of EPAs Australia and New Zealand
IARC	International Agency for Research on Cancer
IBL	Information by location
INND	Institute of Neurotoxicology & Neurological Disorders
ITRC	Interstate Technology Regulatory Council
MTCA	Washington state Model Toxics Control Act
NELAP	National Environmental Laboratory Accreditation Program
NWGA	National Ground Water Association
ODW	Washington State Department of Health Office of Drinking Water
OECD	Organisation for Economic Co-operation and Development
OIA	Outdoor Industry Association

Acronym	Definition
ppt	Part per trillion
PBT	Persistent bioaccumulative toxin
RCW	Revised Code of Washington
SAEPA	South Australia Environment Protection Authority
SBOH	Washington State Board of Health
SEPA	State Environmental Policy Act
SSEHRI	Social Science Environmental Health Institute
SWM	Ecology Solid Waste Management Program
UNEP	United National Environment Programme
WAC	Washington Administrative Code
WTN	Washington Tracking Network
WWTP	Wastewater treatment plant

Chemical Names

Table 2. Chemical name acronyms used in the CAP summary, excluding the general acronyms listed in the table above.

Acronym	Chemical Name
FTOH	Fluorotelomer alcohol
PDSF	Perfluorodecane sulfonate
PFAA	Perfluoroalkyl acid
PFAS	Per- and poly-fluorinated alkyl substances
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonic acid
PFCA	Perfluoro-carboxylic acid
PFD	Perfluorodecanoate
PFDA	Perfluorodecanoic acid
PFDoA	Perfluorododecanoic acid
PFHpA	Perfluoroheptanoic acid
PFHpS	Perfluoroheptane sulfonic acid
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexane sulfonic acid
PFN	Perfluorononanoate
PFNA	Perfluorononanoic acid
PFO	Perfluorooctanoate
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFOSA	Perfluorooctane sulfonamide
PFPeA	Perfluoropentanoic acid
PFPeS	Perfluoropentane sulfonic acid

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Acronym	Chemical Name
PFSA	Perfluorooctane sulfonic acid
PFUnA	Perfluoroundecanoate
POSF	Perfluorooctane sulfonyl fluoride

Appendix 1: Chemistry

1.0 Overview

1.0.1 Findings

PFAS are a class of fluorine-containing chemicals with broad application in commercial products. More than 4,730 PFAS have been registered in the Chemical Abstract Service. EPA's Master List of PFAS Substances includes 7,866 compounds.

Fluorosurfactants are used for their effectiveness in reducing surface energy (of solids) and surface tension (of liquids). Side-chain fluorinated polymers and polyether products help impart oil and grease resistance or soil resistance to food packaging or other substrates. The unique properties of PFAS arise from the strength of the carbon-fluorine bond.

Fluorosurfactants and side-chain polymer PFAS are manufactured from raw materials made by either electrochemical fluorination (ECF) or the telomerization process. Both processes produce end-mixtures of variable composition. The ECF process produces mixtures of various structural shapes (branched chains) and lengths (odd and even). Conversely, telomerization produces a homologous mixture of even chain lengths. Per- and polyfluorinated ethers can be manufactured by several diverse processes, however, comparatively little has been published on the by-products or composition of polyether technical mixtures.

The production and use of long-chain PFAS was voluntarily curtailed in the U.S., Japan, and Western Europe starting in 2002. Following additional regulatory restrictions and voluntary withdrawal campaigns regarding long-chain PFAS, manufacturers in the U.S., Western Europe, and Japan have shifted manufacture primarily to shorter-chain PFAS.

Global PFAS production includes both newer short-chain chemistries and ongoing production of long-chain chemistries in some countries. The transition from legacy products to new chemistries has led to a concurrent increase in what was an already large number of PFAS substances.

This large number of substances—coupled with the fact that products may contain mixtures of target substances, residuals, and contaminants—complicates efforts to understand and characterize PFAS uses, emissions, and impacts.

1.0.2 Introduction

This appendix provides an overview of per- and polyfluoroalkyl substances (PFAS) and a background on their manufacture, and identifies select physical and chemical properties of PFAS relative to their uses. PFAS and their properties have been thoroughly described by others (Buck et al., 2011; ITRC, 2020a, 2020b; Korzeniowski & Buck, 2019a, 2019b, 2019c).

PFAS are a class of fluorine-containing chemicals with broad application in commercial products. More than 4,730 PFAS have been registered in the Chemical Abstract Service (OECD, 2018; Wang et al., 2017). As of November 2019, EPA's Master List of PFAS includes 7,866 chemical compounds (EPA, 2018).

1.1 Subclasses of per- and polyfluoroalkyl substances (PFAS)

1.1.1 PFAS terminology

This section provides a basic definition of PFAS and establishes how these compounds will be described in this appendix.

Definition of PFAS

Buck et al. (2011) and others have provided thorough discussion of PFAS classification (ITRC, 2020b; Knepper & Lange, 2012). Buck et al. (2011) provides the following definition of PFAS:

...the highly fluorinated aliphatic substances that contain one or more C atoms on which all the H substituents (present in the nonfluorinated analogues from which they are notionally derived) have been replaced by F atoms, in such a manner that they contain the perfluoroalkyl moiety C_nF_{2np1} —.

Ordinary **hydrocarbons** contain mostly hydrogen (H) and carbon (C) atoms. However, when the H atoms are completely replaced by fluorine (F) atoms, the substance is described as **perfluorinated**. Figures 2 and 3 provide an example of such perfluorination.

Figure 2 illustrates a non-fluorinated hydrocarbon, octane sulfonic acid. When the hydrogen atoms are replaced by fluorine, one obtains its perfluorinated cousin, perfluorocatane sulfonic acid (PFOS), illustrated in Figure 3.

Figure 3 illustrates the structure of PFOS with all the individual atoms shown. As indicated, PFOS is made up of a chain of carbon (C) and fluorine (F) atoms, with a sulfonic acid tail composed of sulfur (S), oxygen (O), and hydroxyl (OH) components.

Figure 4 simplifies these illustrations. It does not show the C atoms located at the intersection of the straight lines (which represent bonds between the atoms), nor the hydrogens that are attached to carbons. This simplified style will be used throughout the remainder of this appendix.

It is also customary to abbreviate carbon chain-length using the term Cx, where x is replaced by a number indicating the number of carbon atoms in the chain. For example, C6 would represent a chain length of six carbon atoms.

Figure 2. Non-fluorinated hydrocarbon, octane sulfonic acid.

$$\begin{array}{c|c} H & H & H & H & H & H & H \\ \hline H & C & C & H & C & H & C & H \\ \hline H & C & C & H & C & H & C & H \\ \hline H & C & C & H & C & H & C & H \\ \hline \end{array}$$

Figure 3. Fluorinated hydrocarbon, PFOS.

Figure 4. Simplified illustration of octane sulfonic acid (left) and PFOS (right) chemical structure where C and H are not shown.

Moiety (R-group)

A moiety (or R-group) is a part of a molecule that can be found in other types of molecules, and is given a typical name. For convenience, structure illustrations often use R to represent a "functional group" add-on to the main carbon chain. R may represent a single atom or a group of atoms. For the PFOS example used in Figures 2, 3, and 4 above, the sulfonic acid group (SO₃H) is the R-group in PFOS. Figure 5 illustrates the relationship of the R-group (SO₃H) to the remainder of the PFOS chemical structure. Chemical manufacturers may alter the R-group in PFAS to achieve desired properties, such as solubility in a formulation solvent.

Figure 5. PFOS in which the SO₃H functional group is represented by R.

Long-chain vs. short-chain PFAS

Much of the regulatory interest around PFAS in the environment has focused on PFOS and PFOA. Both of these chemicals are long-chain assemblages of fluorine and carbon atoms. In scientific literature, researchers distinguish between long-chain and short-chain PFAS as follows (ITRC, 2020b; OECD, 2013).

Long-chain refers to:

- Perfluoroalkyl carboxylic acids (PFCAs) with eight or more carbons (seven or more carbons are perfluorinated).
- Perfluoroalkane sulfonates (PFSAs) with six or more carbons (six or more carbons are perfluorinated).
- Short-chain refers to:
 - Perfluoroalkyl carboxylic acids with seven or fewer carbons (six or fewer carbons are perfluorinated).
 - Perfluoroalkane sulfonates with five or fewer carbons (five or fewer carbons are perfluorinated).

Regardless of the chain length distinction described above, and although some PFAS exhibit similarities based on chain length, PFAS behavior is not entirely based on chain length (ITRC, 2020b).

1.1.2 Overview of PFAS

PFAS are a large family of compounds with varying physical and chemical properties. In their manufactured form they can be gases (for example, perfluorobutane), liquids (for example, fluorotelomer alcohols), surfactants (for example, perfluorooctane sulfonate), and high-molecular weight polymer solids (for example, polytetrafluoroethylene [PTFE]) (ITRC, 2020a). The family of PFAS has been subdivided into two primary classes (Buck et al., 2011; ITRC, 2020a). These include polymers and non-polymer substances. Table 3, adapted from Buck et al. (2011), describes this classification and identifies substance types within each subclass.

Each of these PFAS classes is described in additional detail below, and illustrates the class based on example "characteristic" properties, substances, and uses. It is important to note that individual PFAS can be raw materials, compounds used in products, or environmental transformation products. In many cases, raw materials, final manufactured products, or treated articles may contain a mixture of related structures, impurities, residual raw materials, and other contaminants. Similarly, environmental transformation products may result in a mixture of compounds at the emission source based on the ambient conditions causing degradation to occur. Some of these substances are known and well-characterized, but many are unknown.

Appendix 4: Fate and Transport addresses environmental transformation of PFAS in more detail.

Table 3. PFAS classes.

Non-polymers	Polymers
Perfluoroalkyl substances Compounds for which all hydrogen atoms on all carbon atoms (except for carbon atoms associated with functional groups) have been replaced by fluorine atoms, such as: • (Aliphatic) perfluorocarbons. • Perfluoroalkyl acids. • Perfluoroalkane sulfonyl fluorides. • Perfluoroalkane sulfonamides. • Perfluoroalkyl iodides. • Perfluoroalkyl aldehydes. • Perfluoroalkyl ether acids Polyfluoroalkyl substances Compounds for which all hydrogen atoms on at least one (but not all) carbon atoms have been replaced by fluorine atoms, such as: • Perfluoroalkane sulfonamido derivatives. • Fluorotelomer-based compounds. • Semifluorinated n-alkanes and alkenes.	Side-chain fluorinated polymers Variable composition non-fluorinated polymer backbone with fluorinated side chains, such as: • Fluorinated acrylate and methacrylate polymers. • Fluorinated urethane polymers. • Fluorinated oxetane polymers. Fluoropolymers Carbon-only polymer backbone with fluorine atoms directly attached, such as: • Polytetrafluoroethylene. • Polyvinylidene fluoride. • Polyvinyl fluoride. Perfluoropolyethers Carbon and oxygen polymer backbone with fluorine atoms directly attached to carbon atoms, such as perfluoropolyethers.

1.1.3 Non-polymer PFAS

Most PFAS of interest at environmental release sites are non-polymers (ITRC, 2020b). Non-polymeric PFAS can be subdivided into two classes: perfluoroalkyl substances and polyfluoroalkyl substances.

Table 4 below provides additional classification of perfluoroalkyl substances, their chemical structures, and their uses. This table is in no way comprehensive. It focuses on those substances which have been more prevalently identified with respect to environmental presence or regulatory control (ITRC, 2020b).

Table 5 presents similar information for polyfluoroalkyl substances.

Perfluoroalkyl substances

Perfluoroalkyl substances are fully fluorinated (perfluoro-) alkane (carbon-chain) molecules. Their basic chemical structure is a chain (or tail) of two or more carbon atoms with a charged functional group "head" attached at one end. The functional groups commonly are carboxylic or sulfonic acids, but other forms have been detected, as indicated in Table 4.

PFOS, illustrated in Figure 2 above, is a perfluoroalkyl substance—where F atoms are attached to all possible bonding sites along the C chain of the tail, except for one bonding site on the last C where the functional sulfonic acid group head is attached. Perfluoroalkyl chains are often represented in a shorthand form as C_nF_{2n+1} , with n>2. As noted in Table 4, these PFAS can be present in the form of raw materials, compounds used as commercial products, or intermediate environmental degradation compounds.

As addressed in <u>Appendix 4: Fate and Transport</u>, biotic and abiotic degradation of many polyfluoroalkyl substances may result in the formation of PFAAs. PFAAs are essentially non-degradable, and are the most tested type of PFAS in the environment (ITRC, 2020b). Polyfluoroalkyl substances that degrade to PFAAs are often called "precursors." PFAAs are sometimes referred to as "terminal PFAS," because no further degradation products will form from them.

Short-chain PFAAs have been developed and are currently marketed as replacements to phased-out long-chain PFAAs such as PFOS and PFOA. These are discussed in <u>Section 1.3.4</u> below.

Polyfluoroalkyl substances

Unlike perfluoroalkyl substances, polyfluoroalkyl substances are not fully fluorinated. Instead, they have a non-fluorine atom (typically hydrogen or oxygen) attached to at least one, but not all, carbon atoms, while at least two or more of the remaining carbon atoms in the carbon chain tail are fully fluorinated (ITRC, 2020b).

Fluorotelomer substances are polyfluoroalkyl substances produced by the telomerization process. Perfluoroalkane substances have a fully fluorinated carbon chain tail, but they also contain one or more CH₂ groups in the head of the molecule attached to a sulfonamido spacer (ITRC, 2020b).

In Figure 6, the six perfluorinated carbons (n=6), could be represented as C_6F_{13} , the hydrocarbon spacer as C_2H_4 , and the end group as SO_3H . This mix of a perfluoroalkyl chain and a hydrocarbon spacer results in a polyfluorinated carbon chain. The polyfluoroalkyl structures have a numerical prefix based on these structural elements to indicate the number of perfluorinated versus non-fluorinated C atoms. Figure 6 illustrates the 6:2 fluorotelomer sulfonic acid.

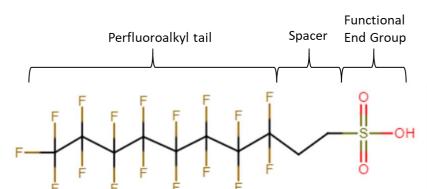


Figure 6. Schematic structure of a polyfluorinated surfactant, the 6:2 fluorotelomer sulfonic acid.

The carbon-hydrogen (or other non-fluorinated) bond in polyfluoroalkyl molecules creates a "weak" point in the carbon chain that is susceptible to biotic or abiotic degradation. As a result, many polyfluoroalkyl substances that contain a perfluoroalkyl C_nF_{2n+1} group are potential precursor compounds that have the potential to be transformed into PFAAs (ITRC, 2020b). See <u>Appendix 4: Fate and Transport</u> for additional discussion of the degradation of these substances.

Table 4. Overview of non-polymer perfluoroalkyl PFAS (Buck et al., 2011).

Class	Sub-class	Functional group C _n F _{2n+1} R, where R =	Examples	Uses
Perfluoroalkyl acids (PFAAs) ^a	Perfluoroalkyl carboxylic acids (PFCAs)	-COOH	Perfluorooctanoic acid (PFOA) Perfluorononanoic acid (PFNA) Perfluorohexanoic acid (PFHxA)	Surfactant
PFAAs ^a	Perfluoroalkyl carboxylates (PFCAs)	-COO	Sodium perfluorooctanoate (Na- PFOA) Ammonium perfluorooctanoate (APFO) Ammonium perfluorononanoate (APFN)	Surfactant
PFAAs ^a	Perfluoroalkane sulfonic acids (PFSAs)	-SO₃H	Perfluorooctane sulfonic acid (PFOS) Perfluorohexane sulfonic acid (PFHxS) Perfluorobutane sulfonic acid (PFBS)	Surfactant

Class	Sub-class	Functional group C _n F _{2n+1} R, where R =	Examples	Uses
PFAAs ^a	Perfluorooctane sulfonic acid (PFSIAs)	-SO2H	Perfluorooctane sulfonic acid (PFOSI)	Intermediate environmental transformation product
PFAAs ^a	Perfluoroalkane sulfonates (PFSAs)	-SO ₃ -	Tetraethylammonium perfluorooctane sulfonate (NEt ₄ -PFOS)	Surfactant
PFAAs ^a	Perfluoroalkyl phosphonic acids (PFPAs)	-P(=O)(OH) ₂	Perfluorooctyl phosphonic acid (C8- PFPA)	Surfactant
PFAAs ^a	Perfluoroalkyl phosphonic acids (PFPIAs)	-P(=O)(OH)	Bis(perfluorohexyl) phosphonic acid (C6/C6-PFPIA)	Surfactant
PFAAs ^c	Perfluoroalkyl ether carboxylic acids (PFEAs) or Perfluoroalkyl ether sulfonic acids (PFECA/PFESA)	Various Example: - (C _m F _{2m})COOH	Hexafluoropropylene oxide (HFPO) dimer acid "GenX"	Polymer processing aid
Perfluoroalkane sulfonyl fluorides (PASFs) ^b		-SO₂F	Perfluorooctane sulfonyl fluoride (POSF) Perfluorobutane sulfonyl fluoride (PBSF)	Raw material for surfactant and surface protection products
Perfluoroalkane sulfonamides (FASAs) ^b		-SO₂NH₂	Perfluorooctane sulfonamide (FOSA)	Raw material for surfactant and surface protection products
Perfluoroalkanoyl fluorides (PAFs) ^b		-COF	Perfluorooctanoyl fluoride (POF)	Raw material for PFOA made by the ECF process; raw material for surfactant and surface protection products
Perfluoroalkyl iodides (PFAIs) ^c (Telomer A)		-1	Perfluorohexyl iodide (PFHxI) Perfluorooctyl iodide (PFOI)	Raw material for surfactant and surface protection products

Class	Sub-class	Functional group C _n F _{2n+1} R, where R =	Examples	Uses
Perfluoroalkyl aldehydes and aldehyde hydrates (PFALs) ^c		-CHO and - CH(OH)2	Perfluorononanal (PFNAL)	Intermediate environmental transformation product

Notes:

- a: Substances originating by either electrochemical fluorination (ECF) or fluorotelomer processes.
- b: Substances originating by the ECF process.
- c: Substances originating by the fluorotelomer process.

Table 5. Overview of non-polymer polyfluoroalkyl PFAS (Buck et al., 2011).

Class	Sub-class	Functional group C _n F _{2n+1} R, where R =	Examples	Uses
Perfluoroalkane sulfonamido substances ^a	Perfluoroalkane sulfonamidoethanols (FASEs, MeASEs, EtFASEs) and N-alkylperfluoroalkane sulfonamidoethanols (BuFASEs)	- $SO_2N(R')CH_2$ CH_2OH where $R' = C_mH_{2m+1}$ (m = 1,2,4)	N-Ethyl perfluorobutane sulfonamidoethanol (EtFBSE) Perfluorooctane sulfonamidoethanol (FOSE)	Raw material for surfactant and surface protection products
Perfluoroalkane sulfonamido substances ^a	N-Alkyl perfluoroalkane sulfonamidoethyl acrylates and methacrylates (MeFAS(M)ACs, EtFAS(M)ACs, BuFAS(M)ACs)	- $SO_2N(R')CH_2$ CH_2OC - $(O)CH=CH_2$ and $SO_2N(R')CH_2$ CH_2OC - $(O)C(CH_3)$ = CH_2 where $R' = C_mH_{2m+1}$ (m=1,2,4)	N-Ethyl perfluorooctane sulfonamidoethyl acrylate (EtFOSAC)	Raw material for surfactant and surface protection products
Perfluoroalkane sulfonamido substances ^a	Perfluoroalkane sulfonamidoacetic acids (FASAAs) and N- alkyl perfluoroalkane sulfonamidoacetic acids (MeFASAAs, EtFASAAs, BuFASAAs)	SO2N(R0)CH 2COOH where R0 ¼CmH2mþ1 (m¼0,1,2,4)	N-Ethyl perfluorooctane sulfonamidoacetic acid (EtFOSAA),	Intermediate environmental transformation product

Class	Sub-class	Functional group C _n F _{2n+1} R, where R =	Examples	Uses
Fluorotelomer substances ^b	Semifluorinated n- alkanes (SFAs) and alkenes (SFAenes)	-(CH2) _m H and - CH=CH(CH2) _{m-2H} , with m = 2–16 and n = 6–16	(Perfluorooctyl)ethane (F ₈ H ₂)	Ski wax, medical applications
Fluorotelomer substances ^b	n:2 Fluorotelomer iodides (n:2 FTIs)	-CH ₂ CH ₂ I	8:2 Fluorotelomer iodide (8:2 FTI)	Raw material for surfactant and surface protection products
Fluorotelomer substances ^b	n:2 Fluorotelomer olefins (n:2 FTOs)	-CH=CH ₂	6:2 Fluorotelomer olefin (6:2 FTO)	Raw material for surfactant and surface protection products
Fluorotelomer substances ^b	n:2 Fluorotelomer alcohols (n:2 FTOHs)	-CH₂CH₂OH	4:2 Fluorotelomer alcohol (4:2 FTOH)	Raw material for surfactant and surface protection products
Fluorotelomer substances ^b	n:2 Unsaturated fluorotelomer alcohols (n:2 FTUOHs)	- CF¼CHCH2O H	8:2 Unsaturated fluorotelomer alcohol	Intermediate environmental transformation product
Fluorotelomer substances	n:2 Fluorotelomer acrylates (n:2 FTACs) and methacrylates (n:2 FTMACs)	- H ₂ CH ₂ OC(O) CH=CH ₂ and - CH ₂ CH ₂ OC(O)C(CH ₃)=CH ₂	6:2 Fluorotelomer acrylate (6:2 FTAC) 6:2 Fluorotelomer methacrylate (6:2 FTMAC)	Raw material for surfactant and surface protection products
Fluorotelomer substances ^b	n:2 Polyfluoroalkyl phosphoric acid esters, polyfluoroalkyl phosphates, fluorotelomer phosphates (PAPs)	(-CH ₂ CH ₂ O) _x - P(=O)(OH) _{3-x} where x = 1 or 2	10:2 Fluorotelomer phosphate monoester (10:2 monoPAP)	Surfactant and surface protection products
Fluorotelomer substances ^b	n:2 Fluorotelomer aldehydes (n:2 FTALs) and unsaturated aldehydes (2 FTUALs)	-CH2CHO and - CF¼CHCHO	8:2 Fluorotelomer aldehyde (8:2 FTAL) 8:2 Fluorotelomer unsaturated aldehyde (8:2 FTUAL)	Intermediate environmental transformation product

Class	Sub-class	Functional group C _n F _{2n+1} R, where R =	Examples	Uses
Fluorotelomer substances ^b	n:2 Fluorotelomer carboxylic acids (n:2 FTCAs) and unsaturated carboxylic acids (n:2 FTUCAs)	-CH2COOH and - CF¼CHCOO H	8:2 Fluorotelomer carboxylic acid (8:2 FTCA) 8:2 Fluorotelomer unsaturated carboxylic acid (8:2 FTUCA),	Intermediate environmental transformation product
Fluorotelomer substances ^b	n:3 Saturated acids (n:3 Acids) and n:3 Unsaturated acids (n:3 UAcids)	- CH2CH2CO OH and - CH¼CHCOO H	7:3 Acid, 7:3 UAcid,	Intermediate environmental transformation product
Fluorotelomer substances	n:2 Fluorotelomer sulfonic acids (n:2 FTSAs)	- CH₂CH₂SO₃H	6:2 Fluorotelomer sulfonic acid (6:2 FTSA)	Surfactant and environmental transformation products
Fluorotelomer substances ^b	n:2 Fluorotelomer sulfonic acid chloride (n:2 FTSAs)	- CH ₂ CH ₂ SO ₂ C I	6:2 Fluorotelomer sulfonyl chloride	Surfactant and environmental transformation products
Miscellaneous	Polyfluoroalkyl ether carboxylic acids & others Perfluoropolyethers	e.g., - O(C _m F _{2m})- OCHF(C _p F _{2p}) COOH e.g., - O(C _m F _{2m} O-) _n CF ₃	4,8-Dioxa-3H- perfluorononanoic acid (parent of the structure in Fig. 23, left)	Perfluoro- polyether oils and lubricants Alternative fluoropolymer processing aid (as ammonium salt)

Notes:

- a: Substances originating by electrochemical fluorination (ECF) process.
- b: Substances originating by fluorotelomer process.

Table 6. Overview of polymeric PFAS (Buck et al., 2011).

Class	Sub-class	Examples	Uses
Fluoropolymers: Carbon-only polymer backbone		-(CF2CF2)n- Polytetrafluoroethylene (PTFE) -(CH2CF2)n- Polyvinylidene fluoride	Plastics
with F directly attached		(PVDF)	
to backbone C atoms		-(CH2CHF)n- Polyvinyl fluoride (PVF)	
		-(CF2CF2)n-(CF(CF3)CF2)m-	
		Fluorinated ethylene	
		propylene (FEP)	
Perfluoropolyethers:		Examples:	Functional fluids,
Ether polymer		F-(CmF2mO-)nCF3	surfactants, and
backbone with F atoms		HOCH2O-[CmF2mO-]nCH2OH	surface
directly attached		-where CmF2mO represents -CF2O-, -	protection
(PFPEs)		CF2CF2O-,	products
		and/or -CF(CF3)CF2O- units	
		distributed randomly	
Side-chain–fluorinated	Fluorinated	along the polymer backbone Acrylate:	Surfactants and
polymers:	acrylate and	Backbone-CH-C(O)O-X-CnF2nb1	surface
Nonfluorinated	methacrylate	Methacrylate:	protection
polymer backbone	polymers	Backbone-C(CH3)-C(O)O-X-CnF2nb1	products
with fluorinated	po.,	-where X is -CH2CH2N(R0)SO2-	p. caaca
side chains, ending		with R0 ¼-CnH2nþ1 (n¼0,1,2,4)	
in -CnF2nþ1		or	
·		-CH2CH2-	
Side-chain-fluorinated	Fluorinated	Backbone-NHC(O)O- X-CnF2nþ1	Surfactants and
polymers	urethane	-where X is either -CH2CH2N(R0)SO2-	surface
	polymers	with R0 ¼-CnH2nþ1 (n¼0,1,2,4)	protection
		or	products
	5 1	-CH2CH2-	
Side-chain–fluorinated	Fluorinated	Backbone-CH2OCH2-R	
polymers	oxetane	-where R = -CF3, -C2F5 or -CH2C4F9	
	polymers		

1.1.4 Polymeric PFAS

Polymers are large molecules formed by combining many identical smaller molecules (monomers) in a repeating pattern (ITRC, 2020b). Polymeric substances in the PFAS family include fluoropolymers, polymeric perfluoropolyethers, and side-chain fluorinated polymers. Table 6 provides an overview of polymeric PFAS, their chemical structures, and their uses.

In general, polymeric PFAS are currently believed to pose less immediate human health and ecological risk relative to some non-polymer PFAS (ITRC, 2020b). However, some polymeric PFAS incorporate one or more PFAS monomer(s) during their synthesis. Any degradation of

these polymers, during or after their useful lifetime, may lead to release of PFAS to the environment (Buck et al. 2011).

Fluoropolymers

Fluoropolymers contain F bound to one or both of the olefinic C atoms, to form a perfluorinated C-only polymer backbone with F atoms directly attached to it (Buck et al., 2011).

Fluoropolymers have been found to have thermal, chemical, photochemical, hydrolytic, oxidative, and biological stability (Henry et al., 2018; Korzeniowski & Buck, 2019a). They are almost insoluble in water and not subject to long-range transport. With very high molecular weight (greater than 100,000 Da), fluoropolymers cannot cross the cell membrane. They are neither bioavailable nor bioaccumulative. Clinical studies of their use in medical devices has demonstrated lack of chronic toxicity or carcinogenicity and no reproductive, developmental, or endocrine toxicity.

Fluoropolymers can only be destroyed or degraded to HF and CO₂ under municipal waste incineration conditions. The manufacture of some fluoropolymers may require use of PFAS monomers as a processing aid, added in very small levels, and traditionally composed of PFOA or PFNA. Although the manufacturing process intends to remove the fluorosurfactants by drying or high cure temperatures, residual surfactants may remain on the product (Guo et al., 2009). Most U.S. manufacturers have discontinued the use pf PFOA (see <u>Appendix 9:</u> <u>Regulations, Section 9.2.1 Environmental Protection Agency</u>) and PFNA salts.

Pefluoropolyethers

Perfluoropolyethers (PFPEs) are polymers in which the backbone -CF2-, -CF2CF2-, and possibly -CF(CF3)CF2- units are separated by O atoms (Buck et al., 2011). Because the repeating units of these PFPEs contain only two or three perfluorinated C atoms per O atom, their degradation cannot lead to the formation of long-chain PFCAs.

Perfluoropolyether polymers have thermal, chemical, photochemical, hydrolytic, oxidative, and biological stability. They are practically insoluble in water and hydrocarbons, and not subject to long-range transport (Korzeniowski & Buck, 2019a).

Side-chain fluorinated polymers

Unlike fluoropolymers and perfluoropolyethers, side-chain fluorinated polymers do not have perfluorinated or polyfluorinated polymer backbones, but are composed of variable composition backbones with polyfluoroalkyl (and possibly perfluoroalkyl) side chains (Buck et al., 2011). In particular, three groups of side-chain fluorinated polymers (acrylate or methacrylate, urethane, and oxetane) may be able to sever from the polymer chain to form PFAS shown in Tables 2a and 2b.

Buck et al. (2011) notes that this transformation process can occur over periods greater than 1,000 years and may result in small amounts of PFAS—meaning a small overall contribution of

long-chain PFAS to the environment relative to other sources. However, other studies have shown degradation of these polymers in shorter time frames (Rankin et al., 2014; Washington & Jenkins, 2015; Washington et al., 2015). This topic is discussed further in Appendix 4: Fate and Transport.

1.2 Select physical and chemical properties of PFAS

Physical and chemical properties of PFAS have been extensively described in scientific literature (for example, but not limited to, Buck et al., 2011). PFAS have some unique and valuable properties when compared with non-fluorinated hydrocarbon chemicals of similar structure (Krafft & Riess, 2015). The purpose of this section is to identify significant PFAS characteristics relevant to their commercial use and significant characteristics impacting how they may enter the environment. Appendix 4: Fate and Transport addresses specific PFAS degradation pathways in detail.

1.2.1 Resistance to extreme environments

Fluorine forms an extraordinarily strong bond with carbon, and when fluorine completely replaces hydrogen in an alkyl chain of carbons, the resulting substance is much more resistant to thermal or chemical attack than a similar fluorine-free hydrocarbon. As a result, PFAS are often preferred for use in extreme environments (high temperatures, strongly reactive conditions, etc.). These same characteristics are responsible for the extreme environmental persistence of perfluorinated substances—they are completely resistant to naturally occurring breakdown mechanisms. See more on this in <u>Appendix 4: Fate and Transport</u>.

1.2.2 Surfactants and emulsifiers

PFAS treatments or polymer coatings are often used to create low surface energy materials, preventing the spread of water or oils on their surface. Fluoropolymers, such as polytetrafluoroethylene (PTFE), are un-wettable in that both oil and water will "bead-up" on PTFE surfaces. Common applications include thin fluoropolymer linings in hydraulic tubing, linings for chemical and pharmaceutical processing equipment, and breathable membranes for garments. Side-chain polymers or perfluoropolyethers derived from PFAS can be used to coat surfaces on a molecular scale, imparting oil and water (i.e., stain) resistance at the individual fiber level in textiles, fabrics, or carpets.

Other PFAS are added to liquid formulations and function mostly as surface-active agents (surfactants) or emulsifiers. Surfactants are commonly used to affect wetting and spreading of liquids (Knepper & Lange, 2012). When a surfactant is added to water, the normally high surface tension is reduced, and droplets behave more like oil droplets, spreading on the polyethylene surface. Fluorinated surfactants are effective at reducing surface tension in both oil- and water-based products to promote wetting and spreading. These properties are

important in many applications, for example paints which must cover surfaces uniformly and completely, or inks which need to achieve full coverage on printing plates.

When surfactant properties are combined with a need for chemical inertness or resistance to high temperature, PFAS can have distinct advantages over traditional hydrocarbon surfactants or materials (Krafft & Riess, 2015).

1.2.3 Modifications for PFAS chemical function

Formulating a product from a mixture of chemical ingredients and solvents is complex. A surfactant may play multiple roles and needs to meet other functional requirements (color, temperature, stability, etc.). In a floor polish, the surfactant improves wetting and spreading, but also helps achieve a smooth, glossy finish through its effect on surface tension as the polish dries. Several surfactants may be used in a single product, with hydrocarbon surfactants used to keep ingredients dispersed and fluorinated surfactants used to promote wetting. The individual constituents must work well together in the complete system of ingredients for the product to function as intended. PFAS products are therefore carefully designed to achieve multiple characteristics upon their intended use.

Hydrocarbon surfactants are often described as having a head and a tail. The tail is often a long alkyl chain and relatively insoluble in water (hydrophobic). In contrast to the tail, the head is typically more compact, and often hydrophilic, or water-loving. Most surfactants for water-based applications orient at the surface of the liquid, with the tail portion extending out and over the surface at the molecular level and the head-only immersed in liquid. The head is equivalent to the R-group described in Section 1.1.1 above.

As described in Section 1.1.1 above, many fluorosurfactants have a similar design, but the fluorocarbon tail is insoluble in both oil and water (both oleophobic and hydrophobic). Most often, the tail is a perfluorinated carbon chain. The head varies more widely and is chosen so that surfactants will perform certain functions in each product application. For example, a fluorinated surfactant for a water-based paint application usually has an R-group that is hydrophilic (water-loving). Sulfonic acid or carboxylic acid R-groups work well in these applications, so both PFOS and PFOA were used for water-based applications.

In some applications, heteroatoms, like oxygen (O), may be introduced into the fluorinated tail. The resulting perfluoroalkyl ether surfactants are currently used as processing aids in emulsion polymerization, where they replace legacy processing aids like ammonium perfluorooctanoate (APFO), the ammonium salt of PFOA. One example is the ammonium salt of perfluoro-2-propoxypropanoic acid (PFPrOPrA), also called hexafluoropropylene oxide dimer acid (HPFO-DA) and known by the trade name used for this process, called GenX. Perfluoroalkyl ether carboxylic acids (PFECAs) and perfluoroalkyl ether sulfonic acids (PFESAs) contain O-atoms interspersed among (typically) short perfluorinated chains (Sun et al., 2016). Figure 7 provides an illustration of the structure of PFPrOPrA.

Figure 7. The ammonium salt of PFPrOPrA/HPFO-DA (also known as GenX).

$$F = F = F$$

$$F = F$$

$$F = F$$

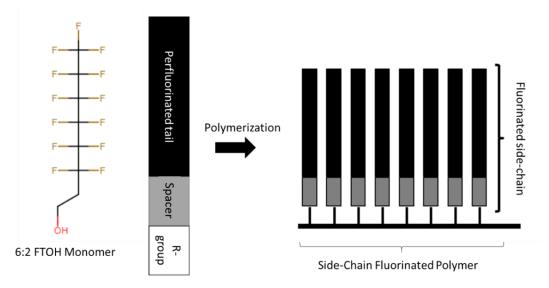
$$F = F$$

The basic head and tail concept is a bit different in polyfluorinated surfactant design. As illustrated in Figure 8, using 6:2 FTOH as an example, manufacturers have introduced a hydrocarbon spacer (grey portion in center) between the perfluorinated tail (black portion, left) and the head-group (white portion, right). The hydrocarbon "spacer," often a two-carbon group, extends the combined surfactant tail length. Some reports suggest that the use of a spacer helps to balance function and toxicity as manufacturers have moved to shorter perfluorinated chains (Renner, 2006).

Very similar fluorinated monomer structures as those used in surfactants are used in the production of polymeric surface treatment or impregnation products for textiles and paper. R-groups such as acrylate or methacrylate form fluoroalkyl acrylate and methacrylate monomers. These may be combined with non-fluorinated monomers. The monomers are polymerized to form a non-fluorinated hydrocarbon backbone with fluorinated side-chains, like teeth on a comb. These are commonly called "side-chain polymers." Side-chain polymers are often sold as aqueous dispersions and used for surface treatment or impregnation of textiles, carpets, and paper products, among other uses. Side-chain polymers are not themselves considered surfactants.

Figure 8 provides an illustration of a side-chain polymer. In the schematic (right), the solid line at the base represents the main, non-fluorinated polymer backbone. Fluorinated side-chains (black bars) are bonded to the backbone through reactions with the hydrocarbon spacer group (gray bar). The treated surface is at the bottom of the figure with the air interface at the top. The structure on the right is a typical example of one "tooth" of the comb.

Figure 8. Fluorinated side-chain polymer, typical of stain-resistant surface treatments for textiles.



Another example of substance tailoring is that of perfluoropolyether (PFPE) substances, which include repeating structural ether units, as illustrated in Figure 9 by the bracketed structures with subscripts. Depending on the number of repeating ether-units, these will vary in molecular weight and in their physicochemical properties. PFPE includes different length ether units that repeat ("n" or "m" times) and variable R groups that can be tailored by application requirements (Solvay Company, 2015). One manufacturer reports that "n" can vary from 10 to 60 (Krytox, 2020). PFPEs are used as surfactants, functional fluids, and to modify properties of other polymers such as polyurethane.

Figure 9. Possible chemical structure of perfluoropolyether (PFPE).

$$F = F$$

$$F =$$

The expertise to fine-tune these surfactant, side-chain fluoroalkyl polymer, and perfluoro-polyether structures is highly valued intellectual property and may be one reason why the details of these structures are often not publicly disclosed. The first chapter in Knepper and Lange (2012) contains many examples of fluorinated chemicals, their associated applications, and relevant literature citations.

1.2.4 Solubility in water

PFAS can have varying solubility in water (Ross & Hurst, 2019). Pancras et al. (2016) compiled solubility data for a variety of PFAS. PFCAs (PFBA, PFPeA, PFHxA, PFHpA, and PFOA) and PFSAs (PFBS, PFPeS, PFHxS, PFHpS, and PFOS), in general, have high solubility, with decreasing solubility as chain length increases. This is one reason why these PFAS have been transported throughout the environment. On the other hand, fluorotelomer alcohols (FTOHs) are, in general, more hydrophobic than PFAAs, and also have decreasing solubility as chain length increases.

Solubility of PFAS is further affected by the chemical composition of the water medium where they are located. The environment determines the protonation state of PFAS, which in turn affects physical and chemical properties, including solubility. For example, PFAAs are anionic, dissociating in water under most environmentally relevant pHs to form a negatively charged version of the acid along with a dissociated proton. However, under conditions of very low pHs, PFAAs will not dissociate (Johansson, 2017), which changes their properties, such as greatly decreasing their solubility.

In this report, we will most often be discussing anionic PFAS under environmental conditions, since they are the chemicals most often studied and used. However, some PFAS are cationic, zwitterionic, or non-ionic, which can lead to different behavior. As described in Appendix 4: Fate and Transport, for example, cationic PFAS are much more likely to associate with soils and sediment (ITRC, 2020c).

1.3 Manufacturing

Complex chemicals like PFAS generally require several sequential manufacturing steps and utilize multiple chemical raw materials, catalysts, and other additives too numerous to detail here. However, the principle perfluoroalkyl building blocks used for making fluorosurfactants and side-chain fluorinated polymers are manufactured using two main processes: electrochemical fluorination (ECF) and telomerization (Buck et al., 2011; Keppner & Lange, 2012).

As addressed in <u>Appendix 3: Sources and Uses, Section 3.1.1 Primary Manufacturing</u>, PFAS were not, nor are they currently, manufactured in Washington state.

1.3.1 Electrochemical fluorination (ECF)

When a hydrocarbon raw material is combined with hydrofluoric acid (HF), application of a strong electric current can break the H – F bond and create reactive fluoride species. These reactive species replace the hydrogens in the hydrocarbon one-by-one with fluorine resulting in a perfluorinated molecule. ECF produces odd and even numbered chains as well as branched and linear mixtures. ECF was the dominant global method of production (principally by the 3M Company) for both PFOS and PFOA from the late 1940s until their U.S. phase-out beginning

around the year 2000, and subsequent 2006 – 2015 stewardship program (De Voogt, 2010; EPA, 2000, 2018).

ECF related production of short-chains PFAS products became available in the 2000s, and ECF production of long-chains was started in Asian countries such as China to fill the void left by the major global manufacturers who exited production (ITRC, 2020a). ECF is still used in both the U.S. and abroad, especially in China, India, and Russia (OECD, 2015).

Perfluorooctane sulfonyl fluoride (POSF) from the ECF process was the basic building block for a wide variety of surfactant and polymer products, including PFOS. Figure 10 illustrates the reaction that produces POSF through ECF.

Figure 10. A schematic of the ECF reaction that forms POSF.

1.3.2 Telomerization

Following the phase-out of PFOS and PFOA production by ECF, telomerization has become a dominant process for producing perfluorinated alkyl chain raw materials. Telomerization is a polymerization reaction that results in products with even-numbered carbon chain lengths and a terminal iodide (I) functional group. PFOA can be subsequently made by oxidizing PFI with sulfur trioxide. Insertion of the hydrocarbon ethylene instead of fluorocarbon reactants converts a perfluorinated molecule to a linear polyfluorinated alkyl chain, such as the 8:2 fluorotelomer iodide (8:2 FTI). Figures 11 and 12 respectively illustrate each of these reactions.

Figure 11. Schematic of telogen (perfluoroethyl iodide) reacting with three taxogen units (tetrafluoroethene) to form a perfluorinated product, perfluorooctyl iodide (PFI).

$$F \stackrel{F}{\longleftarrow} F + 3 \stackrel{F}{\longleftarrow} F \stackrel{F}{\longrightarrow} F \stackrel{F}{\longrightarrow}$$

Figure 12. Schematic of PFI further reacting with the hydrocarbon ethene to form the polyfluorinated 8:2 fluorotelomer iodide (FTI).

FTI can be converted to alcohols (FTOHs) and further functionalized for use as fluorotelomer surfactants. Figure 13 provides the example of an 8:2 fluorotelomer iodide shown on the left, and its alcohol counterpart shown on the right. In the case of this fluorotelomer, the "8" refers

to the eight perfluorinated carbons, and the "2" refers to the two hydrogenated carbons (Hs not shown) adjacent to the end group. A significant share of the fluorotelomer market is for side-chain fluorinated polymers (Grand View Research Inc., 2020) such as the fluorotelomer acrylates (FTACs), which are made from FTOH monomers.

Because FTOH have been manufactured more recently, their presence in environmental media can be an indication of more recent contamination sources.

Figure 13. An 8:2 fluorotelomer iodide (left) can be converted to an 8:2 fluorotelomer alcohol (right).

1.3.3 Other processes

As identified in Tables 4 and 5, the per- and polyfluoroalkyl substances described in the previous sections can be used as raw materials or intermediates for commercial products. Some of the main manufacturing processes used to modify these intermediates, such as the addition of functional groups, are well described in Knepper and Lange (2012).

Since the 1970s, several manufacturers have developed independent production paths to produce the many per- and polyfluorinated ether surfactants and perfluoropolyether products available on the market today (Dams & Hinzter, 2016; Knepper & Lange, 2012). Literature has only recently begun to identify and assess these substances (Sun et al., 2016; Wang et al., 2013).

1.3.4 Technical quality and implications for environmental impacts

While discrete substances, like PFOS or PFOA, are the focus of the discussion of environmental impacts, the ECF and telomerization processes produce a complex mixture of substances rather than pure one-component products. For example, the harsh conditions of the ECF process lead to a variety of unwanted side-reactions. The resulting product mixture may contain both linear and branched chains with both odd and even chain lengths. ECF production targeting PFOA (C8) includes 70-80% linear substances (of differing carbon chain lengths) with 20-30% branched substances, including even cyclic compounds (De Voogt, 2010).

While ECF mixtures randomly vary, they are sufficiently consistent for forensic application. PFAS environmental contaminants collected in China matched the chain-length profile expected for ECF products, suggesting that nearby manufacturing facilities employ the ECF process (Jiang, 2015). Figure 14 illustrates the isomer composition of two ECF products (adapted from Jiang, 2015). As one would expect, the majority of these ECF products are "normal" or straight-chain isomers, but may contain 20 – 30% of various branched isomers. The top bar represents

Chinese ECF production (Defu PFOSK, China). The bottom bar is typical of a 2000-era 3M PFOS. The similarity of the composition confirms that both were manufactured using ECF.

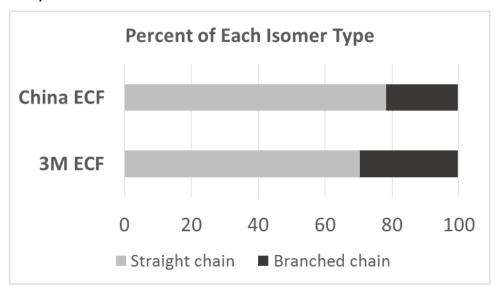


Figure 14. Comparison of two-carbon tetrafluoroethene taxogens manufactured using ECF (Jiang et al., 2015).

The telomerization process also produces a mixture of substances, typically a series of straight chains varying in length by even numbers. For example, production targeting the 6:2 FTI may include minor quantities of 8:2 and 4:2 chain lengths. The two-carbon tetrafluoroethene taxogen (shown in Figure 14) adds about 100 grams per mole (g/mol) in each addition step. The change in properties between the C_n to C_{n+2} homologue allows for purification by distillation (Krafft & Riess, 2015). The extent to which manufacturers purify their products or otherwise control for by-product content is not well understood.

Products have been marketed as mixtures of PFAS isomers or homologues (KEMI, 2015). For example, Surflon® S-111, a now-discontinued surfactant produced by telomerization, contained primarily 9-C perfluorononanoic acid (PFNA), but also significant quantities of 11-C perfluoroundecanoic acid (PFUnDA) (20%) and 13-C perfluoro-tridecanoic acid (PFTrDA) (5%) (Prevedouros et al., 2006). Chemical analysis of "articles of commerce" shows that many formulated products have been composed of complex PFAS mixtures (Figure 15) (Liu et al., 2012).

Products may also be contaminated with residual raw materials, polymerization aids, and unintended by-products. PFOA, higher molecular weight homologues, and PFOA precursors have all been found in fluorotelomer and fluoropolymer products. Similarly, FTOHs and fluorotelomer olefins (FTOs) have been identified in fluorotelomer acrylate and methacrylate products (Lassen et al., 2013).

Figure 15 provides an illustrative example of the various PFAS making up a single commercial product—in this case, a carpet or upholstery protector concentrate. As indicated in the figure,

the sample contains a wide distribution of chain lengths. Many other commercial PFAS-based products may also be composed of multiple PFAS. This shows that PFAS manufacturing and the use of PFAS in products can lead to emission of a multitude of PFAS. As further explained in Appendix 4: Fate and Transport, environmental transformation of manufactured PFAS may lead to an even larger variety of contaminants. Appendix 2: Analytical Methods further addresses the fact that analytical methods approved by EPA and other agencies are not able to detect all PFAS present in a sample, but only those targeted by the analytical method.

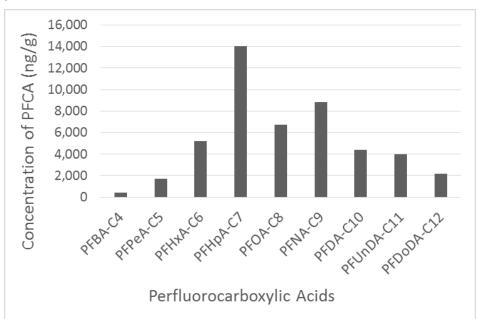


Figure 15. Analytical chemistry data for the PFCA content (C4 – C12) of a U.S. carpet or upholstery protector concentrate (Liu et al., 2012).

1.3.5 Trends in per- and polyfluorinated substance design

PFOS and PFOA, both which are characterized as long-chains, dominate the literature on PFAS due to their well-established PBT properties. These substances are associated with workhorse technologies of the first decades of PFAS development and use. PFOS is both a directly manufactured product and a highly stable degradation product of many legacy POSF-based surfactants. PFOS can also occur as an impurity in derivative products. PFOA was widely used as a polymerization aid in fluoropolymer manufacture. PFOA emissions have historically been linked to releases from these manufacturing operations (Prevedouros et al., 2006), but also occur as breakdown products of PFOA-precursors like the fluorotelomer alcohols. Production of PFOS- and PFOA-associated chemistries has continued in China, India, and Russia. Figure 16 illustrates historical estimated emissions based on manufacturing location. Based on these estimates, production-related PFCA emissions were expected to be substantively eliminated in Japan, Western Europe, and the U.S. by 2002, but have continued in China, India, and Russia (ITRC, 2020a). Articles treated with long-chain PFAS are still imported from these countries to the U.S.

Due to regulatory restrictions and voluntary withdrawal campaigns regarding long-chain PFAS (see <u>Appendix 9: Regulations, Section 9.2.1 Environmental Protection Agency</u>), manufacturers in the U.S., Western Europe, and Japan have shifted manufacture primarily to replacement shorter-chain PFAS. Shorter-chain alternatives include (OECD, 2013):

- Perfluorobutane sulfonyl fluoride (PBSF)-based derivatives.
- Shorter-chain (i.e., 6:2) fluorotelomer-based chemicals.
- Mono- and polyfluorinated-ether compounds.
- Fluorinated oxetanes.
- Other fluorinated polymers.

Both legacy products (which are responsible for much of current-day emissions) and newer chemistries of so-called alternative or replacement products are included in the discussion of commercially used products in Section 1.4 below. It is important to remember that PFOA may be present as a manufacturing impurity in shorter-chain products made by telomerization. If non-target isomers and homologues are not removed by further processing, they will end up in the final product formulation or treated articles.

Information is lacking regarding the effects and fate of short-chain PFAS in general, in the environment, and their toxicological profiles. Ateia et al. (2019) reviewed the information available regarding short-chain PFAS and identified the following challenges in characterizing and quantifying their long-term effects once released in the environment: larger quantities are being used to attain similar performance to long-chain PFAS; these substances can persist in the environment; few of them have been identified because they remain proprietary; and their release may continue indefinitely into the future.

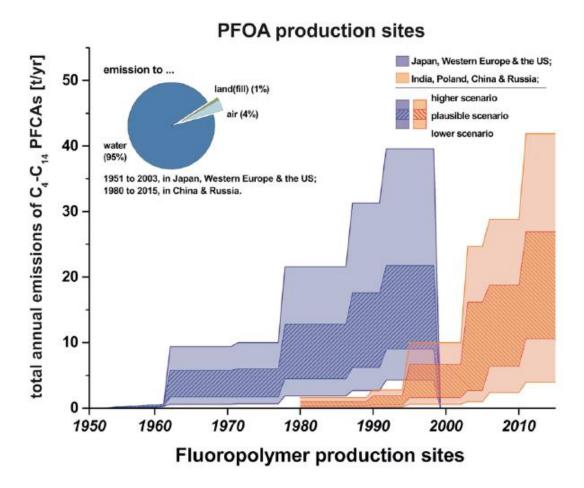


Figure 16. Manufacturing emissions estimates from the OECD (Wang et al., 2014).

1.4 Characteristic product uses of PFAS

Poulsen (2005) discussed legacy product designs in detail. In addition to legacy products, current-use products have also been addressed by OECD (2013), Buck et al. (2011), and Knepper and Lange (2012). <u>Appendix 3: Sources and Uses</u> discusses commercial PFAS uses in detail.

This section focuses on better-known product types and substances more commonly discussed in the environmental literature, as well as their relationships to specific PFAS chemistries and characteristics. While fluoropolymers dominate the market for fluorinated materials, this section will limit the polymer discussion primarily to side-chain polymers and perfluoropolyethers used as surface treatments.

Example substances for both legacy and current-use PFAS in some selected use categories are presented in Table 7. While the term "legacy" suggests an old or outdated use, the terminology is used more loosely here because:

- Some of the identified legacy substances may still be manufactured in foreign markets and imported to the U.S. (as discussed above in <u>Section 1.3.4</u>).
- Some legacy substances were recently withdrawn from the U.S. market and may still be in use or stockpiled, such as long-chain PFAS in treated carpets or firefighting foams.
- Some otherwise widely banned substances have permitted (exempt) uses, such as long-chain PFAS in mist suppressants for chrome-plating operations. This use of PFOS has been phased out by industry in the U.S. (NASF, 2019).

Table 7. Typical examples of legacy and current-use products for selected use categories (Danish Environmental Protection Agency (DEPA), 2015; United Nations Environmental Programme (UNEP), 2013).

See Tables 2. 3	3. an	d 4 for	definitions	of	abbreviations.
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Use category	Example legacy products	Example current-use product
Carpet, textile,	PFOS, N-ethyl perfluorooctane	<c6 and="" fti="" ftoh-="" pbsf-based<="" td=""></c6>
leather, stone and	sulfonamidoethanol (EtFOSE)-based	acrylate, methacrylate and urethane
tile, paints and	acrylate, FTOH-based acrylate,	side-chain polymers
coating additives	methacrylate and urethane side-chain	
and treatments	polymers	
Paper and	EtFFOSE phosphate esters, N-methyl	Perfluoropolyethers, <c6 side-chain<="" td=""></c6>
packaging	perfluorooctane sulfonamido-ethanol	fluorinated polymers
treatment	(MeFOSE) acrylate polymers	
Specialty chemicals	Potassium salt of glycine, N-ethyl-N-	FTOH- and PBSF-based surfactants,
used in oil	[(heptadecafluorooctyl)sulfonyl] (PFOS-	perfluoropolyethers
production	based surfactant)	
Fire-fighting	Perfluoroalkyl sulfonamido amine	6:2 FTAB (FTalkyl iodide-based
chemicals	derivatives and other PFOS-based	surfactant) and 6:2 thiol derivatives
	products	(6:2-SH)
Polymer processing	PFOA, PFNA	Ammonium salts of PFOA
aids		
Metal plating	PFOS	6:2-Fluorotelomer sulfonate (6:2
Wictar plating	1103	FTS)
		113)

1.4.1 Carpet and textile surface treatment

Surface treatment for carpets, upholstery, leather, apparel, and other textiles are the largest market for fluorinated side-chain polymers. Carpeting and upholstery involve large treated areas and stain-resistance treatment is a frequent specification among institutional purchasers (DTSC, 2017).

POSF is a manufacturing precursor for the perfluoroalkane sulfonamido alcohols. These alcohols are converted to acrylates and methacrylates used as monomers in the production of polymeric surface protection products. Acrylates of N-methyl or N-ethyl perfluorooctane

sulfonamido ethanol (MeFOSE and EtFOSE) and related precursors have been phased-out among U.S., Western Europe, and Japanese manufacturers over the last decade. A single MeFOSE-derived side-chain "tooth" is shown in Figure 17. These products were no longer produced in the U.S. after the early 2000s.

Figure 17. Legacy carpet treatment chemistry.

Similarly, fluorotelomer alcohols and ethyl iodides are the basis for acrylate, methacrylate, or urethane substances that are polymerized to form fluorinated side-chain polymers as illustrated previously in Figure 8. Early versions of these telomer-based products contained broad ranges of chain lengths (e.g., recall Figure 15 analytical results) (Dinglasan-Panlilio & Mabury, 2006). Fluorotelomer products (4:2 or 6:2) have replaced the longer chain legacy products in the U.S., Western Europe, Japan, and elsewhere globally.

1.4.2 Paper and packaging treatment

Surface treatment and impregnation products provide water, oil, and grease resistance and non-stick performance for paper and packaging. These include both food-contact materials, like popcorn bags, pizza boxes, and fast-food wrappers, and non-food applications, such as masking papers and folding cartons.

Legacy products include variants of perfluorooctane sulfonamido alcohols (like EtFOSE) in perfluoroalkyl phosphoric acid products, also called SAmPAPs (D'eon et al., 2009; Geueke, 2016). As an example, Figure 18 provides a schematic of a phosphate di-ester formed from EtFOSE. These and related mono- and tri-esters are also called SAmPAPs and are among the

first perfluorinated substances widely commercialized for food packaging. Prior to their removal from commerce in the U.S., SAmPAPs were reportedly the largest source of PFOS precursors in the commercial market (Benskin et al., 2012).

The MeFOSE-based acrylate polymers similar to those used in textiles were also used for paper protection. PFOS-based and other long-chain chemistries are still used for food-contact materials in Thailand and China (Benskin et al., 2012; Geueke, 2016; Yuan et al., 2016). A very recent review from the Nordic Council of Ministers includes a broad survey of PFAS food packaging chemicals worldwide (Trier et al., 2018).

Figure 18. A phosphate di-ester formed from EtFOSE.

Current-use alternatives in food-contact materials may be based on:

- Short-chain replacements for the FOSE-like products, such as N-ethyl perfluorobutane sulfonamidoethanol (EtFBSE) (Geueke, 2016).
- Fluorotelomer acrylate and methacrylate side-chain polymers made with short-chain fluorotelomer intermediates. It should be noted that although these are discussed in literature, none of these types of products (fluorotelomer-based PAPs) are approved products on the U.S. Food and Drug Administration's (FDA) current Food Contact Notification (FCN) listing.
- Fluorotelomer-based mono-, di-, and triPAPs (such as tri-polyfluoroalkyl phosphoric acid) (Zabaleta et al., 2017).
- Perfluoropolyethers (Wang et al., 2013).

1.4.3 Specialty chemicals

Fluorinated surfactants are used in innumerable industrial and consumer products, where they provide advantages both during application and in the final product performance. Paints, coatings, and sealants need to wet the solid substrate and penetrate into crevices or other imperfections. The final finish should be smooth and level. These performance characteristics are all facilitated by the very low surface tension obtained using fluorosurfactants. When appropriately formulated, the same or related surfactants can impart water, oil, and dirt resistance to painted walls, sealed grout, or polished floors.

PFAS are also used in a wide-range of functional fluids. These include lubricants for use in harsh or reactive environments such as space applications, vacuum pump fluids, and heat transfer fluids. Other specialty applications include friction reduction, anti-adhesion products, and anti-squeak products used in automotive applications. Perfluoroethers or polyethers can also be used (as illustrated in Section 1.4.5 below) as polymer processing aids.

Liquid-applied products vary substantially by type, and the specialty chemical market requires a broad range of surfactant designs. Knepper and Lange (2012) provides a number of examples with supporting literature references. A study of commercial products purchased around 2010 (such as the carpet protector in Figure 15 above) often contained a mix of PFAA chain-lengths (4-C to 12-C PFAAs were quantified) (Liu et al., 2012). The potassium salt of glycine, N-ethyl-N-[(heptadecafluorooctyl)sulfonyl] (Chemical Abstract Services Registration Number [CASRN] 2991-51-7, also marketed as Fluorad 129, now discontinued) is a typical legacy POSF-based substance used in cleaning agents and polishing products (Poulsen et al., 2005). Figure 19 provides an illustrative schematic of this compound.

Figure 19. Typical legacy POSF-based surfactant used in liquid-applied products.

As for the applications described above, current-use surfactant products can be similar in structure to the legacy products, but with shorter perfluorinated chains. Product brochures from major manufacturers identify 4-C (PFBS) and 6-C (6:2 FTOH) chemistries for a wide-range of product types (3M, 2016; DuPont, 2008).

1.4.4 Fire-fighting chemicals

PFAS-based AFFF (aqueous film forming foams) were developed in the 1960s to extinguish Class B flammable liquid fuel fires. After extinguishing the fire, the foam-surfactant film acts as a radiation barrier and vapor-sealant to prevent re-ignition of the fuel or "burnback." Impacts of AFFF use are discussed in additional detail in Appendix 3: Sources and Uses, Section 3.2 Aqueous film forming foam.

While PFCAs were used only in the earliest AFFF formulations, POSF-based products dominated the market in the 1970s and later (Prevedouros et al., 2006). Many 1970 – 2000-era AFFF products were PFSA-based, with derivatives of perfluoroalkylsulfamido amines and PFOS as "major presence(s)" (Favreau et al., 2017).

Formulations for the military produced in the 1980s-to-early 2000s contained perfluorinated chains up to 8-, 9-, and 10-C in some cases (Place & Field, 2012). The current generation fluorotelomer-based AFFF products are shorter perfluorinated chains, such as the 6:2 fluorotelomer sulfonamide alkylbetaine (6:2 FTAB, Figure 20) (Wang et al., 2013). Figure 20 provides a schematic of the structure of a 6:2 FTAB surfactant typical of fire-fighting foam products. These can also carry a three-digit prefix indicating three types of carbons: X:Y:Z (perfluorinated-polyfluorinated-non-fluorinated) carbons (Place & Field, 2012). Foam concentrates may contain additional surfactants (PFAS and non-PFAS) as well as other adjuvants.

Figure 20. 6:2 FTAB surfactant typical of fire-fighting foam products.

1.4.5 Polymer processing aids

Fluorinated surfactants are used as emulsifiers in aqueous reaction systems, for example, the emulsion polymerization of PTFE. Sodium and ammonium salts of PFOA and PFNA were widely used in the U.S. and Europe, and their use continues in developing and transitional economies.

Newer processing aids identified in the literature are functionalized ethers or polyethers, which contain single or multiple ether O-atoms. Among these are (Wang et al., 2013):

- Ammonium 3H-perfluoro-3-[(3-methoxy-propoxy)propanoic acid], CASRN 958445-44-8 (illustrated in Figure 21 to the left).
- Ammonium perfluoro-2-propoxypropanoic acid (PFPrOPrA), CASRN 62037-80-3 (illustrated in Figure 21 to the right).

Figure 21. Two processing aids used in fluoropolymer production.

1.5 Data gaps and recommendations

1.5.1 Data gaps

While much of the discussion of PFAS focuses on well-known substances like PFOA, PFOS, and perfluorohexanoic acid (PFHxA), as stated in the introduction, there are thousands of different PFAS in use. The applications, properties, and fate of these is in most cases proprietary or unknown.

1.5.2 Recommendations

Proper understanding of PFAS structures and characteristics is necessary to inform recommended activities described in the PFAS Chemical Action Plan (CAP) recommendations.

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List of acronyms

General acronyms

Table 8. Acronyms found in the chemistry appendix.

Acronym	Definition
CAP	Chemical Action Plan
ECF	Electrochemical fluorination
EPA	United States Environmental Protection Agency
FCN	Food contact notification
FDA	United States Food and Drug Administration
g	grams
g/mol	Grams per mole
ITRC	Interstate Technology Regulatory Council
mol	Mole
OECD	Organisation for Economic Co-operation and Development

Chemical names

Table 9. Chemical name acronyms found in the chemistry appendix, excluding general acronyms listed only in the table above.

Acronym	Chemical name
6:2 FTAB	6:2 fluorotelomer sulfonamide alkylbetaine
8:2 FTI	8:2 fluorotelomer iodide
11CI-PF3OUdS	11-chloroeicosafluoro-3-oxaundecane-1-sulfonic acid
APFO	Ammonium perfluorooctanoate
С	Carbon
diPAPs	Per- or polyfluoroalkyl phosphate di-esters
EtFBSE	N-ethyl perfluorobutane sulfonamidoethanol
EtFOSE	N-Ethyl perfluorooctane sulfonamidoethanol
F	Fluorine
FTAC	Fluorotelomer acrylate
FTI	Fluorotelomer iodide
FTO	Fluorotelomer olefin
FTOH	Fluorotelomer alcohol
Н	Hydrogen
HFPO	Hexafluoropropylene oxide
HFPO-DA(GenX)	Hexafluoropropylene oxide dimer acid
MeFOSE	N-Methyl perfluorooctane sulfonamido-ethanol

Acronym	Chemical name
monoPAPs	Per- or polyfluoroalkyl phosphate esters
0	Oxygen
PAP	Per- or polyfluoroalkyl phosphate ester
PBSF	Perfluorobutane sulfonyl fluoride
PFAA	Perfluorinated alkyl acid
PFAS	Per- and poly-fluorinated alkyl substances
PFCA	Perfluoro-carboxylic acid
PFECA	Perfluoroalkyl ether carboxylic acid
PFHxA	Perfluorohexanoic acid
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFPE	Perfluoropolyether
PFPrOPrA	Perfluoro-2-propoxypropanoic acid
PFSA	Perfluorooctane sulfonic acid
PFTrDA	Perfluorotridecanoic acid
PFUnDA	Perfluoroundecanoic acid
POSF	Perfluorooctane sulfonyl fluoride
PTFE	Polytetrafluoroethylene
SAmPAPs	Perfluorooctane sulfonamidoethanol-based phosphate esters
triPAPs	Per- or polyfluoroalkyl phosphate tri-esters

Appendix 2: Analytical Methods

2.0 Overview

2.0.1 Findings

A variety of analytical methods are available for the analysis of per- and polyfluoroalkyl substances (PFAS) in the environment and consumer products. Analytical methods for PFAS analysis are still evolving. Currently, few methods are formally validated and published.

A multi-laboratory validated method, United States Environmental Protection Agency (EPA) Method 537.1 version 1.0 (EPA, 2018), was published in November 2018 for the analysis of 18 PFAS analytes in drinking water. Method 537.1 is a solid phase extraction (SPE) liquid chromatography/tandem mass spectrometry (LC/MS/MS). Surrogate and internal standards are used to monitor for analyte loss due to sample preparation, instrument drifts, or matrix effects. This method is limited to the analysis of selected PFAS in drinking water samples.

In March 2020, EPA updated Method 537.1. Method 537.1 Revision 2.0 is an editorial update to Method 537.1 Revision 1.0 that includes method flexibility to improve the method performance. Method 537.1 measures PFAS in drinking water using solid phase extraction and LC/MS/MS at low ng/L concentrations.

In December 2019, EPA announced a new validated method for testing additional PFAS in drinking water, EPA Method 533. EPA's Method 533 focuses on those PFAS with carbon chain lengths of 4-12. This method complements EPA Method 537.1 Revision 1.0, and can be used to test for 11 additional PFAS. Both methods (537.1 and 533) can measure a total of 29 PFAS in drinking water.

In June 2019, EPA published a validated SW-846 Method 8327—Per- and polyfluoroalkyl substances (PFAS) Using External Standard Calibration and Liquid Chromatography/Tandem Mass Spectrometry (LC/MS/MS). This method conducts a two-phase study for 24 PFAS analytes and 19 isotopically-labeled PFAS surrogates in four aqueous matrices of reagent water, surface water, groundwater, and wastewater effluent, three of which were intended to represent non-potable water matrices.

Draft Method 8328 is tentatively scheduled to be issued by EPA in 2020. The draft Method 8328 will make use of solid-phase extraction (SPE) for non-drinking water aqueous samples and solvent extraction for solid matrices. Isotope dilution (ID) will also be incorporated into Method 8328.

Other published standard methods for PFAS analysis that have not been multi-laboratory validated include the American Society for Testing and Materials International (ASTM) D7979-17 (ASTM, 2017). This method is a direct injection method that requires very little sample preparation. The method can be applied for wide range of liquid environmental samples such

as surface water, groundwater, and wastewater influent and effluents. Another method, ASTM D7968-17a (ASTM, 2017a), was developed for analyzing PFAS in soil matrices.

The importance of multiplatform approach for accurately characterizing PFAS is discussed in this appendix. The multiplatform approach comprises a novel workflow combining target analysis and non-target screening analysis (NTA), in addition to extractable organic fluorine with combustion ion chromatography (EOF/CIC) for the determination of total fluorine (TF), and inorganic fluoride (IF) analysis to characterize the chemical composition of both known and unknown PFAS. This approach resulted in the identification of more PFAS chemicals that were not included in the targeted analysis, but were prioritized samples from EOF for suspect screening and quantification. By using these approaches, the sum of the targeted PFAS and total organic organofluorine concentration were determined, as well as a mass balance of known and unknown organofluorine.

A specific multiplatform approach could be used to identify and quantify multiple PFAS chemicals, and provide data on PFAS presence in varying types of environmental media. Ecology supports the use of approved validated methods as recommended by EPA for specific targeted PFAS analysis.

An important shortcoming of the multiplatform non-target approach is that these methods are not standardized or multi-laboratory validated. For regulatory purposes, standard validated methods such as EPA-validated PFAS analytical methods are recommended. Non-targeted analysis techniques are not validated, and may not be used for regulatory purposes. The uses of these methods are limited to research and investigation.

2.0.2 Introduction

The objective of this appendix is to evaluate the current available analytical methods for the analysis of PFAS in the environment and consumer products. This review includes an assessment of the standard and non-standard analytical methods for the analysis of PFAS. The performance challenges with current standard methods for PFAS analysis and suggested analytical techniques for measuring PFAS are also discussed.

Buck et al. (2011) provides an expanded overview of PFAS in the environment, terminology, classification, and their contributory sources. EPA has an online resource for PFAS (EPA, 2019). The Interstate Technology Regulatory Council (ITRC) has developed a series of fact sheets that summarize the latest science and emerging technologies regarding PFAS (ITRC, 2018).

The ITRC fact sheet describes methods for evaluating PFAS in the environment, including laboratory analytical methods for PFAS (ITRC, 2018). There are several published papers and literature reviews on analytical methods or techniques for the determination of PFAS in various matrices (Berger et al., 2011; De Voogt et al., 2006; Jahnke et al., 2009). The analytical methods used for PFAS determination are dominated by chromatography, mostly in combination with mass spectrometric detection.

High performance liquid chromatography (HPLC) hyphenated with conductivity or fluorimetric detection and gas chromatography combined with flame ionization or electron capture detection have been used for PFAS analysis (Mahmoud et al., 2009; Moody et al., 2001; Schultz et al., 2004; Trojanowicz et al., 2013). These methods are used for the analysis of specific, targeted PFAS analytes. Most PFAS fractions are quantified during targeted liquid chromatography mass spectrometric (LC/MS/MS) analysis. Commercially relevant internal standards are available for most of the method analytes, however many of the branch isomers are unknown and standards are not available. As the list of PFAS analytes grows, corresponding isotopically labeled internal standards for these analytes may become available. Otherwise, definitive identification and quantitative analysis are difficult or impossible.

2.1 Published standard methods for PFAS analysis

The following standard methods have been used for PFAS analysis. For detailed procedure and quality control requirements for each method, see the referenced standard methods.

2.1.1 Drinking water methods

The following drinking water methods have been tested and validated. Tested and validated methods are important for ensuring that government and private laboratories can accurately and consistently measure PFAS in the environment, which is critical for estimating exposure and risk.

Method 537.1

EPA Method 537—Determination of Selected Per- and Polyfluorinated Alkyl Substances in Drinking Water by Solid Phase Extraction and Liquid Chromatography/Tandem Mass Spectrometry (LC/MS/MS)—was first published in 2009 for the determination of 14 PFAS in drinking water using SPE and LC-MS/MS (Shoemaker et al., 2008). Table 10 lists the original 14 PFAS determined using Method 537.

A multi-laboratory validated method, EPA method 537.1 version 1.0, was published in November 2018 for the analysis of 18 PFAS analytes in drinking water, including the 14 originally determined using Method 537 and four additional PFAS (Shoemaker & Tettenhorst, 2018). New analytes in the updated method, also shown in Table 10, include for example the GenX (hexafluoropropylene oxide dimer acid [HFPO-DA]) and hexafluoropropylene oxide dimer acid (Kato et al., 2008; Strynar et al., 2015). However, non—targeted liquid chromatography with high-resolution mass spectrometer (LC-HRMS) can be applied to identify additional suspected or uncharacterized PFAS if analytical standards are available for PFAS identification and quantification (McDonough et al. 2019).

In March 2020, EPA further updated Method 537.1. Method 537.1 Revision 2.0 is an editorial update to Method 537.1 Revision 1.0 that includes method flexibility to improve the method performance. Method 537.1 measures PFAS in drinking water using solid phase extraction and

LC/MS/MS at low ng/L concentrations (Shoemaker & Tettenhorst, 2020). The method flexibility incorporated into revision 2.0 permits laboratories to modify the techniques in the method such as the evaporation and separation techniques. However, changes may not be made to sample collection and preservation, sample extraction steps, or to quality control requirements. EPA recommends that method modifications should be considered only to improve method performance. Modifications that are introduced in the interest of reducing cost or sample processing time, but result in poorer method performance, should not be used.

Analysis of short-chain PFAS using Method 533

In December 2019, EPA announced a new validated method for testing additional PFAS in drinking water, EPA Method 533. EPA's Method 533 focuses on those PFAS with carbon chain lengths of four to twelve, and complements EPA Method 537.1 version 1.0. It can be used to test for 11 additional PFAS, as shown in Table 10. Used together, Methods 537.1 and 533 can measure a total of 29 PFAS chemicals in drinking water.

EPA Method 533 is a SPE LC/MS/MS method for the determination of select PFAS in drinking water. Method 533 requires the use of MS/MS in Multiple Reaction Monitoring (MRM) mode to enhance selectivity. Method 533 incorporates ID, which can minimize sample matrix interference and improve data quality (Rosenblum et al., 2019).

Table 10. EPA validated Methods 537, 537.1 and 533 analyte list.

Analyte	Abbreviation	CASRN	Method 537	Method 537.1	Method 533
11-Chloroeicosafluoro-3-oxaundecane-1-sulfonic	11Cl- PF3OUdS	763051-92- 9		Х	Х
9-Chlorohexadecafluoro-3-oxanonane-1-sulfonic acid	9CI-PF3ONS	756426-58- 1		Х	Х
4,8-Dioxa-3H- perfluorononanoic acid	ADONA	919005-14- 4		Х	X
Hexafluoropropylene oxide dimer acid	HFPO-DA	13252-13-6		Х	Х
Perfluorobutanesulfonic	PFBS	375-73-5	Х	Х	Х
Perfluorodecanoic acid	PFDA	335-76-2	Х	Х	Х
Perfluorododecanoic acid	PFDoA	307-55-1	Х	Х	Х
Perfluoroheptanoic acid	PFHpA	375-85-9	Х	Х	Х
Perfluorohexanoic acid	PFHxA	307-24-4	Х	Х	Х
Perfluorohexanesulfonic	PFHxS	355-46-4	Х	Х	Х
Perfluorononanoic acid	PFNA	375-95-1	Х	Х	Х
Perfluorooctanoic acid	PFOA	335-67-1	Х	Х	Х

Analyte	Abbreviation	CASRN	Method 537	Method 537.1	Method 533
Perfluorooctanesulfonic acid	PFOS	1763-23-1	Х	Х	Х
Perfluoroundecanoic acid	PFUnA	2058-94-8	Х	Х	Х
1H,1H, 2H, 2H- Perfluorohexane sulfonic acid	4:2FTS	757124-72- 4			Х
1H,1H, 2H, 2H- Perfluorooctane sulfonic acid	6:2FTS	27619-97-2			Х
1H,1H, 2H, 2H- Perfluorodecane sulfonic acid	8:2FTS	39108-34-4			Х
Nonafluoro-3,6- dioxaheptanoicacid	NFDHA	151772-58- 6			Х
Perfluorobutanoic acid	PFBA	375-22-4			Х
Perfluoro(2- ethoxyethane)sulfonic	PFEESA	113507-82- 7			Х
Perfluoroheptanesulfonic acid	PFHpS	375-92-8			Х
Perfluoro-4- methoxybutanoic acid	PFMBA	863090-89- 5			Х
Perfluoro-3- methoxypropanoicacid	PFMPA	377-73-1			Х
Perfluoropentanoic acid	PFPeA	2706-90-3			Х
Perfluoropentanesulfonic acid	PFPeS	2706-91-4			Х
N-ethyl perfluorooctane- sulfonamidoacetic acid	NEtFOSAA	2991-50-6	Х	Х	
N-methyl perfluorooctane- sulfonamidoacetic acid	NMeFOSAA	2355-31-9	Х	Х	
Perfluorotetradecanoic acid	PFTA	376-06-7	Х	Х	
Perfluorotridecanoic acid	PFTrDA	72629-94-8	Х	Х	

Note: X denotes that the method can be used to test for the specified analyte.

These methods (533 and 537.1) measure all forms of the analytes as anions while the identity of the counterion is inconsequential. Method 533 could be used for a variety of environmental monitoring applications, which include the analysis of multiple short-chain PFAS that cannot be measured by Method 537.1 (Rosenblum et al., 2019).

In Method 533, the concentration of each analyte is calculated using the ID technique. For quality control (QC) purposes, the percent recoveries of the ID analogues are calculated using the integrated peak areas of isotope performance standards, which are added to the final extract and function as traditional internal standards, exclusively applied to the ID analogues.

Quantitation of linear and branch isomers of PFAS with drinking water methods

Accurate quantification of PFAS that are mixtures of linear isomers and branched isomers in environmental matrices is useful in understanding both the sources of PFAS and the age of the source, since the production of isomers varies by manufacturing processes. However, such quantification of PFAS can be difficult (Riddell et al., 2009).

With EPA Method 537, laboratories had difficulty in quantifying both linear and branch isomers of perfluorooctanoic acid (PFOA) (Shoemaker & Tettenhorst, 2018). To account for linear and branched isomers of PFOA, EPA recommends that integration and quantitation of drinking water samples include peaks that represent both linear and branched isomers. EPA notes that the correct application of the method is to calibrate using a certified quantitative standard that includes both the linear and branched isomers of each analyte, if available. As of the release of EPA's (2016) technical advisory, there is no certified quantitative mixed standard for PFOA, and the available PFOA standards can be used to account for mixed isomers.

Since there is currently no certified quantitative PFOA standard that contains both linear and branched isomers and that can be used to quantitate in the traditional manner, EPA recommends that until such standard is available, labs use the following approach (EPA, 2016):

- Calibrate instrumentation using a certified quantitative standard containing only the linear isomer.
- Identify the branched isomers by analyzing a qualitative or semi-quantitative PFOA
 mixed standard that includes both linear and branched isomers (Wellington
 Laboratories, cat#: T-PFOA or equivalent), and compare retention times and tandem
 mass spectrometry transitions.
- Quantitate PFOA by integrating the total response (i.e., accounting for peaks that are identified as linear and branched isomers) and relying on the initial calibration with the linear-isomer quantitative standard.

Method 533 includes procedures for summing the contribution of multiple isomers to the final reported concentration. Where standard materials containing multiple isomers are commercially available, laboratories are encouraged to obtain the standards for the method analytes. The technical grade standards are used to identify retention times of branched and linear isomers of method analytes (Rosenblum et al., 2019).

2.1.2 Non-drinking water sample methods

Methods 537.1 Revision 2.0 and 533 are specified for analyzing PFAS in drinking water. As a result, they are not amenable to an expanded list of PFAS compounds or to analysis of other sample matrices without modification of the method. Method 537.1 Revision 2.0 only permitted modification to the method techniques for application to drinking water analysis. For example, it would not work well for the determination of PFAS in consumer products or non-water matrices. Proprietary non-standard methods based on modifications of Method 537 are used by various commercial laboratories for the determination of PFAS in non-drinking water samples. The U.S. Department of Defense (DOD), Environmental Laboratory Accreditation Program (ELAP) maintains a list of laboratories for the determination of PFAS in various environmental media other than drinking water on the <u>Defense Environmental Network Information Exchange (DENIX) server.</u>

With lack of standardization among laboratories performing Method 537 modified, Ecology recommends, as part of the laboratory selection process for non-drinking water analysis (e.g. consumer product), the laboratory analytical procedure should be evaluated based on the DOD Quality Systems Manual (QSM) to ensure all parameters meet acceptance criteria for all analytical QC elements. The QC elements should be evaluated to ensure that they are set at levels that meet the project's measurement quality objectives (MQOs). The laboratories are required to provide an initial demonstration of capability (IDC) consistent with the DOD QSM for Ecology bid evaluation. The QC criteria should not be less stringent than the criteria found in the DOD QSM, Version 5.1, Appendix B, Table B-15 (DOD, 2017a) or later version.

Currently, DOD QSM for Environmental Laboratories, Version 5.1.1, Table B-15 (DOD, 2017a) provides the most current and comprehensive set of quality standards for PFAS analysis. These performance-based standards outline specific quality processes for sample preparation, instrument calibration and analysis when working with PFAS. The DOD QSM, Version 5.1, Table B-15, criteria currently require ID quantitation of PFAS. The ID method accounts for interferences caused by complex sample matrices and bias introduced by sample preparation and instrumental issues.

EPA SW-846 Method 8327

In June 2019, EPA published a validated SW-846 Method 8327—Per- and Polyfluoroalkyl Substances (PFAS) Using External Standard Calibration and Liquid Chromatography/Tandem Mass Spectrometry (LC/MS/MS) (EPA, 2019). This method consists of a two-phase study for 24 PFAS analytes and 19 isotopically-labeled PFAS surrogates in four aqueous matrices of reagent water, surface water, groundwater, and wastewater effluent, three of which were intended to represent non-potable water matrices. As identified in Tables 11, 12, 13, and 14 below, the PFAS targets included sulfonic acids (e.g., perfluorooctane sulfonic acids (PFOS)) (Table 11), carboxylic acids (e.g., PFOA) (Table 12), fluorotelomer sulfonic acids (e.g., 6:2 fluorotelomer

⁹⁰ https://www.denix.osd.mil/edqw/accreditation/accreditedlabs/

sulfonate [FTS]) (Table 11), and sulfonamides and sulfonamidoacetic acids (e.g., N-methyl perfluorooctanesulfonamidoacetic acid [N-MeFOSAA]) (Table 14).

Target compounds are identified by comparing multiple reaction monitoring (MRM) transitions in the sample to MRM transitions in the standards. The retention time (RT) and qualifier ion ratio are compared to a mid-level standard to support qualitative identification. Target compounds are quantitated based on the response of their quantifier MRM transitions utilizing external standard calibration. See reference for method detail (EPA, 2019).

Standards for some target analytes may consist of mixtures of structural isomers; however, the Chemical Abstracts Service Registry Number (CASRN) listed in the tables below is for the normal-chain isomer. All CASRNs in the table are for the acid form. Sulfonic acids in stock standard mixes are typically received as the sodium or potassium salt form. CASRNs for the salt form are not included (EPA, 2019).

Analytes marked with an asterisk (*) in the tables exhibit known difficulties with reproducibility, response, recovery, stability, and/or chromatography that may reduce the overall quality or confidence in the result when using this method. This analyte may require special care to ensure analytical performance will meet the needs of the project and, where necessary, may also require the use of appropriate data qualification. See Section 1.3 of the referenced method for specific information regarding these analytes (EPA, 2019).

Table 11. Method 8327 PFAS analytes: PFAS sulfonic acids.

Analyte	CASRN
Perfluoro-1-butanesulfonic acid (PFBS)	375-73-5
Perfluoro-1-pentanesulfonic acid (PFPeS)	2706-91-4
Perfluoro-1-hexanesulfonic acid (PFHxS)	355-46-4
Perfluoro-1-heptanesulfonic acid (PFHpS)	375-92-8
Perfluoro-1-octanesulfonic acid (PFOS)	1763-23-1
Perfluoro-1-nonanesulfonic acid (PFNS)	68259-12-1
Perfluoro-1-decanesulfonic acid (PFDS)	335-77-3
1H, 1H, 2H, 2H-perfluorohexane sulfonic acid (4:2 FTS)	757124-72-4
1H, 1H, 2H, 2H-perfluorooctane sulfonic acid (6:2 FTS)*	27619-97-2
1H, 1H, 2H, 2H-perfluorodecane sulfonic acid (8:2 FTS)*	39108-34-4

Table 12. Method 8327 PFAS analytes: PFAS carboxylic acids.

Analyte	CASRN
Perfluorobutanoic acid (PFBA)*	375-22-4
Perfluoropentanoic acid (PFPeA)*	2706-90-3
Perfluorohexanoic acid (PFHxA)*	307-24-4
Perfluoroheptanoic acid (PFHpA)	375-85-9
Perfluorooctanoic acid (PFOA)	335-67-1
Perfluorononanoic acid (PFNA)	375-95-1
Perfluorodecanoic acid (PFDA)	335-76-2
Perfluoroundecanoic acid (PFUdA)*	2058-94-8
Perfluorododecanoic acid (PFDoA)*	307-55-1
Perfluorotridecanoic acid (PFTrDA)*	72629-94-8

Table 13. Method 8327 PFAS analytes: Perfluorotetradecanoic acid.

<u>Analyte</u>	CASRN
Perfluorotetradecanoic acid (PFTeDA)*	376-06-7

Table 14. Method 8327 PFAS analytes: PFAS sulfonamides and sulfonamidoacetic acids.

Analyte	CASRN
N-ethylperfluoro-1-octanesulfonamidoacetic acid (N-EtFOSAA)*	2991-50-6
N-methylperfluoro-1-octanesulfonamidoacetic acid (N-MeFOSAA)*	2355-31-9
Perfluoro-1-octanesulfonamide (FOSA)	754-91-6

EPA Method 8328

Draft Method 8328 is tentatively scheduled to be issued by EPA in 2020. Draft Method 8328 will make use of solid-phase extraction (SPE) for non-drinking water aqueous samples and solvent extraction for solid matrices. ID will also be incorporated into this method (Mills & Impellitteri, 2019).

It is a more complex method relative to direct injection. The method will account for matrix effects (e.g., sorption) through isotopically marked standard recoveries, and the options to meet DOD requirements. The method is amenable to the same 24 PFAS as in Method 8327 plus GenX in matrices consisting of non-drinking water sources (surface water, groundwater, wastewater) and solids (soils, sediments, biosolids). Two-lab internal validation is ongoing, and

an additional ten-lab external validation study is planned. EPA is exploring collaborative efforts with DOD on external validation. The target quantitation limit for Method 8328 is 10 nanograms (ng)/liter(L).

Other EPA methods in development

Table 15 below summarizes the description and status of additional methods EPA is developing and validating to detect and quantify selected PFAS in air, water, and soil (EPA, 2020).

Table 15. EPA method development and validation to detect and quantify selected PFAS in air, water, and soil (EPA, 2020).

Title	Description	Status
Clean Water Act	CWA Method 1600 is a draft ID method that	Expect to complete
(CWA) Method 1600	EPA is validating to measure 24 PFAS in	validation in the first
	surface water, groundwater, wastewater,	quarter of 2021 and
	solids (soils, sediments, and biosolids), and	post the final method
	tissues. These 24 PFAS are the same as SW-	in the second quarter
	846 Method 8327. EPA is collaborating with DOD to validate.	of 2021.
OTM Method 45	Draft method to measure PFAS in air	Expected fourth
	emissions.	quarter 2020.
Standard Operating	TOF methods can indicate the total amount of	This work is in its
Procedures for Total	fluorine present and may be a viable approach	early stages.
Organic Fluorine	to quickly screen for PFAS and to identify	
(TOF)	situations where more specific measurements are needed.	
Analytical Model to	High resolution mass spectrometry (HRMS) to	Ongoing.
Identify Novel PFAS	qualitatively identify PFAS substances without	
Using Non-Targeted	a standard method.	
Analysis Data		

International Organization for Standardization (ISO) Method 25101:2009: SPE in water

ISO 25101:2009 specifies a method for the determination of the linear isomers of PFOS and PFOA in unfiltered samples of drinking water, groundwater, and surface water (fresh water and sea water) using high-performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS) (ISO 2009, reviewed 2014). Analytes are extracted from water samples by solid phase extraction (SPE) followed by solvent elution and determined by HPLC-MS/MS. Other isomers may be reported separately as non-linear isomers and qualified as such. The method is applicable to a concentration range of 2 - 10,000 ng/L for PFOS and 10 - 10,000 ng/L for PFOA. Depending on the matrix, the method may also be applicable to higher concentrations ranging from 100 - 200,000 ng/L after suitable dilution of the sample or reduction in sample size.

ASTM D7979: Direct injection—surface and wastewater

ASTM D7979 have been successfully used in the determination of selected PFAS in water matrices (e.g., sludge and wastewater influent and effluent) using liquid chromatography (LC) and detection with tandem mass spectrometry (MS/MS) (ASTM 2017). This method adheres to a technique known as selected reaction monitoring (SRM) or sometimes referred to as multiple reaction monitoring (MRM). This is not a drinking water method—performance of this test method has not been evaluated on drinking water matrices. ASTM D7979 is a performance-based method, and alternative operating conditions can be used to perform this method provided data quality objectives are attained. It is a direct injection method that does not require sample preparation.

ASTM D7979 (2017) currently covers the analysis of 21 PFAS compounds, with ten additional compounds listed for consideration in the appendix of the method. Eight additional PFAS compounds, including three emerging PFAS compound of interest (11-chloroeicosafluoro-3-oxaundecane-1-sulfonic acid [11Cl-PF3OUdS], 9-chlorohexadecafluoro-3-oxanone-1-sulfonic acid [9Cl-PF3ONS], and 4,8-dioxa-3H-perfluorononanoic acid [ADONA]), have been determined by the method to a total of 39 PFAS analytes (Waters, 2018).

Since the publication of this international standard, there have been many comments regarding the method, ranging from applicability to matrices, detection limits, lack of solid phase extraction, and calibration model.

ASTM D7968: Solids (soil)

This method was developed by EPA Region 5 Chicago Regional Laboratory, and has been successfully used for the determination of selected PFAS in a soil matrix (ASTM, 2017a). It is similar in scope to ASTM D7979-17 and uses solvent extraction and filtration, followed by LC/MS/MS to qualitatively and quantitatively determine PFAS in soil. Thirty analytes can be detected with this method, including but not limited to:

- Eleven perfluoroalkyl carboxylic acids.
- Three perfluoroalkyl sulfonates.
- Decafluoro-4- (pentafluoroethyl) cyclohexanesulfonate.
- Six fluorotelomers.

This is also a performance-based method, and alternative operating conditions can be used to perform this method, provided that all data quality objectives defined in the method are attained. It is recommended that QC and quality assurance requirements, if not well defined in the standard methods, must not be less stringent than the PFAS requirement found in DOD QSM (2017), Version 5.1 or later, Appendix B, and Table B-15, for media types.

2.2 Non-specific methods for PFAS analysis

Many of the available standard methods for PFAS analysis do not account for all known PFAS. Human exposures to PFAS are generally not from individual PFAS, but from a complex mixture (Schaider et al., 2017), and analytical techniques are limited for determining which PFAS constituents are in a given mixture. Hence, the full extent of PFAS contamination could be underestimated when targeted analytical methods are used to quantify PFAS concentration. The complexity of PFAS, the production of commercial mixtures, and the tendency to generate intermediate transformation products (Guelfo et al., 2018) present a performance challenge for current targeted methods.

Targeted analytical methods have been used successfully in quantitation of known PFAS chemicals (Lacorte et al., 2006), but they may not be feasible in the quantitation of more than 4,700 PFAS that are recognized today (Thermofisher, 2018). Unknown PFAS—including new alternatives or legacy substances, their transformation products, and residual impurities—may contribute to a substantial proportion of unknown organic fluorine in the environment.

These unknowns represent a great source of uncertainty for ascertaining environmental and human health risks (Liu et al., 2019). Analytical approaches that can discover and characterize such unknown PFAS are a first step to facilitating knowledge on the hazards and environmental behaviors of these unknown chemicals. Studies have indicated that scientists are using techniques that focus on measuring the total exposure of all PFAS instead of one or a limited set of PFAS. This is important to gain a better understanding of exposures to PFAS as a class (Hartmann et al., 2107; Poothong et al., 2017).

In a published study by the Nordic Council of Ministers analyzing PFAS and TOF in products, comparison between analyzed individual PFAS and TOF concentration showed that individual PFAS constitute a small proportion of the TOF (Borg et al., 2017). It indicates a data gap relative to the unknown or potentially uncharacterized PFAS by conventional analytical techniques. The TOF method is capable of measuring TOF at ultra-trace levels and checking the mass balance, but cannot trace the individual analytes present in the sample (Ateia et al., 2019).

Schultes et al. (2019) also compared combustion ion chromatography (CIC) based EOF to target PFAS measurement in food packaging samples by LC/MS/MS. The study revealed large amounts of unidentified organic fluorine not captured by compound-specific analysis.

Liu et al. (2019), in their literature review of HRMS for non-targeted analysis, reported unknown PFAS discovery in commercial products, surfactant concentrates in environmental samples, sediment, soil, airborne particulate matter, and concrete, as well as in biological matrices, polar bears, and human serum.

2.2.1 Non-standard analytical techniques for measuring PFAS

McDonough et al. (2019) evaluated analytical techniques for measuring total (bulk) organofluorine developed for the study and quantification of unidentified fractions of PFAS in environmental and biological samples. These methods or techniques vary in applicability to different sample matrices, and in their selectivity and sensitivity. Description of each technique follows.

Combustion ion chromatography (CIC) methods

Combustion ion chromatography mineralizes and then measures organic fluorine from the EOF and absorbable organic fluorine (AOF) assay. Samples are combusted at a temperature of 900 – 1,000 degree Celsius (C) to convert organic fluorine to hydrofluoric acid, which is then absorbed into a solution of sodium hydroxide (McDonough et al., 2019). The total concentration of the fluoride is subsequently measured by ion chromatography (IC) after calibration with sodium fluoride. The choice of sample preparation is important in isolating organic fluorine from fluoride prior to CIC analysis, since CIC will not differentiate between organic and inorganic fluorine, and does not identify individual PFAS.

In EOF, the organic fluorine fraction is isolated by ion pairing methods and TOF is measured by CIC. The EOF assay is the most commonly used assay found in literature for total organic fluorine measurement in different environmental matrices, in human blood (Miyake et al., 2007, Yeung et al., 2013), and in marine mammals (Yeung et al., 2009).

Wagner et al. (2013) described the AOF assay, which differs in the way the organo-fluorine is extracted from the sample matrix. In AOF, the sample is passed through cartridges containing synthetic polystyrenedivinylbenzene-based activated carbon (AC). Residual fluoride is removed with a sodium nitrate washing solution, and the AC absorbent is then analyzed by CIC. AOF has only been applied to waters and wastewater (Dauchy et al., 2017; Wagner et al., 2013).

Particle-induced gamma ray emission (PIGE)

PIGE is a non-destructive analytical technique that takes advantage of the unique gamma-ray wavelength emission of fluorine when impacted with a proton ion beam. The technique is not compound specific, but it is able to assess total fluorine content of a variety of materials isolated on a thin surface. Fluorine can be detected to a depth of approximately 200 micrometers (ym), but the precise value varies by substrate type (Ritter et al., 2017).

The sample is secured in the instrument and bombarded ex vacuo under a 3.4 Mega electron-volt (MeV) beam with an intensity of 10 nanoampere (nA) for approximately 180 seconds (s). Two gamma rays characteristic of the decay of the F nucleus (110 kiloelectron volt (keV) and 197 keV) are measured and the responses integrated. PIGE has recently been quantitatively applied to the measurement of PFAS-impacted samples by creating calibration standards consisting of textiles soaked in solution of a known organofluorine (Ritter et al., 2017).

PIGE has primarily been used for solid-phase samples such as textiles, paper, and food packaging (Lang et al., 2016, Robel et al., 2017, Schaider et al., 2017). PIGE is a rapid screening technique to measure fluoride, PFAS, and other fluorine-containing compounds in the samples. PIGE does not differentiate between inorganic fluorine and organic fluorine. It is important to understand whether there are significant sources of both organic and inorganic fluorine in a sample. There are techniques to remove inorganic fluorine that can make it specific for organofluorine if the sample does not contain a significant amount of fluoride or if the inorganic fluoride has been removed from the sample.

PIGE can detect a wide range of fluorine treatment chemicals including polymeric fluorine treatments such as polytetrafluoroethylene (PTFE), side-chain fluorinated polymers, and small molecule products.

Total oxidizable precursors (TOP) assay

Houtz and Sedlak (2012) developed the TOP assay method. The TOP assay was developed to infer and indirectly quantify the total amount of chemical precursors to perfluorinated alkyl acids (PFAA) in a sample by comparing the concentrations of specific PFAAs before and after oxidation of the sample by an excess of hydroxyl radicals (Houtz & Sedlak, 2012). It is the most selective of PFAS surrogate analytical methods, in that it selects only PFAS compounds that can be oxidized to form targeted PFAAs (McDonough et al., 2019). The same procedure of sample preparation is followed as traditionally used for targeted LC/MS/MS analysis. The assay is useful with compounds that oxidize to form LC-amenable hydroxyl radical resistant PFAS, however, these oxidation products must then also be detectable by LC/MS/MS. Some oxidation products, such as very short-chain PFAS, will not be detected by standard post-assay detection approaches such as EPA Method 537.

The assay is subject to low and variable recoveries that may lead to false negatives, especially in samples that have very low levels of PFAS (Robel et al., 2017). The limitation of the TOP assay is that it does not easily differentiate between precursors that contain telomer or sulfonamide functionalities, as all of these precursors are chemically oxidized primarily to perfluoroalkyl carboxylates. The TOP assay has not been demonstrated on large molecular weight polymer compounds or newer ether-linked PFAS like GenX. It is unknown if the oxidative process would liberate PFAAs from these types of compounds.

The TOP assay process converts fluorotelomer-based compounds including PFAA precursors into a mixture of PFAA products (Houtz & Sedlak, 2012). The increase in PFAAs measured after the TOP assay, relative to before, is a conservative estimate of the total concentration of PFAA precursors present in a sample, because not all PFAS present will be subject to quantitation or reaction, and will remain as undetected PFAS. The PFAAs generated have perfluoroalkyl chain lengths equal to or shorter than the perfluoroalkyl chain lengths present in the precursors (Dauchy et al., 2017; Houtz et al., 2013; Houtz & Sedlak, 2012; Weber et al., 2017).

The TOP assay has been applied to a number of environmental matrices such as effluent wastewater, stormwater runoff, river water and groundwater, and soil. Houtz and Sedlak (2012), Houtz et al. (2013, 2016), McGuire et al. (2014), and Harding-Marjanovic et al. (2015) have published applications of the TOP assay.

2.3 Challenges of analytical method selection

Detailed descriptions of the non-standard analytical techniques for measuring PFAS are referenced in TOP (Houtz & Sedlak, 2012), PIGE (Ritter et al., 2017), EOF (Miyake et al., 2007), and AOF (Wagner et al., 2013). These methods enable measurement of total precursors, TF, and TOF, respectively. Method choice depends on the selectivity and inclusivity of individual or cumulative PFAS needed for a given application. McDonough et al. (2019) indicated that methods that are highly inclusive—such as PIGE, which does not differentiate between organic and inorganic fluorine—are impractical for measuring PFAS-related organofluorine.

However, EOF has a unique advantage over other methods as its selectivity can be adjusted depending on the sample preparation and fractionation method, and it can be used to measure PFAS-related organofluorine present in a sample. EOF and AOF may have sufficient sensitivity to measure total PFAS in water (Miyake et al., 2007), while the sensitivity of PIGE may be limited by fluoride interferences.

Among these methods, the TOP assay is the most sensitive for individual PFAS (Houtz & Sedlak, 2012), as it utilizes LC/MS/MS of targeted precursors. However, it is limited in its ability to account for emerging PFAS of concern such as GenX and ADONA that do not oxidize. It is also prone to selectivity concerns with reverse phase liquid chromatography, meaning that compounds that are not retained by the LC columns (for example, short-chain PFAS are lost).

Although analysis of PFAS is progressing, significant challenges remain from the fact that the complete list of PFAS relevant to environmental and human exposure scenarios is still unknown. As more research and studies identify novel PFAS and precursor transformation products, an effective, comprehensive technique that is capable of quantitative non-target analysis remains elusive (Nakayama et al., 2019).

Targeted analyses with sensitive and highly specific analytical methods have made great contributions to PFAS discovery and to quantification of concentrations in human and environmental samples (Liu et al., 2019). However, the full extent of PFAS contamination may be underestimated unless non-targeted methods are used for PFAS analysis. The lack of available analytical standards means that precursors, degradation products, and transformation products will not be quantified (D'Agostino & Mabury, 2018).

Recent development in HRMS has made the discovery of unknown or suspected PFAS possible without the need for an authentic standard (Liu et al., 2019). HRMS, using technology such as

quadrupole time of flight (QTOF), generates high mass accuracy data that can be used to identify unknown compounds (Barzen-Hanson et al., 2017b; Strynar et al., 2015).

McDonough et al. (2019) recommended combining total organofluorine measurements by EOF and/or TOP assay with HRMS and with targeted analytical methods (LC/MS/MS) to obtain a full characterization of PFAS composition and sources. Although this recommendation may be specific to water, TOF measurement has been applied to other matrices (Schultes et al., 2019). Guelfo et al. (2018) suggested that coupling AOF/EOF, TOP, or PIGE with LC/MS/MS could help provide a better understanding of the total PFAS load present in a sample, but will not result in identification of all individual PFAS present.

The availability of these techniques (EOF, PIGE, and HRMS—except TOP assay) is mostly limited to non-commercial research facilities or laboratories. The quantification of PFAS that lack standards remains a challenge.

Due to the limitation of available standard methods, non-targeted analytical techniques that can measure the total PFAS concentration in multiple matrices are preferred. The selection of any non-targeted method depends on the selectivity and inclusivity for a given application.

Spaan et al. (2020) highlighted the importance of multiplatform approach for accurately characterizing PFAS. To assess whether PFAS exposure is underestimated in marine mammals, Spaan et al. (2020) performed a combination of targeted ultra performance liquid chromatography analysis tandem mass spectrometry (UPLC-MS/MS) and suspect screening (UPLC-Orbitrap-MS)—in addition to EOF/CIC for the determination of TF. This approach resulted in the identification of 63 more PFAS that were not included in the targeted analysis, but were prioritized samples from EOF for suspect screening and quantification. EOF/CIC remains a tool in determining the total PFAS as TF (except where suspect screening is required to identify the unknown PFAS from the mass balance).

In their study, Dubocg et al. (2020) also used a multiplatform approach comprising of a novel workflow combining target analysis, non-target screening analysis (NTA), TF analysis, and inorganic fluoride (IF) analysis to characterize the chemical composition of 24 firefighting foams marketed as containing PFAS as well as fluorine-free foams. By using these approaches, the study determined the sum of the targeted PFAS and total organofluorine concentration, as well as a mass balance of known and unknown organofluorine. In this study, five fluorinated substances were tentatively identified, and non-fluorinated zwitterionic betaine compounds, which are considered to be replacement substances for PFAS, were tentatively identified in the organofluorine-free foams.

Miaz et al. (2020) developed a combined method for quantitative analysis, along with suspect and non-target screening of PFAS using ultra-high pressure liquid chromatography and ultra-high resolution (Orbitrap) mass spectrometry as reported in (Spaan et al. 2020). The method was applied together with measurements of TF and EOF to pooled serum samples. This study found that targeted PFAS accounted for a smaller fraction of the EOF in the serum, indicating an

increased contribution from unidentified PFAS. Non-targeted screening found three unidentified features with neutral masses, but the authors could not confirm if they are fluorinated without structural elucidation and NTA data base mining confirmation (Miaz et al., 2020).

A multiplatform approach allows for the comparison of the sum PFAS concentrations from targeted analysis to EOF and total fluorine (Miaz et al., 2020; Spaan et al., 2020). An important shortcoming of the non-targeted methods is that they are not standardized or multi-laboratory validated. The use of these methods is limited to research and investigation. Their results cannot be used for estimating toxicological effects, preventing the use of these methods, or for regulatory purposes.

2.4 Data gaps and recommendations

2.4.1 Data gaps

Progress has been made in the targeted analysis of PFAS. However, significant challenges remain, in that the complete list of PFAS relevant to environmental and human exposure scenarios is still unknown. It is estimated that there are more than 4,000 known registered PFAS compounds (Miaz et al., 2020). Targeted PFAS analysis can only quantify a limited amount of known PFAS, and most of the targeted analytical techniques only address the anionic forms of PFAS, unable to identify cationic, zwitterionic, and neutral forms of PFAS.

These unknown PFAS represent a great source of uncertainty for ascertaining environmental and human health risks (Liu et al., 2019). Analytical approaches that can discover and characterize such unknown PFAS are a first step to facilitating knowledge on the hazards and environmental behaviors of these unknown substances.

Addressing these challenges requires analytical tools that are both selective and inclusive (analytical methods that are able to detect thousands of known and unknown PFAS). Targeted analysis using LC with either HRMS (e.g., quadrupole time-offlight; Q-TOF) or MS/MS can capture many known PFAS. Non-targeted analysis using HRMS could also be used to identify many additional suspected or previously uncharacterized PFAS.

Using LC/MS/MS or LC-HRMS for PFAS identification and quantification requires analytical standards, and standards are currently only available for about 100 of the more than 3,000 potentially relevant PFAS (Liu et al., 2019). HRMS can be used with a number of techniques for measuring TOF to study and quantify the unidentified portion of PFAS in environmental samples. CIC and PIGE can be used to identify samples with high organic fluorine content, which can then be selected for non-target HRMS analysis.

CIC has been used to measure the TF in firefighting foams, and when combined with HRMS, can quantify the unidentified fraction of the PFAS that were unaccounted in targeted analysis (Dubocq et al., 2020). Although useful, CIC and PIGE have low sensitivity, limiting their direct

application to many environmental samples (Liu et al., 2019). EOF/CIC remains a tool in determining the total PFAS as TF (except where suspect screening is required to identify the unknown PFAS from the mass balance).

Spaan et al. (2020), Dubocq et al. (2020), and Miaz et al. (2020) highlighted the importance of multiplatform approaches for accurately characterizing PFAS. The multiplatform approach combines target analysis, non-target screening analysis (NTA), EOF/CIC for the determination of total fluorine (TF), and inorganic fluoride (IF) analysis to characterize the chemical composition of both known and unknown PFAS. This approach resulted in the identification of more PFAS chemicals that were not included in the targeted analysis but were prioritized samples from EOF for suspect screening and quantification. By using these approaches, the sum of the targeted PFAS and total organic organofluorine concentration were determined, as well as a mass balance of known and unknown organofluorine.

The multiplatform approach has been used in the study of fluorinated substances that were tentatively identified, and non-fluorinated zwitterionic betaine compounds (which are considered to be replacement substances for PFAS) that were tentatively identified in the organofluorine-free foams (Dubocq et al., 2020).

Non-targeted screening techniques are semi-quantitative and require structural elucidation and NTA data base mining confirmation. Sample pre-treatment and data analysis are not standardized. A multiplatform approach allows for the comparison of the sum PFAS concentrations from targeted analysis to EOF and total fluorine. Although these approaches are used in the discovery of unidentified PFAS, they are also useful for screening fluorinated substances in the environment and other matrices. For regulatory purposes, standard validated methods such as EPA-validated PFAS analytical methods are recommended. Non-targeted analysis techniques are not validated, and may not be used for regulatory purposes.

A specific multiplatform approach could be used to identify and quantify multiple PFAS chemicals, and provide data on PFAS presence in varying types of environmental media. Ecology supports the use of approved validated methods as recommended by EPA for specific targeted PFAS analysis. Modification of an approved standard analytical method will require Ecology approval, provided such modification is consistent with the DOD QSM.

2.4.2 Recommendations

There are no specific recommendations resulting from our analysis of analytical methods, neither of those available now nor those in process of development. Implementation of several recommendations in the CAP will require sampling and assessment of PFAS in various environmental media. This includes but is not limited to the following:

- 1.2 Technical support for site characterization, source investigation and mitigation at contaminated sites.
- 4.1 Evaluate PFAS in wastewater treatment.

- 4.2 Evaluate PFAS in landfill leachate and air emissions.
- 4.3 Evaluate Washington biosolids management.

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List of acronyms

General acronyms

Table 16. Acronyms found in the analytical methods appendix.

Acronym	Definition
AC	Activated carbon
AOF	Absorbable organic fluorine
ASTM	American Society for Testing and Materials International
CAS	Chemical Abstracts Service
CIC	Combustion ion chromatography
CWA	Clean Water Act
DENIX	Defense Environmental Network Information Exchange
DOD	Department of Defense
EOF	Extractable organic fluorine
ELAP	Environmental Laboratory Accreditation Program
EPA	United States Environmental Protection Agency
HPLC	High performance liquid chromatography
HPLC-MS/MS	High-performance liquid chromatography-tandem mass
	spectrometry
HRMS	high-resolution mass spectrometer
IC	Ion chromatography
ID	Isotope dilution
IDC	Initial demonstration of capability
IF	Inorganic fluoride
ISO	International Organization for Standardization
ITRC	Interstate Technology Regulatory Council
keV	Kiloelectron volt
L	liter
LC	Liquid chromatography
LC-HRMS	Liquid chromatography with high-resolution mass spectrometer
LC/MS/MS	Liquid chromatography/tandem mass spectrometry
MeV	Mega electron-volt
MQO	Measurement quality objective
MRM	multiple reaction monitoring
MS	Mass spectrometry
MS/MS	Tandem mass spectrometry
nA	Nanoampere
ng	nanogram

Acronym	Definition
NTA	non-target screening analysis
PIGE	Particle Induced Gamma Emission
QC	Quality Control
QSM	Quality Systems Manual
QTOF	Quadrupole time of flight
QTOF-MS	Quadrupole Time of Flight-Mass Spectroscopy
RN	Registry Number
RT	Retention time
S	second
SPE	Solid phase extraction
SRM	Selected reaction monitoring
TF	Total fluorine
TOF	Total organic fluorine
ТОР	Total Oxidizable Prec
UPLC	Ultra performance liquid chromatography
ųm	micrometer

Chemical names

Table 17. Chemical name acronyms found in the analytical methods appendix, excluding the general acronyms listed in the table above.

Acronym	Chemical name
11Cl-PF3OUdS	11-chloroeicosafluoro-3-oxaundecane-1-sulfonic acid
4:2FTS	1H,1H, 2H, 2H-Perfluorohexane sulfonic acid
6:2FTS	1H,1H, 2H, 2H-Perfluorooctane sulfonic acid
8:2FTS	1H,1H, 2H, 2H-Perfluorodecane sulfonic acid
9CI-PF3ONS	9-chlorohexadecafluoro-3-oxanone-1-sulfonic acid
ADONA	4,8-dioxa-3H-perfluorononanoic acid
FTS	6:2 fluorotelomer sulfonate
HFPO-DA (GenX)	Hexafluoropropylene oxide dimer acid
NFDHA	Nonafluoro-3,6-dioxaheptanoic acid
NEtFOSAA	N-ethyl perfluorooctanesulfonamidoacetic acid
NMeFOSAA	N-methyl perfluorooctanesulfonamidoacetic acid
PFAA	Perfluorinated alkyl acid
PFAS	Per- and poly-fluorinated alkyl substances
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonic acid

Acronym	Chemical name
PFDA	Perfluorodecanoic acid
PFDoA	Perfluorododecanoic acid
PFEESA	Perfluoro(2-ethoxyethane)sulfonic
PFHpA	Perfluoroheptanoic acid
PFHpS	Perfluoroheptanesulfinic acid
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexane sulfinic acid
PFMBA	Perfluoro-4-methoxybutanoicacid
PFMPA	Perfluoro-3-methoxypropanoicacid
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFPeA	Perfluoropentanoic acid
PFPeS	Perfluoropentanes ul fonicacid
PFTA	Perfluorotetradecanoic acid
PFTrDA	Perfluorotridecanoic acid
PFUnA	Perfluoroundecanoic acid
PTFE	Polytetrafluoroethylene

Appendix 3: Sources and Uses

3.0 Overview

3.0.1 Findings

Primary manufacturing of per- and polyfluoroalkyl substances (PFAS), involving wastewater discharges, waste disposal, and air emissions, can release PFAS into the environment. There are no known primary PFAS manufacturing operations in Washington state.

Secondary manufacturing, where PFAS are used as part of the manufacturing or industrial process, manufacturing emissions, or waste management could result in PFAS releases. These operations can include aerospace, automotive, aviation, building and construction, cable and wiring, electronics, energy, food processing, paper production, leather and textile, oil and mining, medical products, and metal plating. An estimated 1,200 Washington businesses could use PFAS or a PFAS-containing product in their operations.

Firefighting foam can release PFAS to the environment during use, storage, training, and annual testing. We estimate that as of 2011, an estimated 389,000 liters of aqueous film forming foam (AFFF) was maintained in Washington state by fire departments, civilian airports, military installations, and petroleum-related facilities. As part of the implementation of Chapter 70.75A⁹¹ Revised Code of Washington (RCW), Ecology is collecting additional information regarding current AFFF stocks.

Waste management related PFAS releases have been documented in other states, including air emissions, wastewater discharges, land application of industrial sludges, and landfill disposal. However, no data is available regarding releases of PFAS from waste management facilities in Washington state.

Household products that are sources of PFAS include:

- Cosmetics and personal care products.
- Treatments on textiles, upholstery, carpets, and leather.
- Coatings and floor finishes.
- Cleaning agents.
- Automobile and ski waxes.
- Nonstick cookware.

Occupational exposure to PFAS has been documented at retail stores where products containing PFAS are sold, and service industries that use products containing PFAS.

⁹¹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

Historic releases in Washington are estimated based on global estimates published in the literature.

3.0.2 Introduction

This appendix presents information about PFAS in Washington, and estimates historical PFAS releases to the environment.

As presented in Appendix 1: Chemistry, PFAS describes a class of more than 4,730 chemicals (Organisation for Economic Co-Operation and Development (OECD), 2018). This appendix provides information on the variety of consumer products that are known to contain PFAS. This appendix also addresses the main sources of PFAS in the environment resulting from manufacturing, consumer use, and product disposal. Past PFAS production, use, and disposal have resulted in PFAS contamination of soil, surface water, and groundwater (see Appendix 4: Fate and Transport).

As of April 2020, known PFAS contamination in the U.S. includes approximately 339 sites and 393 water systems in more than 40 states (Social Science Environmental Health Institute (SSEHRI), 2020; Walker, 2018). These compilations identify three impacted sites (City of Issaquah, Fairchild Air Force Base, Joint Base Lewis McChord) and three water systems (City of Dupont, Fort Lewis Cantonment, City of Issaquah) in Washington state. However, as discussed in Section 3.2.3 below, several other locations are being investigated.

Nationwide, groundwater contamination sites are impacted by firefighting foam use and training at military installations, civilian airports or fire stations, as well as use during a few fire events. Other activities reported to impact groundwater include manufacturing of PFAS and secondary manufacturing use of PFAS. Impacts to groundwater are also reported from waste disposal, landfill leachate, land application of industrial sludge, and discharges of wastewater to treatment facilities or septic systems—discussed in more detail in Section 3.4, Waste Management, below.

3.1 Manufacturing

3.1.1 Primary manufacturing

<u>Appendix 1: Chemistry, Section 1.3 Manufacturing</u>, addresses the methods used to manufacture PFAS compounds. Although raw and intermediate PFAS compounds have been, and continue to be, manufactured in the U.S. (<u>see Appendix 9: Regulations, Section 9.2.1 Environmental Protection Agency)</u>, we do not know of manufacture which was conducted in Washington state.

3.1.2 Secondary manufacturing

Release of PFAS can occur at manufacturing sites where PFAS are used as part of the manufacturing or industrial process. More than 3,000 PFAS may have been commercially used

since their inception (Naturvårdsverket, 2016, as cited by Banzhaf et al., 2017). EPA has identified 602 PFAS which are currently commercially active in the U.S. (EPA, 2019a) and EPA's Significant New Use Rules still allow use of certain long-chain PFAS in specific low-volume applications where substitute chemicals are limited or absent (for example, but not limited to, photographic imaging and semiconductor manufacturing) (See <u>Appendix 9: Regulations, Section 9.2.1</u>).

However, use of PFAS in secondary manufacturing operations is not typically reported to regulatory agencies. Its presence in air and aqueous industrial waste streams is not regulated with numeric standards under the Clean Air Act (CAA) or the Clean Water Act (CWA) respectively. However, certain solid wastes may qualify for reporting if they meet the threshold of state designation as dangerous waste (Section 3.4.4 below).

Fourteen investigated contaminated sites across the U.S. indicate PFAS releases from automobile, carpet, cable or wire, footwear, metal plating, paper, plastics, and textiles manufacturing (SSEHRI, 2018). PFAS releases and release mechanisms differ among the manufacturing processes. Appendix 4: Fate and Transport provides additional information about release mechanisms to the environment. PFAS releases during manufacturing operations could result from industrial air emissions, wastewater discharges, stormwater runoff, or waste disposal.

Examples of secondary manufacturing using PFAS include (Gaines, 2017; Interstate Technology Regulatory Council (ITRC), 2020a; SSEHRI, 2018; United Nations Environment Programme (UNEP), 2012, 2015a, 2015b, 2016):

- **Automotive**: Coatings on mechanical components, surface treatments for textiles, upholstery, carpets, and leather and automobile surface protectants and finishes.
- Aviation and aerospace: Coatings on mechanical components; hydraulic fluids.
- **Electroplating and etching**: Corrosion prevention; mechanical wear reduction; aesthetic enhancement; surfactant; wetting agent/fume suppressant for chrome, copper, nickel and tin electroplating; and postplating cleaner.
- Industrial surfactants, resins, molds, and plastics: Manufacture of plastics and fluoropolymers, rubber, and compression mold release coatings; plumbing fluxing agents; fluoroplastic coatings, composite resins, and flame retardants for polycarbonate.
- Medical products: Coatings on surgical products and medical fabrics.
- Oil and mining: Surfactants; evaporation inhibitors; solvents; fire suppression.
- Paper products and packaging: Surface coatings to repel grease and moisture. Uses
 include non-food paper packaging (for example, cardboard, carbonless forms,
 masking papers) and food-contact materials (for example, pizza boxes, fast food
 wrappers, microwave popcorn bags, baking papers, pet food bags).
- Semiconductor industry: Top anti-reflective coatings; bottom anti-reflective coatings; etchants, with other uses including surfactants, wetting agents, and photoacid generation.

- **Textiles and leather treatments**: Factory or consumer-applied coatings to repel water, oil, and stains. Examples include protective clothing and outerwear, umbrellas, tents, sails, architectural materials, carpets, footwear, and upholstery.
- Wire manufacturing: Coating; insulation.

Ecology collected information to estimate how many businesses in Washington state might be operating in a sector known to employ PFAS in the manufacturing process. The U.S. Census Bureau listed 577,445 businesses in Washington state in 2015 (U.S. Census, 2015). Table 18 lists the number of Washington businesses in selected North American Industry Classification System (NAICS) codes that include potential PFAS use (Infogroup, 2012). Figure 22 shows the general location of the businesses in Table 18 in each county of the state. There is no evidence that any of these operations use PFAS or have released PFAS during their operations. Also, PFAS use is not an indication that a release could have occurred.

Table 18: Secondary manufacturing in Washington.

NAICS code name	Count of businesses
All other plastics product manufacturing	241
Automobile manufacturing (plating activity)	13
Aviation and Aerospace	165
Carpet rug mills	13
Corrugated solid fiber box manufacturing	28
Electroplating, plating, polishing, anodizing	60
Leather hide tanning finishing	12
Medical products	249
Other fabricated wire product manufacturing	74
Oil (petroleum) and mining	128
Paper mills (except newsprint)	54
Paper bag coated treated paper manufacturing	69
Paperboard mills	10
Pulp mills	18
Semiconductors related devices manufacturing	33
Textile fabric finishing mills	46
Total of secondary manufacturing by NAICS code	1,213

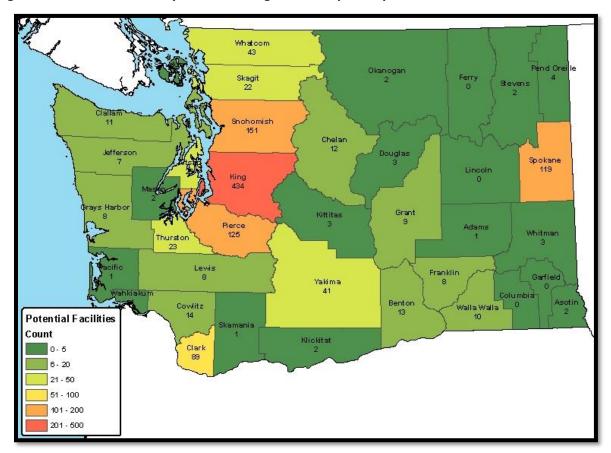


Figure 22. Count of secondary manufacturing facilities by county based on NAICS code.

EPA has also compiled the typical industry sectors which may produce or employ PFAS in manufacturing processes or components in its Enforcement and Compliance History (ECHO) database (EPA, 2020). The facility data presented within displays a subset of the universe of facilities subject to CAA, CWA, or Resource Conservation and Recovery Act (RCRA) regulations. EPA identified these industry sectors from literature reviews and other investigations.

Inclusion in the compilation, however, is not an indication that a business has ever used any PFAS component, or if it has, that any emissions have occurred. For Washington, ECHO identifies 1,095 businesses across 20 industry categories including airports and defense installations. The industry categories are similar to those listed in Table 18. Of the businesses identified, ECHO categorizes them against activity status: 508 are listed as active, 603 inactive, and seven with unknown activity status. Some facilities may be listed in multiple industry categories.

In addition to the manufacturing businesses in Table 18, use of PFAS-containing products like car polishes have been identified in one case as a source of groundwater contamination (Kernan, 2018). A variety of products containing polytetrafluoroethylene (PTFE) are marketed in the automobile washing and detailing industry, as well as to individual consumers. There are

more than 700 car washes listed in Washington state, however the extent of use of such PFAS products in the state is unknown. Car washes are not included in Table 18 or Figure 22.

3.2 Aqueous film forming foam

<u>Appendix 1: Chemistry, Section 1.4.4 Fire-fighting chemicals</u>, describes the chemical characteristics of PFAS used in the manufacture of AFFF.

AFFF, while not a large use category, is often used in uncontrolled circumstances with little or no barrier to direct environmental release. PFAS-containing firefighting foams have been implicated in many cases of groundwater contamination (Hu et al., 2016). Environmental releases of firefighting foam can occur during emergency response, mandatory firefighting equipment testing, emergency activation of fire suppression systems, and training exercises. These releases can occur at airports, refineries, bulk storage terminals, and other facilities handling large volumes of flammable liquid hydrocarbons (Heads of EPAs Australia and New Zealand (HEPA), 2018).

Typical facilities that could store AFFF for use at the facility are listed below. Listing of these categories does not imply that PFAS releases have occurred from such activities in Washington, with the exception of specific sites that are discussed below.

- Electrical power generation from coal, diesel, or gas.
- General chemical storage.
- Military installations, civilian airports, or fire departments.
- Mineral, oil, or gas extraction.
- Mining for coal or minerals.
- Petroleum production, exploration, storage, or refining.
- Production of aluminum, batteries, bitumen, brewing and distilling, coal works, dangerous goods, explosives, paints, polishes, or adhesives.

As identified in <u>Appendix 9: Regulations, Section 9.1.1 Washington state laws</u>, the Firefighting Agents and Equipment Toxic Chemical Use Law, Chapter <u>70.75A</u>⁹² RCW, now applies restrictions to the use of PFAS-containing firefighting foam and PFAS use in firefighting personal protective equipment.

The following subsections describe our estimates of AFFF held in the state. The data below was derived from the following sources:

- Information gathered directly by Ecology.
- Data estimated based on Ecology's regulatory requirements.
- Estimates based on a compilation of AFFF use by Darwin (2004).
- An update to Darwin's 2004 data completed in 2011.

⁹² https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

3.2.1 Fire departments and fire training

According to the Washington Fire Chiefs Association, there are approximately 350 public fire agencies within the state (Senter, 2019). Fire agencies are better known as fire departments, fire districts, regional fire authorities, and port fire departments. In addition to these public agencies, there also exists U.S. Department of Defense (DOD) and private or industrial firefighting forces. Each fire agency has one or more fire stations to serve their community. Fire agencies typically have training facilities located at a one of their facilities for in-service training. Fire agencies frequently create regionalized training centers where resources are pooled for multi-agency out-of-service training.

Use of AFFF for fire training has occurred both locally and at regional fire training sites across the state. The following lists some of the larger and frequently used regional training facilities, however we have not identified all fire training centers at this time:

- Big Bend Community College Air Rescue Firefighting Training, Moses Lake.
- City of Seattle Joint Training Facility, Seattle.
- Kitsap County Regional Training Center, Bremerton.
- Mark Noble Regional Fire Training Center, Olympia.
- North Bend Fire Training Academy, North Bend.
- Puget Sound Regional Fire Authority Fire Training Center.
- Spokane Regional Training Center, Spokane.
- Tacoma Fire Department Training Center, Tacoma.
- Yakima Fire Department Training Center, Yakima.

Other uses of AFFF include portable and wheeled fire extinguishers available for DOD, residential, commercial, and industrial users. Estimates of this type of fire extinguisher availability or use are currently not available.

In early 2018, the Washington Fire Chiefs Association polled its membership to begin to quantify impacts of the proposed legislation that would eliminate PFAS-containing AFFF from training exercises and curtail sales a year later. Feedback, while limited, indicated that most large fire agencies had moved away from using PFAS-containing AFFF. Other feedback related to the availability of reasonable alternatives and how to safely dispose of PFAS-containing AFFF. In response, the Washington Fire Chiefs Association held presentations on the subject at its annual conference and raised awareness through its newsletter and other various mediums.

In 2019, as part of the implementation of Chapter 70.75A⁹³ RCW (see Appendix 9: Regulations, Section 9.1.1 Washington state laws), Ecology surveyed municipal fire departments, fire districts, fire authorities, port authority fire departments, and fire training facilities about volumes of AFFF currently stored and interest in state-funded AFFF disposal options (Ecology, 2020a). As of February 2020, Ecology received 61 responses. A summary of responses is

⁹³ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

expected to be completed by fall 2020 (Smith, 2020). Ecology is currently identifying locations where these AFFF stocks can be safely disposed (Smith, 2020).

Pending the compilation of state-wide survey information, Ecology estimated fire agency storage of AFFF in Washington from the 2004 and 2011 Darwin reports. In 2004, Darwin estimated U.S. public fire departments (excluding airports) possessed 5.14 million liters of AFFF (all measurements are reported in metric units—Darwin reported 1,360,000 gallons of AFFF). This estimate included a 35% margin of error. Adjusting the national estimate in Darwin's study to Washington state (23% of the U.S. by population), the fire service possessed 118,577 liters of fluorinated and non-fluorinated firefighting foam in 2004 (the margin of error represents a range from 77,075 to 160,078 liters of foam).

Darwin's (2011) estimate took into account two factors. First, Darwin estimated total 2014 holdings by fire departments nationally to be lower, at 120,000 gallons. Second, Darwin estimated that by 2011, holdings were reduced by 50%. Applying these same reductions to Washington's 2004 estimate above results in 52,240 liters held in 2011.

3.2.2 Civilian airports

U.S. airports have been required to procure and use AFFF that meets the standards set by the Federal Aviation Administration (FAA), which currently requires the use of AFFF that meets military specifications (required to be fluorinated). In October 2018, the U.S. Congress passed legislation directing the FAA to allow airports to use non-fluorinated firefighting foam. The change is required to be implemented within three years using the latest version of the National Fire Protection Association (NFPA) 403 Standard for aircraft rescue and firefighting services at airports. NFPA 403 includes a fluorine-free synthetic foam option. There are fluorine-free foams that have been certified by GreenScreen. These products meet the bronze standard indicating that their ingredients are not found on any restricted substances lists (GreenScreen, 2020).

The FAA issues operating certificates to airports that comply with certain operational and safety standards. Current regulatory requirements related to firefighting at airports are found in 14 Code of Federal Regulations (CFR) Aeronautics and Space, Part 139: Certification of Airports, specifically 139.317: Aircraft rescue and firefighting: Equipment and agents. FAA provides guidance in Advisory Circulars. The most recent, on Aircraft Fire Extinguishing Agents (AC 150/5210-6D), states that foam concentrates must meet the performance test requirements of U.S. Military Specification (MIL-SPEC) MIL-F-24385F, which includes the requirement that the foam be fluorinated (FAA, 2004).

The eleven airports in Washington certified by the FAA to handle aircraft rescue and firefighting are listed below (FAA, 2018). In addition to airports listed below, there are 124 general aviation, reliever, and private airports and airstrips around the state (Washington State Department of Transportation (WSDOT), 2017).

WSDOT Aviation has reached out to several larger general aviation airports that do not have a requirement for AFFF under the FAA Part 139 requirement and have found that they do not possess any firefighting foam or personal protective equipment (PPE) that contained PFAS (Wright, 2019).

The amount of AFFF at airports is based on the amount carried on aircraft rescue and firefighting vehicles as well as the reserve available at the airport. Aircraft rescue and firefighting indexes indicate ascending order of aircraft length: A for aircraft less than 18 meters in length and up to E for aircraft longer than in 60 meters in length. Estimated quantities of AFFF stored at civilian airports based on each aircraft rescue and firefighting index are as follows (Darwin, 2004):

- Index A: 2,101 liters.
- Index B: 4,088 liters.
- Index C: 11,564 liters.
- Index E: 25,434 liters.

The following list identifies the index classification of larger civilian airports in Washington.

- Bellingham International, Bellingham, Index B.
- Boeing Field/King County International, Seattle, Index A.
- Grant County International, Moses Lake, Index A.
- Pangborn Memorial, Wenatchee, Index A.
- Pullman/Moscow Regional, Pullman, Index B.
- Seattle-Tacoma International, Seattle, Index E.
- Snohomish County (Paine Field), Everett, Index A.
- Spokane International, Spokane, Index C.
- Tri-Cities, Pasco, Index B.
- Walla Walla Regional, Walla Walla, Index A.
- Yakima Air Terminal (McAllister Field), Yakima, Index A.

Table 19 summarizes the volumes of AFFF held by civilian airports in Washington based on Darwin's (2004) assumptions.

Darwin re-estimated volumes of AFFF held by airports in 2004 based on volumes of 3M concentrate only, and also determined rate of usage drawing down the amounts held through 2011. Darwin's national 2004 estimate was only 37% of the 2011 estimate (i.e., a total of 26,824 gallons). Darwin determined that between 2004 and 2011, national civilian airport AFFF stocks were further reduced by approximately 85% (i.e., a total of 3,992 gallons). Darwin also received confirmation that SeaTac airport no longer held any AFFF with perfluorooctane sulfonic acid (PFOS). Table 20 provides a conservative update for Washington AFFF holdings based on an 85% reduction of 2004 volumes by index and excluding SeaTac. Based on these assumptions, civilian airports in Washington would have held 5,465 liters.

AFFF is also used in airplane hangars, according to NFPA standard 409 "Standard on Aircraft Hangars." Aircraft hangars require overhead foam sprinkling for the entire hangar if the floor area exceeds 1,858 square meters (m²): 11,356 liters of AFFF concentrate. Foam capacity increases for a hangar floor greater than 3,716 m²: 22,712 liters of AFFF concentrate. Darwin estimated hangar AFFF storage for airport index categories C and E at 43,721 and 289,205 liters per airport respectively (Darwin, 2004). These totals assumed AFFF storage in hangars were proportional to the FAA index estimates. Estimated AFFF stored in hangars in Washington in 2004 is summarized in Table 19. Darwin estimated that by 2011, volumes previously provided in 2004 were reduced by 37% overall. This same assumption is applied to AFFF maintained in hangars in Washington state in 2011, resulting in 123,183 liters, as shown in Table 20.

FAA regulations (14 CFR Part 139) establish the minimum aircraft firefighting capability for each index. AFFF quantities stored at FAA certified airports are estimated from Darwin (2004) using the estimates for A, B, C, and E aircraft rescue and firefighting indexes and for associated storage for hangars. There are additional users that maintain supplies of AFFF, such as airplane manufacturers, overnight shipping aircraft hangars, and fuel storage. Darwin (2004) provided quantities of AFFF stored by Boeing at 217,472 liters and FedEx at 378,541 liters at all U.S. locations.

PFAS-containing AFFF quantities stored at Washington certified airports are listed in Tables 19 and 20 for 2004 and 2011 respectively.

Table 19. 2004 estimated AFFF storage at certified airports and hangars (combined totals).

Airports in each FAA Index code	AFFF storage (liters)	AFFF hangar storage (liters)
A = 6 airports	12,605	-
B = 3 airports	12,265	-
C = 1 airport	11,564	43,721
E = 1 airport	25,434	289,205
TOTAL	61,868	332,926

Table 20. 2011 Estimated AFFF storage at certified airports and hangars (combined totals).

Airports in each FAA Index code	AFFF storage (liters)	AFFF hangar storage (liters)
A = 6 airports	1,891	-
B = 3 airports	1,840	-
C = 1 airport	1,735	16,177
E = 1 airports	0	107,006
TOTAL	5,465	123,183

Many airports have instituted best management practices associated with the testing of aircraft rescue and firefighting equipment required for use of AFFF (FAA, 2004; NFPA, 2014; Thalheimer et al., 2017). Certified airports must annually test the AFFF proportioning equipment to

maintain their Part 139 Certification. These tests require spraying the foam for 30 seconds and collecting a sample of the foam to verify that the proper concentration of AFFF is dispensed. AFFF best management practices recommend collection and proper disposal of the foam and any impacted soil. Recent FAA guidance allows testing to be performed in a closed system, some airports may opt to use this system for future annual tests (FAA. 2019). Fire response training can be conducted at the airport or at other fire training locations.

3.2.3 Defense installations

As required by law, DOD installations in Washington no longer use AFFF containing PFAS in training operations (Shirley, 2018). AFFF storage and use at DOD sites includes ships, shore facilities, and firefighting vehicles (Darwin, 2004). We assumed that nationally there are 242 Navy installations, 245 Army installations, 384 Air Force installations, and 400 Coast Guard installations. There are 19 active military installations in Washington state, including ten operated by the U.S. Coast Guard. Ecology calculated Washington AFFF volumes by proportionally reducing Darwin's national data against the number of installations in Washington. PFAS-containing AFFF quantities stored at active Washington military installations estimated from Darwin (2004) are shown in Table 21.

For his 2011 estimate, Darwin focused on 3M AFFF inventories. On this basis, he first revised the national 2004 holdings from 2,836,497 gallons to 2,080,000 gallons. Darwin further estimated various drawdown percentages between 2004 and 2011 for each of the defense branches and for specific types of uses within a branch. Ecology updated its 2004 estimate using national volume totals by installation type determined by Darwin for 2011, proportioned against the number of installations in Washington. These are also presented in Table 21.

Table 21. Military AFFF storage (c	ombined totals) in 2004 and 2011.

Military installations	2004 estimated AFFF concentrate stored (liters)	2011 estimated AFFF concentrate stored (liters)
4 Navy	78,184	19,623
3 Army	3,585	3,121
2 Air Force	26,173	12,442
10 Coast Guard	13,438	7,823
TOTAL	121,380	43,008

The DOD and Department of the Navy (DON) continue to inventory fire and crash training sites at U.S. installations. The military is assessing the risk of groundwater contamination from firefighting foam at many of its locations including those in Washington state (DOD, 2014, 2018, 2019a, 2019b, 2019c; DON, 2016a). The following is a partial listing of defense installation sites in Washington state where PFAS use or releases may have occurred:

- Four Lakes Communications Air Guard Station (closed), Cheney.
- Fairchild Air Force Base, Spokane (DOD, 2019c).
- Joint Base Lewis-McChord, Tacoma (DOD, 2018).

- Yakima Training Center, Yakima (DOD, 2014).
- Naval Base Kitsap (DON, 2020a, 2020b).
- Naval Air Station Whidbey (DON, 2018, 2019).

<u>Appendix 7: Health, Section 7.4 Known areas of PFAS contamination in drinking water aquifers in Washington state</u>, provides additional information regarding impacts of AFFF releases.

3.2.4 Petroleum storage and transport

Petroleum is refined, stored, and transported from and around Washington state. Petroleum products stored at gas stations are not included in this discussion. Transport and storage of fuel from railcar, tanker, pipeline, or refinery has the potential for fire or explosion, requiring the availability and use of fire suppression. Fire suppression systems at these facilities may include PFAS-containing AFFF.

Ecology regulates equipment and oil transfer, storage, and handling at 121 facilities to ensure protection of environmental and public health. There are three facility types, shown in Figure 23. Each facility has different types of requirements, depending on their classification, but all are required to have some type of spill prevention plan. Regulated facilities are trained to prevent, prepare for, and respond to spills when they occur. Ecology does not track the firefighting foam stored at these facilities. Darwin (2004) estimated 59,052 liters (15,600 gallons) of AFFF concentrate per refinery in the U.S. For the five refineries in Washington, that amounts to 295,262 liters of AFFF. The following brands of AFFF have been reported to be stored or used at these refineries:

- 3M Light Water 3X6 AR-AFFF
- Aer-O-Foam XL-3
- Chemgard 3 percent AR-AFFF
- FireAde 2000
- National Foam (Universal Plus 3/6 percent AR-AFFF)
- Thunderstorm 1 X 6, 3 X 6 and 1 X 3 AR-AFFF Ansul/Williams
- Thunderstorm FC601A

Darwin revised the 2004 estimate to 4,724 liters (1,248 gallons) per refinery based on consideration of 3M AFFF holdings in 2004 and a consumption of 86% between the years 2004 and 2011. This resulted in an estimated total of 23,621 liters for the five refineries in Washington state.

Mobile facilities transporting petroleum product into Puget Sound are required by federal shipping regulations to maintain a supply of fire suppressant on the tanker (46 CFR). That volume of foam liquid must be sufficient to provide a minimum of 20 minutes of flow through nozzles across the cargo tank deck. Darwin (2004) estimated 3,785 liters (1,000 gallons) of AFFF are maintained per oil tanker and 189 liters for other merchant ships. Darwin further estimated that holdings associated with mobile facility uses would have been reduced by half from 2004 – 2011. International shipping regulations require fire extinguishing systems adequate for the fire

hazard that may exist, but fire extinguishing systems using perfluorocarbons are prohibited (International Maritime Organization, 2007).

In addition to refineries, other petroleum facilities include blending facilities, tank farms, loading and fueling terminals, and other flammable liquid storage. Fire protection at these facilities include AFFF systems constructed according to NFPA standards. AFFF storage at these facility types, in Table 22, are estimates. Ecology regulates these facilities in four categories:

- Class 1 facilities are large, fixed shore-side facilities such as refineries and refueling terminals. This definition includes facilities that transfer to or from tank vessels and pipelines.
- Class 2 facilities are mobile facilities, such as tanker trucks and portable tanks.
- Class 3 facilities are small tank farms and terminals that transfer oil to non-recreational vessels that have a fuel capacity of 39,746 liters (10,500 gallons) or more. This definition does not include facilities that transfer to tank vessels and pipelines, as they are Class 1 facilities.
- **Class 4** facilities are marinas or other small fueling facilities that transfer oil to non-recreational vessels with a total oil capacity of less than 39,746 liters.

In line with Darwin's estimates, we also assumed that from 2004 – 2011, stocks of these AFFF holdings were reduced by half. Tables 22 and 23 summarize AFFF volumes estimated in Washington state for the petroleum refinery sector for years 2004 and 2011 respectively.

Table 22. 2004 AFFF storage at petroleum related facilities.

Description	Count of facilities	AFFF/facility (liters)	Estimated AFFF (liters)
Refineries	5	59,052	295,262
Large refueling terminal, pipeline	20	7,570	151,400
Mobile facility	24	3,785	90,840
Transfer >10,500 gal capacity	5	3,785	18,925
Transfer <10,500 gal capacity	67	1,892	126,764
TOTAL	121		683,191

Table 23. 2011 AFFF storage at petroleum related facilities.

Description	Count of facilities	AFFF/facility (liters)	Estimated AFFF (liters)
Refineries	5	4,724	23,621
Large refueling terminal, pipeline	20	3,785	75,700
Mobile facility	24	1.893	45,420
Transfer >10,500 gal capacity	5	1,893	9,463
Transfer <10,500 gal capacity	67	946	63,382
TOTAL	121		217,585

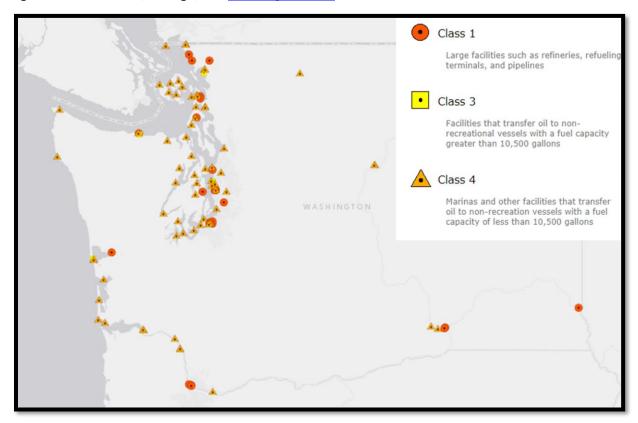


Figure 23. Oil transfer, storage, and handling facilities.94

Oil spill response can also involve the use of products which may contain PFAS, for example AFFF stored in caches. Oil spill response resources are tracked on the Worldwide Response Resource List (WWRL). Records indicate BNSF holds 2,082 liters (550 gallons) AFFF at each of its Pasco, Seattle, and Vancouver cache locations, for a total of 6,246 liters (Ecology, 2020).

Ecology funds oil spill response equipment located around the state (including AFFF) and provides training to local responders on how to safely and effectively deploy the equipment. Cached equipment has been used a number of times since deployment, and has effectively limited the spreading of and environmental damage from oil spills, and reduced the time and costs associated with oil spill cleanup. AFFF covered under this grant funding is limited to non-fluorinated products.

3.2.5 Tunnels

NFPA standard 502 provides fire protection and fire safety requirements for road tunnels, bridges, and other limited access highways (NFPA, 2011). In Seattle, tunnels using a deluge foam fire suppression system are the I-90 Mercer Island, I-90 Mt. Baker, and the I-5 Convention Center (Cox, 2019). Other Seattle tunnels use a non-PFAS based fixed water firefighting system: Battery Street, downtown Seattle transit for bus and train, and SR99 Replacement Tunnel.

⁹⁴ https://fortress.wa.gov/ecy/coastalatlas/storymaps/spills/spills sm.html?&Tab=nt3

Table 24. Road tunnels with fixed foam firefighting systems in Seattle.

Tunnel	Route	Length (meters)	Lanes	Estimate of AFFF storage (liters)
Mercer Island	I-90	914	8	48,510
Mt Baker	I-90	1067	8	28,334
Convention Center	I-5	167	12	11,735
TOTAL				88,579

3.2.6 Summary of AFFF quantities

Table 25 summarizes the estimates firefighting foam quantities in Washington state in 2004 and 2011. The table also estimates average annual use over the seven years.

Table 25. 2004 and 2011 estimated AFFF quantities in Washington state.

AFFF use sector	2004 (liters)	2011 (liters)	Estimated annual use
Fire departments	118,577	52,240	8% or 9,477 liters
Fire extinguishers*	Not able to estimate*	Not able to estimate*	Not able to estimate*
Civilian airports	61,867	5,465	13% or 8,057 liters
Airport hangars	332,926	123,183	9% or 29,963 liters
U.S. Military installations	121,380	43,008	9% or 11,196 liters
Petroleum refineries	295,262	23,621	13% or 38,806 liters
Other petroleum facilities	388,003	193,965	7% or 27,720 liters
Merchant ships/Oil cargo tankers*	189 to 3,785 per vessel*	189 to 3,785 per vessel*	Not able to estimate*
Oil response storage	76,011**	76,011**	Not able to estimate*
Seattle tunnels	88,579**	88,579**	Not able to estimate*
TOTAL storage	1,482,605	606,702	(11%) or 125,219 liters

Notes:

- * = Not included in total.
- ** = 2004 and 2011 data are not available; data represents 2019 2020 storage.

3.2.7 Spill reports

When oil or other hazardous substances are spilled, a report must be submitted to Ecology. Since 2007, Ecology has maintained the Emergency Reporting Tracking System (ERTS) for these reports. Reports entered into that system that refer to releases of firefighting foam are summarized in Table 26. Most of these reports were related to activities that occurred on or

near water, or where firefighting foam entered a waterway. These voluntary reports refer to fuel, water, and foam but do not specify if the material released contains PFAS. These reports are shared with local agencies and other response personnel. Information in these reports is not independently verified.

Table 26. Firefighting release incidents voluntarily reported to Ecology's ERTS.

Year	Number of reported incidents	Released fuel, water, AFFF (liters)
2007	1	76
2009	3	30
2010	3	15
2011	4	1,908
2012	2	34,163
2013	3	2,468
2014	2	15
2015	1	38
2016	9	1,177,535*
TOTAL	28	1,216,248

Note: * = One incident in August 2016 reported the use of 1,173,477 liters (310,000 gallons) of water with firefighting foam at an industrial facility.

3.3 Consumer products

3.3.1 PFAS in children's products

As identified in <u>Appendix 9: Regulations, Section 9.1.1 Washington state laws</u>, the Children's Safe Products Act (CSPA—Chapter <u>70.240</u>⁹⁵ RCW) requires manufacturers to annually report the presence of PFOS or perfluorooctanoic acid (PFOA) in children's products sold in Washington state. Manufacturer reports are <u>available online</u>.⁹⁶

A summary of the PFOS manufacturer data available through 2019 is provided in Table 27. For all products, PFOS was reported to be present at concentrations less than 100 parts per million, except for the artists accessories report from 2014, which reported PFOS at 100 to 500 parts per million. PFOA was only reported once in 2019 in the Belts/Braces/Cummerbunds product category, present at less than 100 ppm, with a stain prevention function.

⁹⁵ https://app.leg.wa.gov/RCW/default.aspx?cite=70.240

⁹⁶ https://hpcds.theic2.org/Search

Table 27. Reports of PFOS in children's products, at concentrations below 100 parts per million unless noted.

Product category	2014	2016	2017	2018	2019	Chemical function
Artists Accessories (PFOS reported at 100 to 500 parts per million)	1					UV stabilizer
Baby Feeding – Bibs	1					Contaminant
Belts/Braces/Cummerbunds					1	Protective Coating
Blankets/Throws (Non Powered)	1					Contaminant
Board Games/Cards/Puzzles Variety Packs	1					Contaminant
Dresses	1	1				Contaminant
Full Body Wear Variety Packs		1				Manufacturing additive
Indoor Footwear – Fully Enclosed Uppers	1					Contaminant
Jackets/Blazers/Cardigans/Waistcoats	1	1	1			Colorant; Contaminant
Overalls/Bodysuits	1	1	14			Colorant; Flame retardant; Contaminant
Pants/Briefs/Undershorts	1		14			Flame retardant; Contaminant
Pantyhose/Stockings	1					Contaminant
Shirts/Blouses/Polo Shirts/T–shirts	1		22			Colorant; Flame retardant; Contaminant
Shoes – General Purpose	1					Contaminant
Skirts	1		1			Flame retardant; Contaminant
Sleepwear Variety Packs	1					Contaminant
Socks	1					Contaminant
Sportswear – Full Body Wear	1	4				Colorant; Contaminant
Sportswear – Lower Body Wear	1	1	2			Colorant; Flame retardant; Contaminant
Sportswear – Upper Body Wear	1	5	4			Colorant; Flame retardant; Waterproofing
Sweaters/Pullovers	1					Contaminant
Trousers/Shorts	1					Contaminant
Upper Body Wear/Tops Variety Packs		1				Colorant
Total reports	20	15	58	0	1	

3.3.2 PFAS in a typical home

PFAS exposure in the home occurs during product use and exposure to house dust containing PFAS. The greatest portion of the chronic exposure to PFAS for the general public, specifically to PFOS and PFOA, results from the intake of contaminated drinking water and foods—more discussion is provided in Appendix 7: Health, Sections 7.3.1 Drinking water and 7.3.2 Food (Trudel et al., 2008). Other sources of exposure could occur from PFAS-containing products in the home and in some occupations (Guo et al., 2009, ITRC, 2020a). Studies of indoor air and house dust indicate exposure to PFAS from consumer products in the home such as carpet care liquids, nonstick cookware, packaged fast food, and waterproof clothing (see Appendix 7: Health, Section 7.3.3 Consumer products).

In a study published in 2009, EPA evaluated 116 products available in the typical home and tested each product for perfluorocarboxylic acid (PFCA) (Guo et al., 2009). The main goal of that study was to identify and rank potentially important indoor sources based on PFCA content in articles of commerce. In the following tables, the Guo et al. (2009) study data is provided and supplemented with data from more recent consumer products PFAS studies.

The EPA study estimated quantities of product categories present in a typical American home (Guo et al., 2009). For example, in the EPA study, a typical home was assumed to contain 150 m² of PFAS-treated carpet and 50 m² of PFAS-treated textiles and upholstery.

- Treated carpet: 60% of the U.S. home floor area of 250 m² is carpeted.
- Textile and upholstery of 50 m²: 10 20 m² of fabric for an upholstered chair or sofa and 2 3 m² of fabric for a jacket, shirt, or pants.

Carpeting and upholstery involve large treated areas and stain-resistance treatment is a frequent specification among institutional purchasers (Department of Toxic Substances Control, California (DTSC), 2017). Textile-related products that use fluorinated applications include: home furnishings, outer garments, umbrellas, bags, sails, tents, parasols, car seats, covers, leather articles, and shoes.

Investigations indicate a variety of PFAS are present in a wide range of cosmetics, including sunscreen, foundations, concealers, hair spray, eye liners, creams, lotions, and powders. The results varied widely across product types and brands, with highest measured PFAS concentrations in sunscreen and foundation (Danish Environmental Protection Agency (DEPA), 2018). Examples of fluorinated ingredients in cosmetic products include: per/polyfluorinated acrylate polymers, naphthalenes, alkanes/alkenes, alcohols, siloxanes, silanes, sulfonamides, ethers, esters, phosphate esters, acrylates, and acids. According to the European Commission's database on cosmetic ingredients, these substances are used in cosmetic products as emulsifiers, antistatics, stabilizers, surfactants, film formers, viscosity regulators, and solvents (Schultes, 2018).

Using the process developed by EPA, recent product testing study data are added to the 2009 data (Guo et al., 2009; Fujii, 2013; Herzke et al., 2012; Kotthoff, 2015; Liu et al., 2015). Tables 28

and 29 list the top ten products for the sum of PFCA and FTOH/fluorotelomer sulfonate (FTS). Supplement 1 to this appendix provides estimates for more product testing data. The amount of PFAS in the typical home from each product will not directly correlate with exposure. Some PFAS such as fluoropolymers in non-stick cookware have been shown to be stable (see Appendix 1: Chemistry, Section 1.1.4 Polymeric PFAS).

Table 28. Estimated PFCA in consumer products in a typical home.

Category name	Concentration of PFCAs in product	Typical quantity of product used	Total PFCA in typical home (microgram µg)	Reference
Pre-treated carpeting	484 μg/ m²	150 m ²	72,600	Guo et al., 2009
Treated home textile and upholstery	346 μg/ m ²	50 m ²	17,300	Herzke et al., 2012
Waterproofing agents	29,889 μg/Liter (L)	0.5 L	14,945	Herzke et al., 2012
Pre-treated carpeting	57.2 μg/kilogram (kg)	50 kg	2,860	Kotthoff, 2015
Food contact material (paper)	2,859.9 μg/kg	1 kg	2,860	Kotthoff, 2015
Treated floor waxes and stone/wood sealants	2,430 μg/kg	1 kg	2,430	Guo et al., 2009
Sunscreen	19,000 μg/kg	0.1 kg	1,900	Fujii, 2013
Treated home textile and upholstery	336 μg/kg	5 kg	1,680	Guo et al., 2009
Non-stick cookware	1,234.74 μg/kg	1 kg	1,235	Herzke et al., 2012
Household carpet/fabric-care liquids and foams	953 μg/kg	1 kg	953	Guo et al., 2009
Dental floss and plaque removers	31.3 μg/kg	0.005 kg	0.2	Guo et al., 2009

Table 29. Estimated FTOH or FTS in consumer products in a typical home.

Category	Concentration of FTOH/FTS in product	Typical quantity of product used	Total FTOH/FTS in typical home (microgram μg)	Reference
Cleaning agents	667,700 μg/kg	1 kg	667,700	Kotthoff, 2015
Treated floor waxes and stone/wood sealants	423,000 μg/kg	1 kg	423,000	Liu et al., 2015
Waterproofing agents	464,774 μg/L	0.5 L	232,387	Herzke et al., 2012

Category	Concentration of FTOH/FTS in product	Typical quantity of product used	Total FTOH/FTS in typical home (microgram μg)	Reference
Treated home textile and upholstery	42,900 μg/kg	5 kg	214,500	Liu et al., 2015
Carpet	4,010 μg/kg	50 kg	200,500	Liu et al., 2015
Impregnating sprays (waterproofing)	1,857,300 μg/kg	0.1 kg	185,730	Kotthoff, 2015
Treated home textile and upholstery	757 μg/ m ²	50 m ²	37,850	Herzke et al., 2012
Carpet samples	73.5 μg/kg	50 kg	3,675	Kotthoff, 2015
Membranes for apparel	1,590 μg/kg	1 kg	1,590	Liu et al., 2015
Treated apparel	464 μg/kg	2 kg	928	Liu et al., 2015

Based on the method used by Guo et al. (2009) and Liu (2015), sources in a typical home include:

- PFCA from carpet, carpet care products, textiles and upholstery, and floor waxes and polishes.
- Fluorotelomer alcohols (FTOH) and fluorotelomer sulphonates (FTS) from cleaners, carpet-care products, waterproofing spray, textiles, floor waxes and polishes, and carpet.

3.3.3 Consumer product priorities

We have identified several consumer product categories that merit additional consideration based on their contribution to PFAS in homes, potential human exposure (see <u>Appendix 7: Health, Section 7.3 Sources and pathways for human exposure</u>), and environmental release pathways (see <u>Appendix 4: Fate and Transport</u>).

Table 30. Consumer products that have the potential to contribute to human and environmental exposures of PFAS in Washington state.

Product	Contribution to human exposure	Contribution to environmental exposure
Stain resistant	PFAS in carpet is associated with indoor air	An estimated 14,300 metric tons of
carpet	concentrations of PFAS (Fraser, 2012) and	PFAS from carpet end up in
	PFAS biomarkers in children (Harris, 2017).	Washington landfills annually.
	Carpet can cover a large portion of home and	
	commercial floor space. Since young children	
	spend more time on or near the floor, they	
	are particularly vulnerable to PFAS exposure	
	from carpet (Tian, 2016; Trudel, 2012).	

Product	Contribution to human exposure	Contribution to environmental exposure
Carpet treatments	Frequent carpet treatments were associated with elevated house dust and blood concentrations of PFHxS, PFOA, and PFOS in a case study (Beesoon, 2012). Once treated, there is similar exposure potential as pretreated stain-resistant carpet. Since application is done in residential settings, the use of protective equipment and ventilation may be insufficient. PFAS are semi-volatile and can be inhaled. Exposures to PFNA is higher in people when they use wax, polish, or water-resistant materials (Lee, 2017).	An estimated 14,300 metric tons of PFAS from carpet end up in Washington landfills annually. Most of this comes from pretreated carpet (50 – 90%), however the use of carpet treatments also contributes to this burden.
Waterproofing sprays	Waterproofing sprays can have high PFAS concentrations (1,857,300 microgram/kg) (Kotthoff, 2015). Since application is done in residential settings, the use of protective equipment and ventilation may be insufficient. PFAS are semi-volatile and can be inhaled. They also penetrate the skin. Exposures to PFNA is higher in people when they use wax, polish, or water-resistant materials (Lee, 2017).	An estimated 2,066 metric tons of PFAS from textiles end up in Washington Landfills annually. PFAS used in waterproofing textiles can be released during the laundering process (CEC, 2017). They are found in WWTP influent and are difficult to remove, resulting in direct environmental releases (Pan, 2016).
Furniture	PFAS can be released from furniture over time and accumulate house dust and be inhaled or ingested by babies and children. Women with treated carpets or furniture in their homes had higher concentrations of some PFAS in their bodies (Boronow et al., 2019).	59,842 metric tons of furniture are disposed of in Washington state each year. If furniture (on average) is approximately 2.4 mg/kg PFOS, 0.17 metric tons of PFOS are disposed of each year (KEMI, 2015).
Waterproof textiles	PFAS in textiles is associated with indoor air and dust concentrations (Wu, 2015). Children who wear waterproof clothing more frequently have higher concentrations of PFOS and PFNA in their serum (Clara, 2008).	An estimated 2,066 metric tons of PFAS from textiles end up in Washington landfills annually. PFAS can also be released from washable textiles during the laundering process (CEC, 2017). They are found in WWTP influent and are difficult to remove, resulting in direct environmental releases (Pan, 2016).
Cosmetics	PFAS are found in some sunscreen, concealers, hair spray, lotions, shampoo, creams, and powders (DEPA, 2018). These products are applied directly to the skin and body. PFOA can be dermally absorbed, leading to increased serum concentrations (Franko, 2012).	PFAS in rinse-off products can be washed down the drain. PFAS are found in WWTP influent and are difficult to remove, resulting in direct environmental releases (Pan, 2016).

3.3.4 Service and retail settings

<u>Appendix 7: Health, Section 7.2.2 Populations with elevated PFAS exposure</u>, addresses potential occupational PFAS exposure routes. With the exception of firefighter exposure, a primary occupational exposure route in Washington state is exposure to products containing PFAS in retail- and service-oriented occupations.

High levels of PFAS have been reported in some occupational settings, including retail stores where products containing PFAS are sold, and service industries that use products containing PFAS, for example stores selling outdoor equipment, furniture shops, and carpet shops (Langer et al., 2010; Schlummer et. al., 2013). We have estimated that approximately 10,400 and 6,500 retail trade workers are employed in home furnishing and sporting goods stores in Washington respectively (Washington State Employment Security Department (ESD), 2017a).

An estimated 255,515 Washington workers could be exposed at work when using PFAS-containing products (ESD, 2019). The estimated number of workers in specific occupations are listed below.

Automotive workers that could use PFAS-containing car polishes or products used on the textiles in the car:

- Automotive & Watercraft Service Attendants: 2,446
- Automotive Body & Related Repairers: 2,545
- Automotive Glass Installers & Repairers: 559
- Automotive Service Technicians & Mechanics: 13,421
- Cleaners of Vehicles & Equipment: 8,116

Carpet and furniture workers that could use PFAS-containing oil, stain, and water repellents:

- Cabinetmakers & Bench Carpenters: 2,330
- Carpet Installers: 1,204
- Floor Layers, Except Carpet, Wood, & Hard Tiles: 291
- Floor Sanders & Finishers: 149
- Upholsterers: 380
- Furniture Finishers: 518

Textile workers that could use PFAS-containing oil, stain, and water repellents:

- Fashion Designers: 495
- Shoe & Leather Workers & Repairers: 68 (2017 data; no data reported for June 2018)
- Textile Cutting Machine Setters, Operators, & Tenders: 114
- Textile Wind/Twist/Draw-Out Machine Setters, Operators, & Tenders: 226
- Fabric & Apparel Patternmakers: 56
- Textile, Apparel, & Furnishings Workers, All Other: 128

Food service workers that could use PFAS-containing food packaging or paper:

- Food Service Managers: 2,297
- Food Preparation & Serving Worker Supervisors: 21,030
- Food Preparation Workers: 20,088
- Combined Food Preparation & Serving Workers, Inc. Fast Food: 80,587
- Counter Attendants, Cafeteria/Concession, & Coffee Shop: 13,766
- Food Servers, Non-restaurant: 4,828
- Dining Room & Cafeteria Attendants & Bartender Helpers: 9,429
- Food Preparation & Serving Related Workers, All Other: 1,646
- Paper Goods Machine Setters, Operators, & Tenders: 2,081

Other workers that could use PFAS-containing cleaning products or cosmetics:

- Janitors/Cleaners, Except Maids & Housekeeping: 45,378
- Maids & Housekeeping Cleaners: 17,617
- Housekeeping & Janitorial Worker Supervisors: 2,421
- Skincare Specialists (cosmetics): 1,301

Other occupations may use PFAS treated clothing or fabric, including but not limited to:

- Medical field and medical emergency responders
- Firefighters
- Retail

3.4 Waste management

3.4.1 Manufacturing waste

Release of PFAS has been shown to occur at manufacturing sites where PFAS are used as part of the industrial process. Approximately 60 contaminated sites across the U.S. are linked to PFAS releases from automobile, carpet, cable or wire, metal plating, paper, plastics, and textiles manufacturing (SSEHRI, 2020). PFAS releases and release mechanisms differ among the manufacturing processes. PFAS releases could result from air emissions, wastewater discharges, stormwater runoff, or waste disposal. SSHERI has not identified any manufacturing-related contaminated sites in Washington. Neither Ecology nor Health have conducted any manufacturing industry surveys in Washington to determine whether PFAS may have been used, and if so, whether discharges of PFAS-contaminated wastes may have occurred.

3.4.2 Wastewater treatment plants

Wastewater is the water "waste" that results from domestic uses, such as restroom use, bathing, food preparation, and laundry, or industrial uses such as, but not limited to, manufacturing, mining, and commercial businesses. Some wastewaters are treated on site (for example single family septic systems or industries that treat their own wastewater prior to disposal to the environment), but large volumes of waste are treated by publicly owned wastewater treatment plants (WWTPs).

Wastewater treatment standards

Different contaminants enter wastewater depending on how and where water is used. Wastewater that contains pollutants (for example, chemicals or organic matter) must be treated before it can be released back into the water environment.

Effluent limits for industries are based on technology requirements and water quality-based standards. Neither federal nor state treatment requirements address criteria for PFAS in industrial wastewater discharges. Effluent limits for publicly owned WWTPs are also largely based on meeting the technology and water quality-based requirements. In cases where effluent is discharged to ground, it is regulated to meet the Washington Groundwater Quality Standards (Chapter 173-2009 Washington Administrative Code (WAC)). Effluent discharged to surface waters must meet the state's Surface Water Quality Standards (Chapter 173-201A9 WAC). Industrial users who discharge to publicly owned WWTPs must comply with national and state pre-treatment requirements, however there are neither state nor federal pre-treatment requirements addressing PFAS in wastewater effluents. In Washington, WWTP effluent can be discharged to surface waters or to ground, and all WWTPs must meet a high level of pollutant removal technology under the requirements of All Known Available and Reasonable Technology (AKART), WAC 173-218-030.99

At this time, EPA has not developed numeric nationally recommended surface water quality criteria for PFAS. States generally adopt EPA's nationally recommended water quality criteria into state surface water quality standards instead of developing state-specific criteria, largely because of the high cost of criteria development and because of the lack of available and adequate toxicological data to support the equations used to calculate criteria.

In the case of PFAS, some states have adopted, or are developing, surface water quality criteria for some PFAS. For example, Michigan adopted a surface water criterion of 12 ng/L for PFOS (Michigan Department of Environmental Quality (MIDEQ), 2019). Washington has not adopted water quality-based numeric standards and regulations for PFAS in effluents. In addition, EPA-approved methods for monitoring compliance with effluent limits for PFAS have not yet been developed and adopted by EPA.

Routine wastewater influent and effluent monitoring is required by federal and state regulations and laws, and small and large systems have different monitoring requirements. The specific pollutants that are generally sampled for under the CWA (for large discharges that reach surface waters) include priority toxic pollutants (126 specific substances), conventional pollutants (five-day biochemical oxygen demand, total suspended solids, pH, fecal coliform, and oil and grease), and non-conventional pollutants (such as ammonia, chlorine, color, iron, and total phenols). State regulations frequently include additional pollutants that must be

⁹⁷ https://apps.leg.wa.gov/wac/default.aspx?cite=173-200

⁹⁸ https://apps.leg.wa.gov/wac/default.aspx?cite=173-201A

⁹⁹ https://apps.leg.wa.gov/wac/default.aspx?cite=173-218-030

addressed (e.g., temperature). PFAS are not included in the lists of pollutants that require monitoring under either the federal or state CWAs. PFAS are also not included in the Washington Groundwater Standards, which sets groundwater quality based requirements for discharges to land.

Industrial wastewater treatment

As identified above, many industries treat their wastewater under state and federal regulatory programs before discharging it. Typically, when industrial wastewater treatment occurs on site, the waste streams produced include both liquids (effluent) and solids (sludge). Treated effluent is typically discharged to surface water at specifically permitted locations (point discharge) or to a WWTP, and on rare occasions to ground. Any PFAS in the effluent, if not removed by, or degraded during, the treatment process enters the environment at the discharge point of the treated effluent. Industries that discharge to publicly owned WWTPs must comply with federal and state pre-treatment requirements.

Publicly owned WWTPs

There are approximately 15,500 operational public WWTPs in the U.S., and approximately 72% of these are considered small systems (serving a population of 10,000 or fewer people and an average daily wastewater flow of less than one million gallons per day) (EPA, 2019). According to Ecology's Water Quality Permitting and Reporting System, there are more than 600 WWTPs in Washington.

PFAS are found in numerous products that contribute to domestic and non-domestic waste streams, as well as in contaminated drinking water supplies. Because PFAS sources are so pervasive, the wastewaters that arrive at WWTPs contain these compounds. As identified above, in addition to sewage, publicly owned WWTPs in many cases accept wastewater from local industries and businesses that contain higher levels of toxic compounds than found in domestic waste. Publicly owned treatment systems that receive wastewater have traditionally been designed and constructed to meet technological requirements to remove solids from the influent (primary treatment) and to further remove some conventional pollutants (secondary treatment) to meet a "technology-based" standard of effluent quality.

These systems did not incorporate specific design considerations for PFAS or other toxics removal. Beyond the technology-based treatment requirements, water quality based toxics regulation is an ongoing process, as WWTPs and others work to reduce levels of toxics entering WWTPs, and as WWTPs work to optimize operations of current infrastructure and to evaluate additional technologies and approaches to reduce toxics. Significant challenges exist in this effort because of the extremely low concentrations that are being targeted for many pollutants, as well as the lack of known technology to assess these concentrations. Because PFAS is a relatively newly identified pollutant, and is gaining attention at the state and national level, some states have begun to sample WWTP effluent for PFAS.

Some WWTP effluents in Washington have been sampled for PFAS compounds as parts of special studies. <u>Appendix 5: Environmental Occurrence, Section 5.1.5 WWTP effluent</u>, documents such measurements conducted in Washington. Where PFAS compounds have been sampled for, they have been found at levels similar to WWTPs in other areas of the U.S., and at lower concentrations than plants treating wastewater containing AFFF.

When PFAS enter wastewater treatment plants there is a mix of long- and short-chain compounds, as well as a large number of precursor compounds that can form perfluorooctanesulfonic acids (PFAA). This mixture is subject to bacterial degradation during the treatment process (see Appendix 4: Fate and Transport, Section 4.1 Non-polymer PFAS). Prior to the development of improved analytical methods used to identify this phenomenon (see Appendix 2: Analytical Methods), it appeared as though WWTPs were increasing the mass balance of PFAS during the treatment process. However, through a better understanding of a fuller list of measurable PFAS, it has since been confirmed that degradation and transformation of influent PFAS to different individual PFAS are the cause of greater total PFAS concentrations in WWTP effluent. This is especially true with PFAAs such as PFOS and PFOA.

Studies show that conventional activated sludge treatment does not effectively remove most PFAS, though some specialized treatments can remove a large percentage of longer chained compounds (Eschauzier, Beerendonk, Scholte-Veenendaal & De Voogt, 2012; Pan, Liu & Ying, 2016).

Solids that are part of the influent wastewater and also generated during secondary treatment of wastewater are largely removed prior to discharge of the treated effluent. These solids, called sludge, are either treated as waste for disposal or treated as a resource. Sludge from many domestic WWTPs is transformed into the product termed "biosolids." Biosolids are used in agriculture to improve the quality of agricultural lands for crop production. Application of biosolids is regulated under state and federal regulatory programs. PFAS in biosolids is discussed in detail in <u>Appendix 8: Biosolids</u>.

Onsite wastewater treatment systems

Onsite wastewater treatment systems (commonly called septic systems) can be sources that release pollutants, including chemical contaminants, to groundwater. Leachate from septic systems can contaminate domestic drinking water wells in areas with high septic system density. Incomplete degradation or sorption during treatment in septic tanks and leach fields allow some contaminants to percolate to the groundwater. PFAS were reported in domestic wells in a Massachusetts study where septic systems were prevalent (Schaider et al., 2016).

3.4.3 Landfilled products

Local health districts directly regulate landfills in Washington under rules authored by Ecology, specifically Chapters <u>173-350</u>¹⁰⁰ and <u>173-351</u>¹⁰¹ WAC. Ecology reviews and approves landfill permits issued by local health districts. Ecology's landfill rules do not require monitoring for PFAS. Ecology's rules allow health districts to include stipulations in permits that require landfills to sample for additional constituents.

Waste disposal in Washington includes all waste that goes to landfills or incinerators in the state, including waste brought from out-of-state, but does not include waste sent out-of-state for disposal. Table 31 illustrates typical annual waste tonnages handled in Washington, based on data collected in 2016. A total of 9,540,438 metric tons of waste were disposed in all types of landfills and incinerators in Washington in 2016 (Ecology, 2016).

Landfill type	Facilities in Washington	Metric tons disposed
Municipal Solid Waste Landfills	17	8,667,147
Inert Waste Landfills	23	1,570,957
Limited Purpose Landfills	12	521,884
Waste to Energy Facility	1	251,879

11,011,867

Table 31. Summary of waste disposed in 2016 in Washington state (Ecology, 2017).

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As described in <u>Appendix 9: Regulations, Section 9.1.2 Washington state rules</u>, wastes containing halogenated organic compounds, such as PFAS at concentrations above 100 parts per million, are designated and managed as state dangerous waste.

Landfill Leachate

TOTAL

Landfill leachate has been recognized as a potential source of PFAS into the environment. Various types of landfills can be used for long term storage of wastes that may contain PFAS. For example, privately and publicly operated landfills which receive and store commercial and residential wastes are likely to receive products which contain PFAS. Limited Purpose Landfills (see WAC <u>173-350-100</u>¹⁰²) may also be a source if they store industrial wastes where PFAS occurred in a manufacturing process.

Landfill storage conditions can result in PFAS degradation and mobilization, including migration into landfill leachate (Hamid et al., 2018). Uncontrolled leachate can migrate into groundwater, resulting in groundwater contamination. In Washington state, solid waste landfills are regulated under different administrative codes, depending on the type of landfill. Only Limited Purpose

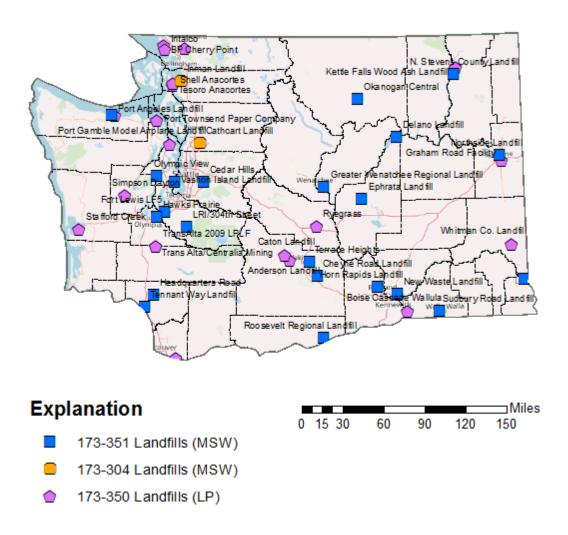
¹⁰⁰ https://apps.leg.wa.gov/wac/default.aspx?cite=173-350

¹⁰¹ https://apps.leg.wa.gov/wac/default.aspx?cite=173-351

https://apps.leg.wa.gov/wac/default.aspx?cite=173-350-100

Landfills (Chapter 173-350¹⁰³ WAC) and Municipal Solid Waste Landfills (Chapter 173-351¹⁰⁴ WAC) are required to have leachate collection systems. This includes most of the active landfills, with the exception of Inert Waste Landfills. A few closed landfills regulated under Chapter 173-304¹⁰⁵ WAC also have leachate collection systems. Older landfills, whether still operating or not, may not have been constructed with liner systems to capture leachate.

Figure 24. Locations of municipal solid waste (MSW) and limited purpose (LP) landfills in Washington.



Under current State requirements, landfill leachates that are collected are sent either to WWTPs or evaporation ponds. Ecology does not collect data regarding volumes of leachate produced. Ecology staff conducted an informal survey of MSW landfill operators to collect data

¹⁰³ https://apps.leg.wa.gov/wac/default.aspx?cite=173-350

¹⁰⁴ https://apps.leg.wa.gov/wac/default.aspx?cite=173-351

¹⁰⁵ https://apps.leg.wa.gov/wac/default.aspx?cite=173-304

quantifying the volumes of leachate typically produced (Carter, 2020). Table 32 summarizes the data collected.

Table 32. Landfill leachate production from select landfills located in Washington (Carter, 2020).

Landfill	County	Туре	Million liters collected	Collection period covered
Headquarters Road	Cowlitz	MSW	212.48	2018, January through November
LRI/304 th Street	Pierce	MSW	30.81	2018, January through March
Tenant Way	Cowlitz	MSW	17.45	2018, January through November
Hawks Prairie	Thurston	MSW	11.85	2018, January through November
Port Angeles	Clallam	MSW	4.73	2017, entire year
Hidden Valley	Pierce	MSW	0.42	2018, January through June
Fort Lewis LF5	Pierce	MSW	0.004	2017, July through December

There is no information regarding the incidence of PFAS in landfill leachate in Washington state. Landfills store wastes containing PFAS representative of items manufactured or in commerce in the state, including but not limited to: industrial wastes, carpeting and upholstery, waterproof clothing, food packaging waste, and—under very specific circumstances—biosolids. PFAS could also enter landfill leachate as a result of fire response using fluorinated foams at a landfill—landfills are required to provide for fire suppression in accordance with state regulation, and in some cases, AFFF may be recommended (California Department of Resources Recycling and Recovery, 2018). No data is available regarding such fire response activities in Washington.

Surveys were conducted in other parts of the U.S. to quantify PFAS contributions to solid waste streams and their impact to PFAS in landfill leachate in particular (Lang et al., 2017). Others have reported data collected from around the world, including (Hamid et al., 2018):

- A study in Germany which identified 44 PFAS in landfill leachate.
- A study showing a range of PFOA in leachate in U. S. landfills ranging from 0.15-9.2 $\mu g/l$.
- Measurements in Chinese landfills as high as 214 μg/L.

Adverse impacts to drinking water resulting from improperly managed landfill leachate have also been documented, in particular when landfills accepted manufacturing wastes known to contain high levels of PFAS (Michigan PFAS Action Response Team, 2020).

Finally, contaminants such as volatile organic compounds (including sufficiently volatile PFAS, which can partition from aqueous solutions) can be transported in landfill gas formed during waste decomposition. As reported by Hamid et al. (2018), studies have demonstrated elevated concentrations of airborne PFAS near certain landfills. EPA plans additional investigation of

PFAS in landfill gas emissions via a grant issued to North Carolina State University at Raleigh and Oregon State University (EPA, 2019c).

Waste characterization studies

A waste characterization study involves sampling, sorting, and surveying waste material delivered to landfills over a one year period. Ecology conducted waste characterization studies in 2009 and 2015 (Ecology, 2010, 2016b). Wastes were separated into 130 material types in 2009 and 156 material types in 2015. A few of those material types include products that may contain PFAS: carpet, furniture, textiles, and paper and packaging. The landfilled quantity for those products reported in 2016 are summarized in Table 33.

The amount of PFAS-containing materials landfilled in Washington is unknown. The disposed volumes listed in Table 33 are used to estimate PFAS disposal in Washington.

National Control	A second section to self-the d	Barrari after all d'accord
Material type	Annual metric ton landfilled	Percent of total disposed
Carpet	64,873	1.4
Furniture	59,842	1.3
Textiles	167,357 *	3.7
Paper packaging	332,543 ^	7.2
TOTAL four types	624,615	12.17
TOTAL waste landfilled	4.589.537	

Table 33. 2015 - 2016 waste characterization data.

Notes:

- * = Excludes footwear.
- ^ = Kraft/cardboard that is less likely to contain PFAS.

Carpet

PFAS used in flooring products include carpet and carpet cleaning and treatment products. From 1970 to 2002, carpet applications included perfluorooctanesulfonyl fluoride (POSF)-derived substances, including PFOS (DEPA, 2013). Currently, another PFAS subgroup—termed fluoropolymers or fluorotelomer-based acrylate polymers—are generally used for carpet stain resistance and carpet care treatments (Bowman, 2018; KEMI, 2015). However, other PFAS can also be present as impurities (for example, PFHxA and PFBA) (Bowman, 2018), or can be formed during environmental degradation (FTOHs and PFCAs) (Washington & Jenkins, 2015).

More than 90% of carpets used in homes and 100% of commercial carpeting is made from plastic. Carpets remain in place for 10 to 12 years or longer before disposal. Between 50% and 90% of carpet is treated for stain resistance with fluorinated substances (DEPA, 2013). Stain resistance treatments are lost each year through vacuuming, steam cleaning, and eventual disposal. Carpet in landfills can take hundreds of years to degrade. Compared to places without carpet, homes and offices with carpet can have higher concentrations of various PFAS in the indoor environment (Fraser et al., 2013; Gewurtz et al., 2009; Kubwabo, Stewart, Zhu, & Marro, 2005).

Based on two reports, a wide range of estimates can be applied to the 65,000 metric tons of carpet annually disposed in Washington landfills:

- One study reported an average concentration of 75 mg/kg (part per million) of PFOS in a mix of treated and untreated carpet (DEPA, 2013). Applying that concentration to the carpet annually disposed results in an estimate of 7 metric tons of PFOS annually disposed in Washington. That would result in a total of 214 metric tons of PFOS over a 30 year period.
- A Swedish estimate reported that treated synthetic carpet contains up to 15% PFAS (KEMI, 2015). That concentration would reflect a total of 14,300 metric tons of PFAS annually disposed in Washington. That would result in a total of 430,000 metric tons of PFAS landfilled over a 30 year period.

Furniture

PFAS are used to treat leather and upholstered furniture for stain resistance—from 1970 to 2002 using PFOS, and after 2002 using perfluorobutane sulfonyl fluoride-based products (PFBS). The U.S. imports 70% of its upholstered furniture from China—other imports come from Vietnam, Mexico, Canada, and Italy (World Furniture Online, 2017). Furniture usually remains in use for more than 15 years before landfilling.

Based on data from a Danish study, the following are estimates for landfill disposal of PFOS in the 71,424 metric tons of upholstered furniture disposed annually (DEPA, 2013):

- Using an average concentration of 80 mg/kg of PFOS in treated leather amounts to an annual disposal of 5.7 metric tons of PFOS. That would result in a total of 171 metric tons landfilled over a 30 year period.
- Based on a concentration of 2.4 mg/kg in a mix of treated and non-treated furniture amounts to an annual disposal of 0.17 metric tons of PFOS. That would result in a total of 5 metric tons of PFOS over a 30 year period.

Textiles

In 2015, the Swedish Chemicals Agency (KEMI) reported treatments or membrane construction of textiles, including:

- Fluoropolymer dispersions (like polytetrafluoroethylene or PTFE) used in industrial fabrics and professional apparel as well as highly porous fabrics like outdoor clothing and camping equipment.
- Side-chain fluorinated polymers (like PASF or fluorotelomer-based acrylate polymers) used as surface treatments on textiles and leather.

Current polymer chemistry used for textiles includes polyfluorinated (meth) acrylate polymers (C2 - 20). However, in the U.S., C8 - C20 polymers have been discontinued since 2015 under voluntary stewardship programs, being largely replaced by C6 based acrylate products. Other polymers include fluorinated urethanes (C4 - C18). Other raw materials include various polyfluorinated or perfluorinated substances. These are alkyl sulfonamide derivatives (C4 - 9),

alkyl ammonium compounds (C4 – C7), alkyl alcohols (C3 – C14), and a smaller number of alkyl sulfonic acids/sulfinic acids (C8), alkyl thiols (C8 – C20), alkyl sulfonamides (C8), alkyl esters (C8 – 14), alkanes/alkenes (C6), and alkanoyl/sulfonyl chlorides or fluorides (C8). Protective clothing uses surface treatments of side-chain fluoropolymers or woven fluoropolymer textiles. Examples include fire retardant clothing used for medics, pilots and firemen. As indicated in Appendix 9: Regulations, Section 9.2.1 Environmental Protection Agency, U.S. manufacturers have voluntarily discontinued production and use PFOS and PFOA, however products entering from other countries may still contain long-chain PFAS.

Table 34 lists 137,755 metric tons of textiles annually disposed—not all of these textiles are PFAS-treated. For the purpose of this estimate, 50% of that total is assumed to be PFAS-treated, which represents 68,877 metric tons of textiles disposed annually:

- 43 metric tons of PFAS annually landfilled based on 627.3 mg/kg perfluoro-carboxylic acid (PFCA) in textiles (Khotoff et al., 2013). Over a 30 year period, this represents 1,300 metric tons of PFCA.
- 2,066 metric tons of PFAS annually landfilled based on 3% by weight of PFAS in treated textiles (KEMI, 2015). Over a 30 year period, this represents 62,000 metric tons of PFAS.

Food packaging

Surface treatment and impregnation products provide water, oil, and grease resistance, and non-stick performance for paper and packaging. These include both food-contact materials (e.g., popcorn bags, pizza boxes, and fast-food wrappers) and non-food applications (e.g., masking papers and folding cartons). Paper, cardboard, and packaging has a very short lifespan from use to disposal. Treated food contact material is generally limited to a one-time use.

In 2015, the Swedish Chemicals Agency (KEMI) reported fluorinated applications in paper packaging, including mainly side-chain fluorinated polymers and polyfluoroalkyl phosphonic acids (PAPs and diPAPs). Other major substance groups were poly- or perfluorinated alkyl thiols (C4 – 20), poly- or perfluorinated alkyl sulfonamide derivatives (C4 – C9), and poly/perfluorinated alkyl phosphorus compounds (C8), as well as smaller number in the substance groups alkyl esters (C6 – 14), alkyl silicones/siloxanes (C6), and alkyl sulfonic/sulfinic acids (C8). As discussed in Appendix 9: Regulations, Section 9.2.2 Food and Drug Administration, FDA regulates the use of FPAS in food packaging. FDA revoked its food additive regulations for use of three long-chain perfluorinated compounds in 2015 (FDA, 2015). Current products on the FDA food contact notification (FCN) list are short-chain fluorotelomer-based polymers and perfluoropoly ethers (FDA, 2019, 2020).

An estimated 17% of disposed paper products and packaging are treated (Trier et al., 2011). For this estimate, 20% of 223,771 metric tons of paper and packaging was used to estimate impacts from landfilled textiles. An estimated 44,751 metric tons of PFAS-treated textiles are used for the estimates below:

- 1.13 metric tons of PFAS annually landfilled based on 25.2 mg/kg fluorotelomer alcohol (FTOH) in treated paper and packaging (Liu et al., 2015). Over a 30 year period, this amounts to 33.83 metric tons of FTOH.
- 671 metric tons of PFCA annually landfilled based on the conservative upper end of 1.5% by weight of PFCA in treated paper products (KEMI, 2015; UNEP, 2015b). Over a 30 year period, this amounts to 20,139 metric tons of PFCA.

Summary

The low and high PFAS disposal estimates are based on limited information from the waste sort data and available product testing data. The greatest sources of PFAS disposal appears to come from carpet and textiles. These estimates are based on the information available in the literature. It is important to note that we received input that the KEMI estimation of 15% is too high, and industry representatives report use at around 0.1%, which would lead us to estimate that between 90 and 140 metric tons of PFAS are used in Washington carpets each year.

Material type	Low estimate of PFAS disposal	High estimate of PFAS disposal
Carpet	7.15 metric ton/year*	14,300 metric ton/year
Textiles	43.21 metric ton/year	2,066 metric ton/year
Furniture	0.17 metric ton/year	5.71 metric ton/year
Compostable paper,	1.13 metric ton/year	671 metric ton/year
packaging		
TOTAL	51.66 metric ton/year	17.043 metric ton/year

Table 34. Annual PFAS disposal estimates by material type.

3.4.4 Dangerous waste disposal reports

Washington's Dangerous Waste Regulation requires businesses to properly manage, store, and dispose of hazardous waste (Chapter 173-303¹⁰⁶ WAC). This regulation identifies halogenated organic compounds as a state-only "dangerous waste" due to persistence. Fluorine is a halogen, therefore PFAS are halogenated organic compounds. PFAS present in a waste above 100 ppm must be properly managed and disposed as dangerous waste (WAC 173-303-040¹⁰⁷).

Dangerous waste disposal must be reported to Ecology. Since 2010, those reports have been entered into the TurboWaste database. PFAS is not specifically reported to the database. Waste data entered into TurboWaste that may contain PFAS include wastes described as AFFF, fire debris, and suppressant. Those reports are summarized in Table 35—the submitted reports do not all indicate the presence of PFAS.

TurboWaste data is reported in pounds. For consistency throughout this appendix, the data was converted to kilograms.

¹⁰⁶ https://apps.leg.wa.gov/wac/default.aspx?cite=173-303

¹⁰⁷ https://apps.leg.wa.gov/wac/default.aspx?cite=173-303-040

Table 35. Dangerous waste disposal reports from 2010 to 2016 (kilograms).

Waste	2010	2011	2012	2013	2014	2015	2016
AFFF	1,252	6,762	877	931	1,528	5,640	40,632
Fire debris	1,316	722	784	8,634	6,378	504	1,555
Suppressant	1,946	6,112	2,445	25,908	96,272	2,867	0
TOTAL	4,514	13,596	4,105	35,473	104,179	9,010	42,187

3.4.5 Compost

Testing for the presence of PFAS chemicals in the environment has been primarily directed at water and biosolids. According to EPA guidance, the threshold for concern in drinking water is 70 parts per trillion (ppt). Currently, there is no national PFAS threshold for soils or compost. The general consensus is that inclusion of food scraps, food packaging, and biosolids in composting operations will introduce some amount of PFAS, but testing has shown the levels to be low (Croker, 2020; Beecher & Brown, 2018; Choi et al., 2019).

Some concern has been expressed that PFAS in compost may be taken-up by plants, but research to date suggests that the concern for plant uptake is minimal (Beecher & Brown, 2018). There is currently minimal regulatory concern with regard to inhalation, ingestion, dermal contact, or other possible routes of exposure related to organic residuals (NEBRA, 2019).

Attention to reducing the use of PFAS chemicals has already resulted in a reduction in PFAS in human blood serum by 50% over the past 15 years (Rainey & Beecher, 2018). Recognizing the impact that PFAS in food packaging is having on human and environmental health, in 2018 the Washington State Legislature passed a bill that prohibits the use of PFAS in paper food packaging (Ecology, 2018). These efforts are considered to be good steps toward reducing the amount of PFAS in biosolids and composts, but additional research specific to compost will add to this understanding.

3.5 Global estimate: Washington proportion

PFAS emissions have not been tracked in Washington state. We neither know historical emission rates nor current emission rates statewide. We reviewed available assessments of historical global emission rates to estimate historical emission rates in Washington.

Global releases of PFAS are estimated in Prevedouros et al. (2006), Wang et al. (2014a), Paul et al. (2009), and Boucher et al. (2019). Others have also summarized global emission inventories (OECD, 2015). We used a proportion of the global use and disposal estimates to determine historic releases of PFAS in Washington. Global estimates related to manufacture of PFAS are not applicable to Washington because no primary PFAS manufacturing occurred in the state. As

such, we have excluded these from our estimates below. A brief summary of each estimate and its associated Washington proportion is provided in the subsections that follow.

To calculate the Washington state proportion of global emissions, we assumed that the U.S. represents 25% of worldwide consumption and Washington represents 2.6% of the U.S. by population. Therefore, the state's emissions would represent 0.65% of global emissions.

These estimates do not reflect all PFAS that may have been present in the global market or released to the environment. The OECD indicated that, "Identifying and understanding production, use, releases, and environmental presence of the various PFAS on the global market has been limited due to the complexity of the issue, data scarcity and fragmentation, and data confidentiality" (OECD, 2018). Researchers have recognized the lack of available information and have only been able to qualitatively assess the emissions of certain PFAS (Wang et al., 2014b).

Wang et al. (2014a) and Boucher et al. (2019) also estimated emissions of certain PFAS following the voluntary stewardship reductions in the U.S. and other countries. These are presented in <u>Section 3.5.5</u> below.

3.5.1 Estimate of PFCA and FTOH emissions

Prevedouros et al. (2006) described the sources, fate, and transport of PFCA in the environment. Prevedouros estimated PFCA and FTOH releases to the environment from direct (manufacture, use, consumer products) and indirect (impurities, precursors) sources. These estimates were based on total emissions from 1960 – 2002. The global estimates of use, disposal, and emissions from consumer and industrial products and firefighting foam are presented in Table 36. Table 36 also identifies the estimated proportion attributable to Washington state, excluding manufacturing emissions. For the 42-year period from 1960 – 2002, this would represent average emissions of 0.10 metric tons per year for Washington state.

Table 36. Global and Washington state estimated PFCA and FTOH emissions for the period 1960 – 2002.

Use, disposal, and emissions*	Global emissions (metric tons)	Washington emissions (metric tons)
Consumer and industrial	520	3.38
AFFF	131	0.85
TOTAL	651	4.23

Note: * = Not including manufacturing emissions.

Wang et al. (2014a) expanded on the Prevedouros (2006) study, estimating that indirect degradation sources in the period 1951 – 2002 could have been five times higher than those presented by Prevedouros.

3.5.2 Estimate of POSF and PFOS emissions

Paul et al. (2009) estimated global historic manufacture, consumer use and disposal of POSF, and environmental releases of POSF and PFOS from 1970 – 2002. Manufacture estimates in the Paul et al. (2009) study do not apply to Washington for reasons stated above. Total global consumer use and disposal of perfluorooctanesulfonyl fluoride (POSF) from direct (use and consumer products) and indirect PFOS (precursors and/or impurities) sources are presented in Tables 37 and 38 respectively. These tables also present the estimated Washington proportion.

Estimates indicate that direct emissions from POSF-derived products are the major source to the environment resulting in releases into wastewater streams, primarily through losses from stain repellent treated carpets, waterproof apparel, and aqueous firefighting foams. For the 32-year period from 1970 – 2002, this would represent average direct emissions of 29.4 metric tons per year for Washington.

Table 37. Global and Washington state POSF direct use and disposal emissions for the period 1970 – 2002.

Direct use and disposal emission category	Global (metric tons)	Washington (metric tons)
Carpet	48,000	312
Paper and packaging	24,000	156
Apparel	12,500	81
AFFF	10,000	65
Performance chemicals (hydraulic fluids)	6,000	39
TOTAL	100,500	653

Table 38. Global and Washington state POSF and PFOS indirect emissions to water and air for the period 1970 – 2002.

Indirect consumer emission category	Global (metric tons)	Washington (metric tons)
Carpet	21,500	140
Apparel	12,600	82
Performance chemicals (hydraulic fluids)	9,610	62
Paper and packaging	367	2.4
AFFF	47	0.3
TOTAL	44,124	286.7

3.5.3 Estimate of PHxSF and PFDS emissions

Boucher et al. (2019) estimated global historic manufacture, consumer use, and disposal of perfluorohexane sulfonate (PFHxS), perfluorodecane sulfonate (PFDS), perfluorohexane

sulfonyl fluoride (PHxSF), and perfluorodecane sulfonate (PDSF) from 1958 – 2015. Manufacture estimates in the Boucher et al. (2019) study do not apply to Washington for the reasons stated above. Total global use and disposal of PFHxS and PFDS and degradate emissions are summarized in Table 39 and reflect the Washington proportion.

Table 39. Global and Washington PFHxSF and PFDS Emissions in the period 1958 – 2015 (Boucher et al., 2019).

Emissions from use, disposal, and degradates*	Global (metric tons)	Washington (metric tons)
PFHxS	32 – 126	0.2 – 0.8
PDFS	34 – 372	0.2 - 2.4
TOTAL	66 – 498	0.4 – 3.2

Note: * = Not including manufacturing emissions.

3.5.4 Summary of historical emissions

We summed all of the Washington proportions of historical PFAS emissions calculated above. Uncertainty is introduced regarding total emissions over a certain period of time because different time accounting periods were considered in each of the studies above. Nevertheless, we can estimate Washington state's average annual contribution over this historical period at approximately 29.5 metric tons per year. Table 40 provides the summation of Washington's proportion of PFAS emissions.

Table 40. Average annual historical Washington state PFAS emissions based on global estimates.

PFAS emission type	Period of estimate	WA emissions during period (metric tons)	Average annual WA emissions (metric tons)	Reference
PFCA and FTOH	1960 –	4.23	0.10	Prevedouros
	2002			et al., 2006
Direct POSF	1970 –	653.00	20.41	Paul et al.,
	2002			2009
Indirect POSF	1970 –	286.70	8.96	Paul et al.,
and PFOS	2002			2009
PFHxS and PDFS	1958 –	0.40 – 3.20	0.01 – 0.06	Boucher et
	2015			al., 2019
TOTAL			29.47 – 29.52	

3.5.5 Current emissions

Both Wang et al. (2014a) and Boucher et al. (2019) recognized that estimates previously performed by others were limited to historical emissions during periods when PFAS manufacturing was not limited in the U.S. However, as identified in Appendix 1: Chemistry,

Section 1.3.4 Technical quality and implications for environmental impacts, following voluntary phase-outs of PFOA and fluoropolymer manufacturing in the U.S., Japan, and Europe, production of these PFAS moved to other countries. Estimates by Wang et al. (2014a) and Boucher et al. (2019) indicate that although production and degradation emissions of some PFAS identified above may have decreased (PFNA products and FTOH-based products as impurities), overall emissions after 2015 may have remained similar to those during the period 2003 – 2015. The estimates in Boucher et al. (2019) also emphasize that even though PFHxS and PFDS production emissions have significantly decreased, degradation emissions of PFHsX continue.

3.6 Data gaps and recommendations

3.6.1 Data gaps

Secondary manufacturing use of products containing PFAS

As identified in <u>Section 3.1.2</u> above, various information sources indicate that PFAS can be used in manufacturing industries that operate, or have operated in Washington. Although some uses have been readily identifiable (for example, AFFF use), we do not know the extent of PFAS used in Washington manufacturing, the types of PFAS which were historically used, or whether manufacturers have transitioned to new generation PFAS or ceased use altogether. We also do not know whether manufacturing uses resulted in human exposure or emissions to the environment, and if so, the risks associated.

Information regarding PFAS use may be readily available for certain sectors. For example, the electroplating industry identified its use of PFOS early, and sought to eliminate or replace such usage industry-wide in the U.S. (National Association of Surface Finishers (NASF), 2019a). The industry, however, has identified that legacy use of PFOS can continue to be a source of emissions, and has researched the effects of replacement PFAS used such as 6:2 FTS (NASF, 2019b). Electroplaters are regulated in Washington state via various regulations—therefore, more is likely known about past and current electroplating locations and can be learned about their PFAS usage practices. However, use in other sectors is simply derivative of component manufacturing—for example the medical industry uses PFAS coated components which may or not be manufactured in Washington (UNEP, 2015a). Similarly, we do not know if semiconductor manufacturing in the state employs PFAS substances. More research is needed to identify industrial sectors that contribute to PFAS use, and which of those have a connection to environmental or human exposure.

Use of AFFF in industry

Use of PFAS at military installations and the extent of resulting environmental impacts are under investigation by the DOD at a number of sites in Washington state (see <u>Section 3.2.3</u>). AFFF use in firefighting, and especially use by public fire departments, is being addressed

through the implementation of Chapter 70.75A¹⁰⁸ RCW (see Appendix 9: Regulations, Section 9.1.1 Washington state laws). Ecology is already working with public fire departments to assess quantities of AFFF stored and means for its collection and disposal. Over time, Chapter 70.75A RCW will require civilian airports and other industry to find non-PFAS fire-fighting products. However, Ecology has insufficient information about AFFF holdings by civilian airports and petrochemical industries. Ecology also lacks a complete list of regionalized fire training centers.

WWTPs

At this time Ecology has limited data confirming the presence of PFAS in WWTP effluents. Ecology does not know the range of PFAS concentrations in WWTP influent, effluent, and sludge, and therefore cannot assess the relative contribution of these sources to the environment. This information is also needed to determine the efficacy of possible treatment technologies at WWTPs to remove PFAS. Ecology also needs more information regarding PFAS removal performances of different treatment technologies (e.g. secondary, secondary with nutrient removal, tertiary membrane filtration), and the role of multiple-benefits of different technologies, including nutrient removal and removal of a broad spectrum of contaminants of concern.

Landfill PFAS emissions

Although information about PFAS emissions from landfills is being collected elsewhere, we do not have information regarding the incidence of PFAS in landfill leachate in Washington. We also do not know if PFAS-contaminated landfill leachate is a source of groundwater contamination. Finally, we do not know the extent to which PFAS entering landfills partitions into gaseous emissions, preventing us from understanding the true mass balance of PFAS emissions from this source.

3.6.2 Recommendations

The following recommendations result from the analyses presented in this appendix:

Recommendation 2.3: Work to prevent PFAS releases from firefighting foam use and manufacturing processes.

Ecology will work proactively with industry, manufacturers, and businesses to eliminate releases to the environment from the use of PFAS-containing AFFF or other manufacturing processes using PFAS.

To address PFAS in AFFF, Ecology would:

 Collaborate with users of firefighting foam to develop and share outreach materials and best management practices that address the proper use, storage, and disposal of PFAS-containing AFFF.

¹⁰⁸ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

- Ensure that industrial use of PFAS-containing AFFF provides for containment procedures along with collection of this foam and contaminated soil or sediment for proper designation and disposal. Costs to industrial users to collect and dispose of released PFAS-containing AFFF include plan development, employee training, methods for containment, and disposal of waste.
- Continue identification of organizations and industries that store and use AFFF in training and emergency firefighting, including use of AFFF in tunnels.
- Assist state and local governments, airports, industry, and fire districts with prioritizing the quantification, disposal, and replacement of PFAS-containing AFFF, especially in communities with cumulative impacts, health disparities, and environmental justice considerations.
- Share information about PFAS-free Class B firefighting foam with users of firefighting foam as information or research is available, including GreenScreen® certifications (New York State Pollution Prevention Institute, 2019).
- Conduct compliance and enforcement actions to ensure the law is being followed.

Ecology will work proactively with industry, manufacturers, and businesses to eliminate releases to the environment from the use of PFAS in manufacturing or other processes.

- Ecology will review data from other states and countries to identify industrial or manufacturing uses of PFAS. Ecology will use this information to identify industries in Washington that have used or continue to use commercial quantities of PFAS. Ecology will also track future TRI reports (starting in 2021) for industries.
- Ecology will evaluate PFAS release potential from those industries which may have used, or continue to use, PFAS.
- Ecology will reach out to these industries to discuss their use of PFAS, identify opportunities to switch to safer alternatives, implement best practices, and ensure proper waste management.

Recommendation 3.1: Reduce PFAS exposure from carpets, water and stain resistance treatments, and leather and textile furnishings.

Ecology identified these products as significant sources and uses of PFAS under Chapter 70.365¹⁰⁹ RCW (Ecology, 2020c). This identification was based on the volume estimates presented in this chapter. We recommend that Ecology determine whether safer alternatives are feasible and available. These determinations should be made by June 2022 and would be accompanied by proposed regulatory actions to reduce exposure.

Regulatory actions could include:

- Requesting that carpet and carpet care manufacturers:
 - o Identify the products containing PFAS.
 - Disclose the use of priority chemicals in product ingredients.
 - Disclose information regarding exposure and chemical hazard.

¹⁰⁹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365

- Describe the amount and function of PFAS in carpet and carpet care products.
- Proposing restrictions of PFAS-containing carpet and carpet care products when a safer alternative is available and feasible, and the restriction will reduce a significant source of PFAS or protect sensitive populations or species.
- Proposing actions to reduce legacy PFAS-containing carpet and carpet care products remaining in homes, especially in low-income households where items may be retained past their typical product lifespan.

In addition to the above, we recommend the following actions:

- Implement a purchasing preference policy for PFAS-free carpet. Work with vendors
 on the state flooring contract to offer PFAS-free carpet on all state master contracts
 and all agency contracts. Purchasing PFAS-free carpet could result in increased costs
 to the state.
- If safer alternatives are available, include them in Ecology's <u>Product Replacement Program</u>¹¹⁰ to replace PFAS-containing carpet in community centers, libraries, daycares, and other environments where children may be disproportionately exposed.

Recommendation 3.2: Identify additional sources and uses of PFAS to consider in the second Safer Products for Washington cycle.

The priority products identified in 2020 under the Safer Products for Washington program do not account for all sources and uses of PFAS. We will continue research to better understand the contribution of other products to PFAS concentrations in homes, workplaces, and the environment. These include PFAS in:

- Water resistant clothing and gear.
- Nonstick cookware and kitchen supplies (e.g., baking paper).
- Personal care products.
- Cleaning agents.
- Floor waxes and sealants.
- Ski waxes.
- Car wash products.

Ecology should engage with overburdened communities regarding consumer products that may contain PFAS. Communities use consumer products differently. It is important that the team identify consumer products which might be disproportionately exposing overburdened communities.

In addition to determining whether these products are significant sources or uses of PFAS, preliminary investigations into the availability and feasibility of safer alternatives should be conducted. This will help the Safer Products for Washington team identify opportunities to reduce additional sources and uses of PFAS. If safer alternatives are identified, outreach should

¹¹⁰ https://ecology.wa.gov/Waste-Toxics/Reducing-toxic-chemicals/Product-Replacement-Program

be conducted to increase voluntary adoption. If these products are identified as significant sources or uses, they should be added as priority products in 2025 and regulatory actions should be determined by 2027.

Recommendation 3.3: Implement additional reduction actions for PFAS from consumer products.

In addition to the activities described above, we recommend the following actions:

- Gather input from low income and other historically overburdened communities, including communities of color. Develop a list of ways to reduce exposure that include low cost and subsidized approaches. These may be particularly important measures to employ in communities with higher exposure from drinking water. No cost estimate is provided to conduct this evaluation or to develop exposure reduction recommendations.
- Establish a purchasing preference policy for PFAS-free products. Work with vendors
 to offer PFAS-free textiles and furniture. Apply this policy to all state master
 contracts and all agency contracts. Purchasing PFAS-free products could increase
 state costs.
- Propose a ban on the import or sale of all products in Washington containing
 phased-out long-chain PFAAs. Long-chain PFAAs include perfluorinated carboxylates
 (PFCAs) with eight or more fully fluorinated carbons (for example, PFOA) and
 perfluorinated sulfonates (PFSAs) with six or more fully fluorinated carbons (for
 example, PFHxS and PFOS), their salts, and precursor compounds capable of forming
 long-chain PFAAs.

Recommendation 4.1: Evaluate PFAS in wastewater treatment.

Ecology should evaluate PFAS in wastewater treatment plant (WWTP) effluent and influent to develop a greater understanding of PFAS in discharges in Washington:

- Ecology should develop a study design to sample PFAS in three different types of plants: WWTPs with secondary treatment, nutrient removal, and advanced solids removal. Sampling should include products of selected WWTP unit processes (for example, primary and secondary clarifiers or dechlorination) to help differentiate removal efficiencies of the different treatment types.
- The study design should ensure that the WWTPs that are sampled receive industrial discharges that are likely to contain PFAS, or that have drinking water sources with known PFAS contamination.
- Ecology should identify industries that are likely to generate wastewater containing PFAS.
- Based on the information from the study, Ecology should consider additional
 monitoring requirements for WWTP dischargers. This should include consideration
 of whether EPA has developed approved analytical methods for PFAS suitable for
 WWTP effluent and a regulatory target (a nationally recommended water quality
 criterion for PFAS) for waters of the state.

 Based on this evaluation, Ecology should require possible PFAS monitoring for some or all domestic and industrial WWTPs.

Recommendation 4.2: Evaluate landfill PFAS emissions.

Ecology will develop and conduct a sampling program at selected landfills throughout the state to test for the presence of PFAS in leachate, groundwater, and air emissions.

Leachate

The Solid Waste Management program (SWM) developed Phase I of the program, leachate sampling. This phase has received funding and approval, and is undergoing final planning. Sampling will begin when the COVID-19 restrictions are withdrawn and normal field operations resume. Ecology has developed a study to better characterize landfill leachate. The study design will:

- Sample leachate at selected landfills in the state.
- Determine the range of values for 33 PFAS substances in leachate, and compare to landfills throughout the country.
- Arrive at an estimate of the total PFAS materials in the landfill leachate through Total Oxidized Precursor (TOP) analyses.
- Determine if differences in amount of PFAS occurs in landfill cells of different ages.
- Determine if specific types of waste streams lead to higher PFAS values.
- Identify disposed wastes that are likely to generate PFAS releases to leachate.
- Perform a one-time testing of leachate from approximately 23 landfills.
- Consider additional sampling of leachate for landfills not yet sampled after the initial Phase I is completed. This second step of Phase I may include landfills that are undergoing MTCA cleanups, or landfills that contain specific refuse streams that have shown to have high PFAS values from the Phase I sampling.

If warranted, Ecology would manage PFAS in landfill leachate long term by:

- Considering additional monitoring requirements for landfills to test leachate for PFAS using information from the study mentioned above.
- Potentially updating the rules (Chapters <u>173-350</u>¹¹¹ and <u>173-351</u>¹¹² WAC) to require PFAS testing of leachate during landfill monitoring.

Groundwater and gaseous emissions

Phase II of the program will sample groundwater and gaseous emissions at landfills for PFAS. This phase of the program is in the conceptual stage. Landfills to be sampled will be based on the results of the Phase 1 leachate study. Groundwater will be sampled from existing monitoring wells.

The Solid Waste Management program (SWM), in conjunction with the Air Quality Program (AQ), will develop the gas emissions sampling portion of the program. Ecology will also monitor

¹¹¹ https://apps.leg.wa.gov/wac/default.aspx?cite=173-350

¹¹² https://apps.leg.wa.gov/wac/default.aspx?cite=173-351

Per- and Polyfluoroalkyl Substances Draft Chemical Action Plan

landfill gas emissions monitoring being conducted by North Carolina State University and Oregon State University (EPA, 2019).

Landfill waste makeup

In parallel to landfill gas emission sampling above, Ecology will continue to research the makeup of PFAS waste entering and potentially currently stored in landfills.

Supplement 1: Estimated PFAS in Consumer Products in a Typical Home

Table 41. Estimated PFCA in consumer products in a typical home.

Category name	Total PFCA	Typical Quantity	PFAS in the home	Reference
Pre-treated carpeting	484 μg/ m²	150 m ²	72,600 μg	Guo et al., 2009
Commercial carpet- care liquids	12,000 μg/kg	6 kg	72,000 µg	Guo et al., 2009
Treated home textile and upholstery	346 μg/ m ²	50 m ²	17,300 μg	Herzke et al., 2012
Waterproofing agents	29,889 μg/L	0.5 L	14,945 μg	Herzke et al., 2012
Pre-treated carpeting	57.2 μg/kg	50 kg	2,860 μg	Kotthoff, 2015
Food contact material (paper)	2,859.9 μg/kg	1 kg	2,860 μg	Kotthoff, 2015
Treated floor waxes and stone/wood sealants	2,430 μg/kg	1 kg	2,430 μg	Guo et al., 2009
Sunscreen	19,000 μg/kg	0.1 kg	1,900 μg	Fujii, 2013
Treated home textile and upholstery	336 μg/kg	5 kg	1,680 μg	Guo et al., 2009
Non-stick cookware	1,234.74 μg/kg	1 kg	1,235 μg	Herzke et al., 2012
Household carpet/fabric-care liquids and foams	953 μg/kg	1 kg	953 μg	Guo et al., 2009
Leather samples	627.3 μg/kg	1 kg	627 μg	Kotthoff, 2015
Foundation cosmetic	5,900 μg/kg	0.1 kg	590 μg	Fujii, 2013
Treated apparel	198 μg/kg	2 kg	396 μg	EPA, 2009
Compounding agent	35,000 μg/kg	0.01 kg	350 μg	Fujii, 2013
Talc	2,500 μg/kg	0.1 kg	250 μg	Fujii, 2013
Outdoor textiles	187.8 μg/kg	1 kg	188 μg	Kotthoff, 2015
Membranes for apparel	124 μg/kg	1 kg	124 μg	Guo et al., 2009
Ski waxes	11,365.5 μg/kg	0.01 kg	113 μg	Kotthoff, 2015
Gloves	169.4 μg/kg	0.2 kg	34 μg	Kotthoff, 2015
Awning cloth (outdoor)	31.6 μg/kg	1 kg	32 μg	Kotthoff, 2015

Category name	Total PFCA	Typical Quantity	PFAS in the home	Reference
Treated food contact paper	3,100 μg/kg	0.01 kg	31 μg	Guo et al., 2009
Electronics and electronic parts	25.51 μg/kg	1 kg	26 μg	Herzke et al., 2012
Thread sealant tapes and pastes	603 μg/kg	0.02 kg	12 μg	Guo et al., 2009
Paints and inks	9.36 μg/kg	1 kg	9 μg	Herzke et al., 2012
Waterproofing agents	80.6 μg/kg	0.1 kg	8 μg	Kotthoff, 2015
Treated non-woven medical garments	795 μg/kg	0.01 kg	8 μg	Guo et al., 2009
Household carpet/fabric-care liquids and foams	3.5 μg/kg	1 kg	4 μg	Kotthoff, 2015
Non-stick cookware	0.28 μg/ m ²	1 m ²	0.3 μg	Guo et al., 2009
Dental floss and plaque removers	31.3 μg/kg	0.005 kg	0.2 μg	Guo et al., 2009

Table 42. Estimated FTOH or FTS in consumer products in a typical home.

Category	FTOH/FTS	Quantity	FTOH/FTS in the home	Reference
Cleaning agents	667,700 μg/kg	1 kg	667,700 μg	Kotthoff, 2015
Commercial carpet care liquids	105,000 μg/kg	6 kg	630,000 μg	Liu et al., 2015
Treated floor waxes and stone/wood sealants	423,000 μg/kg	1 kg	423,000 μg	Liu et al., 2015
Waterproofing agents	464,774 μg/L	0.5 L	232,387 μg	Herzke et al., 2012
Treated home textile and upholstery	42,900 μg/kg	5 kg	214,500 μg	Liu et al., 2015
Carpet	4,010 μg/kg	50 kg	200,500 μg	Liu et al., 2015
Impregnating sprays (waterproofing)	1,857,300 μg/kg	0.1 kg	185,730 μg	Kotthoff, 2015
Treated home textile and upholstery	757 μg/ m ²	50 m ²	37,850 μg	Herzke et al., 2012
Carpet samples	73.5 μg/kg	50 kg	3,675 μg	Kotthoff, 2015
Membranes for apparel	1,590 μg/kg	1 kg	1,590 μg	Liu et al., 2015
Treated apparel	464 μg/kg	2 kg	928 μg	Liu et al., 2015

Category	FTOH/FTS	Quantity	FTOH/FTS in the home	Reference
Outdoor textiles	799.3 μg/kg	1 kg	799 μg	Kotthoff, 2015
Household carpet/fabric-care liquids and foams	372 μg/kg	1 kg	372 μg	Liu et al., 2015
Treated food contact paper	25,200 μg/kg	0.01 kg	252 μg	Liu et al., 2015
Treated home textile and upholstery	1.35 μg/m ²	50 m ²	68 μg	Herzke et al., 2012
Electronics and electronic parts	25.51 μg/kg	1 kg	26 μg	Herzke et al., 2012
Thread sealant tapes and pastes	1,220 μg/kg	0.02 kg	24 μg	Liu et al., 2015
Food contact material (paper)	23.4 μg/kg	1 kg	23 μg	Kotthoff, 2015
Gloves	98.3 μg/kg	0.2 kg	20 μg	Kotthoff, 2015
Treated nonwoven mediCal garments	1,460 μg/kg	0.01 kg	15 μg	Liu et al., 2015
Non-stick cookware	10.55 μg/kg	1 kg	11 μg	Herzke et al., 2012
Electronics and electronic parts	0.57 μg/kg	1 kg	0.6 μg	Herzke et al., 2012

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List of acronyms

General acronyms

Table 43. Acronyms found in the sources and uses appendix.

Acronym	Definition
ACRP	Airport Cooperative Research Program
AFFF	Aqueous film forming foam
AKART	All Known Available and Reasonable Technology
CAA	Clean Air Act
CFR	Code of Federal Regulations
CSPA	Children's Safe Products Act
CWA	Clean Water Act
DEPA	Danish Environmental Protection Agency
DOD	Unites States Department of Defense
DON	Department of Navy
DTSC	Department of Toxic Substances Control, California
ЕСНО	Enforcement and Compliance History
ESTCP	Environmental Security Technology Certification Program
EPA	United States Environmental Protection Agency
ERTS	Emergency Reporting Tracking System
ESD	Washington State Employment Security Department
EWG	Environmental Working Group
FAA	Federal Aviation Administration
HEPA	Heads of EPAs Australia and New Zealand
ITRC	Interstate Technology Regulatory Council
kg	Kilogram
L	Liter
μg	Microgram
m ²	Square meter
MIDEQ	Michigan Department of Environmental Quality
MIL-SPEC	U.S. Military Specification
MSRC	Marine Spills Response Corporation
NAICS	North American Industry Classification System
NASF	National Association of Surface Finishers
NFPA	National Fire Protection Association
NHDES	New Hampshire Department of Environmental Service
NRCNW	National Response Corporation Northwest
OECD	Organisation for Economic Co-Operation and Develop

Acronym	Definition
PPE	Personal protective equipment
RCRA	Resource Conservation and Recovery A
RCW	Revised Code of Washington
SERDP	Strategic Environmental Research and Development Program
SSEHRI	Social Science Environmental Health Institute
UNEP	United Nations Environment Programme
WAC	Washington Administrative Code
WSDOT	Washington State Department of Transportation
WWRL	Worldwide Response Resource List
WWTP	Wastewater treatment plant

Chemical names

Table 44. Chemical name acronyms found in the sources and uses appendix, excluding the general acronyms listed in the table above.

Acronym	Chemical Name
ADONA	Ammonium 4,8-dioxa-3H-perfluorononanoate
AFFF	Aqueous film forming foam
FTOH	Fluorotelomer alcohol
FTS	Fluorotelomer sulphonates
PDSF	Perfluorodecane sulfonate
PFAA	Perfluorooctanesulfonic acid
PFAS	Per- and polyfluoroalkyl substances
PFCA	Perfluorocarboxylic acid
PFDS	Perfluorodecane sulfonate
PHXsF	Perfluorohexane sulfonyl fluoride
PFOS	Perfluorooctane sulfonic acid
POFS	Perfluorooctanesulfonyl fluoride
PFHxS	Perfluorohexane sulfonate

Appendix 4: Fate and Transport

4.0 Overview

4.0.1 Findings

Transformation:

- All PFAS monomers are either perfluoroalkyl acids (PFAAs) or perfluoroalkyl acid (PFAA) precursors.
- PFAA precursors represent a large group of per- and polyfluoroalkyl substances (PFAS), which contribute terminal PFAS such as perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) to the environment.
- It is believed that all PFAA precursors will transform to PFAAs, with a timeframe that could range from hours to hundreds of years.
- Some PFAS polymers will likely serve as a continued source of PFAS emission due to polymer breakdown.

Fate:

- PFAS can be released to the ambient environment as a solid, liquid, or gas, depending on the source of the release (manufacturing or environmental degradation).
- Gas phase and aerosol particulate transport can cause PFAS contamination to occur at long distances from emission sources.
- With the exception of polymers, most PFAS are at least slightly water soluble and can be transported by water movement.
- Adsorption to carbon compounds in soil and sediments can slow PFAS transport by groundwater and surface water.
- Short-chain PFAS are more mobile, less bioaccumulative, and equally as persistent as long-chain versions.
- Chemical transformation of precursor compounds may change preferential partitioning into transport media and rate of transport.
- Landfill waste and biosolids from composting and wastewater treatment plants (WWTPs) may serve as continued sources of PFAS emissions into the environment.
- Some PFAS can bioaccumulate in plants and animals, and biomagnify in higher organisms in the food chain.

4.0.2 Introduction

The purpose of this appendix is to review PFAS transformation in the environment, and address how PFAS transformation products are transported and portioned in various environmental media.

As identified in <u>Appendix 1: Chemistry</u>, because there are hundreds of different PFAS currently on the market, their environmental fate and transport—which describes the chemical transformation and geographic distribution of compounds after release to the environment—

can vary greatly. Commercially manufactured PFAS and their subsequent transformation compounds can exist in many different forms (gas, water, solid) and will partition (i.e., group with separate media) differently depending on the type of compound and the surrounding ambient conditions.

Rate of PFAS chemical transformation can also vary quite dramatically depending on the chemical in question, the phase, and the environment where it is located. Some compounds have a half-life as low as hours in the environment, while others do not transform naturally.

4.1 Non-polymer PFAS

As presented in in <u>Appendix 1: Chemistry, Section 1.1.3</u>, many non-polymer PFAS have been identified and characterized.

PFOS and PFOA, both PFAAs, have been a primary source of attention in research and investigations regarding PFAS impacts. The presence of PFOS and PFOA in the environment results directly from their use and emission from manufacturing processes, or as a result of the degradation of long-chain or polymer substances—usually called precursors. PFAAs are very stable in the environment, and are referred to as terminal substances. As discussed in additional detail below, precursors can undergo several degradation steps prior to forming terminal PFAAs. PFAAs have not been shown to degrade or transform under natural conditions (Ochoa-Herrera, Field, Luna-Velasco & Sierra-Alvarez, 2016; Liou, Szostek, DeRito & Madsen, 2010).

The stability of PFAAs is due to the strength of the high energy carbon-fluorine bond (531.5 kilojoule per mole [kJ/mol]) (Hudlicky & Pavlath, 1995) and the shielding effect of the carbon backbone conformation (Torres, Ochoa-Herrera, Blowers & Sierra-Alvarez, 2009). Precursor compounds, which will eventually turn into PFAAs, have additional moieties added on to the carbon-fluorine chain where other substances and organisms can attack and degrade them. After this process, all that is remaining is the carbon-fluorine backbone and a headgroup: a PFAA. Thus, most scientists consider PFAAs terminal chemicals because they will not undergo further transformation in the natural environment, and will most likely exist on a time frame longer than can be reliably calculated.

There has been one controversial study showing slight degradation of PFAAs under extreme natural conditions (Taniyasu, et al., 2013; Wang, Cousins, & Scheringer, 2015). There has also been successful decomposition of PFAAs in the lab using experimental techniques (Luo, Lu & Zhang, 2015; Luo, Yan, Lu & Huang, 2018; Trojanowicz, Bojanowska-Czajka, Bartosiewicz & Kulisa, 2018) such as fungal treatment (Tseng, 2018) and high temperature reaction with persulfate (Park, Lee, Medina, Zull & Waisner, 2016). However, current research suggests that all PFAS ever produced will either transform into a PFAA and never degrade, or will itself not degrade under common conditions in the environment.

The time frame for the transformation from precursor to PFAA depends on the compound and the conditions. Half-lives are not known or studied for most precursors, with some calculated values ranging from hours to more than a thousand years (Figure 26) (Dassuncao, Hu & Zhang, 2017; Rankin, Lee, Tseng & Mabury, 2014; Wang, Huang & Yang, 2013). With the vast number of potential starting materials and environments, the exact mechanism and changes that occur for each precursor is unknown. However, scientists have studied many changes in the laboratory, and are starting to characterize transformations in the field. The result of these ongoing transformation processes is that the presence and amount of certain PFAS will evolve over time at any one specific sampling location.

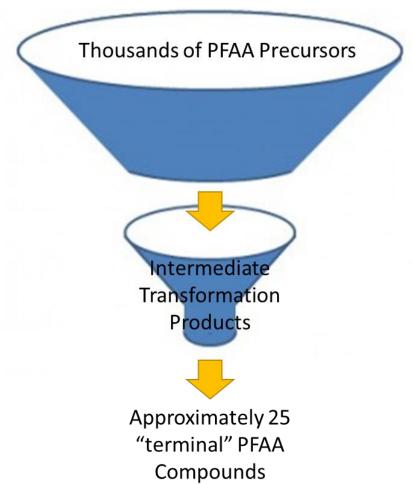


Figure 25. Illustration of precursor transformation leading to PFAAs.

Figure 26. Examples of precursor aerobic biotransformation to PFAAs with half-lives (as described in Section 4.1).

4.1.1 Abiotic transformation

Abiotic transformation (transformation without living organisms) can form both perfluoro-sulfonic acids (PFSAs) and perfluoro-carboxylic acids (PFCAs)—the two main types of PFAAs—from a wide range of precursors. For example, reaction with hydroxyl radicals gives N-methyl perfluorobutane sulfonamidoethanol (NMeFBSE) a half-life of two days, and creates both sulfonic and carboxylic acid byproducts (D'Eon, Hurley, Wallington & Mabury, 2006). Additionally, fluorotelomer alcohol (FTOH) compounds have been found to transform in the atmosphere through reactions with chlorine and hydroxyl radicals to form PFCAs (Ellis, Martin & De Silva, 2004; Ellis et al., 2003). N-ethyl perfluorobutanesulfonamide (NEtFBSA) can transform into PFCAs through a similar mechanism in the atmosphere, with a lifetime in air of 20 – 50 days (Martin, Ellis, Mabury, Hurley & Wallington, 2006).

4.1.2 Biotic aerobic transformation

Researchers have demonstrated aerobic biotransformation (transformation by organisms with access to oxygen) of PFAA precursors several times, and this type of modification is probably the most prevalent form of PFAS chemical transformation. Several studies have been performed with focus on transformation that may occur at WWTPs or aqueous film forming foam (AFFF)-impacted sites (Arvaniti & Stasinakis, 2015). Laboratory studies have shown degradation of FTOHs into PFCAs. In addition, N-ethyl perfluorooctane sulfonamido ethanol (N-EtFOSE) is biodegraded into perfluorooctane sulfonate (PFOS) with a half-life of 0.7 – 44 days (Benskin et al., 2013; Mejia-Avendaño & Liu, 2015; Rhoads, Janssen, Luthy & Criddle, 2008; Zhao, Ma, Fang & Zhu, 2016) and perflurorooctaneamido quaternary ammonium salt (PFOAAmS) transforms into perfluorooctanoic acid (PFOA) with a half-life of 142 days (Mejia-Avendaño, Duy, Sauvé & Liu, 2016). Perfluoroacylphosphates (POPs) have also been shown to biodegrade into FTOHs and eventually to PFCAs (Lee, D'Eon & Mabury, 2009). All precursors tested have shown the ability to be aerobically biotransformed to PFAAs, with most perfluoroalkane sulfonyl fluoride (PASF)-based substances eventually being biotransformed into PFSAs while all FTOH based substances are eventually transformed into PFCAs (Martin, Ellis, Mabury, Hurley & Wallington, 2006).

4.1.3 Biotic anaerobic transformation

Anaerobic biotransformation (transformation by organisms without oxygen) has been studied much less than aerobic biotransformation. Most evidence suggests that it is slower and transformation into final PFAA forms is less complete. For instance, some PFAA precursors have been shown to remain stable for long periods of time under anaerobic conditions (Boulanger, Vargo, Schnoor & Hornbuckle, 2005; Lange, 2018; Yi, Harding-Marjanovic & Houtz, 2018), with most fluorotelomer sulfonates (FTSAs) remaining more stable than FTOHs (Zhang, Lu, Wang & Buck, 2016). However, in general, anaerobic studies have had similar results to aerobic studies, with PFAAs not biodegrading and other compounds eventually leading to PFAAs.

4.1.4 Consequences of chemical transformation

Because of the transformation processes outlined above, even though U.S. production of PFOS was phased out in 2002 and most production of PFOA was phased out in 2015 through the Environmental Protection Agency's (EPA) PFOA stewardship program, levels of PFAAs have continued to increase in wildlife (Dassuncao, Hu & Zhang, 2017; Roos, Berger, Järnberg, Van Dijk & Bignert, 2013). Manufacturers continue to produce precursor compounds, which will change into PFAAs (including PFOS and PFOA if of sufficient chain length) once released to the environment.

Tracking changes in environmental levels of all PFAS is difficult because there are a large number of precursors, and it is only practical to test for a small fraction in each experiment. Most precursors require advanced analytical methods to detect. These are expensive, available in only a few labs around the world, and often cannot accurately measure quantities of compounds. Additionally, there are very few validated methods endorsed by governmental bodies, so much of the testing done uses experimental techniques. With different methods used by different researchers, comparing results from different studies can be poorly reliable.

Terminal PFAA are the most prevalent and the most persistent type of PFAS, so they have been studied the most. However, the fact that several precursors have measurable levels in both surface waters (Gebbink, Van Asseldonk & Van Leeuwen, 2017; Pan, Zhang & Cui, 2018) and wildlife (Shi at al., 2015) shows that it is not only PFAAs that have to be considered when evaluating impact and risk, since exposure to precursors can be significant. A study in the Baltic Sea found PFAAs and precursors in most aquatic organisms, but concluded that PFAA levels were not necessarily correlated with precursor intake (Gebbink, Bignert & Berger, 2016). This suggests that it is important to evaluate exposure to precursors and PFAAs separately when considering risk.

In another example of precursor exposure, North Atlantic pilot whales do not contain the enzyme to convert perfluorooctanesulfonamide (PFOSA) to PFOS like most animals do, so when they adsorb PFOSA, they are exposed to its effects for much longer than other species (Dassuncao, Hu & Zhang, 2017). Scientists will need to consider the rate of a chemical's transformation to PFAA in addition to the chemical hazards of both the chemical itself and the terminal PFAA to get the full picture of risks involved with use and emission.

The PFAS released to the environment from products and manufacturing operations transform over time into a variety of chemical transformation products. The lifetimes and toxicity of these individual transformation products and the final terminal degradates all contribute to a still uncertain environmental impact.

4.2 Polymeric PFAS

There are three different classes of polymeric PFAS to consider when looking at transformation and hazard: fluoropolymers, side-chain fluorinated polymers, and polymeric perfluoropolyethers.

An important consideration is how (or whether) the polymer backbone may degrade, and what unreacted monomers and catalysts may be present. There is evidence that bacteria or light can degrade some fluorotelomer-based PFAS polymers (side-chain fluorinated polymers). This would release soluble monomer or other PFAS fragments to the environment with a half-life of decades to two centuries (Rankin, Lee, Tseng & Mabury, 2014; Washington, Ellington, Jenkins & Yoo, 2010; Washington & Jenkins, 2015; Washington, Jenkins, Rankin, Naile, 2015; Washington, Rankin, Libelo, Lynch & Cyterski, 2019). However, this finding is still unsettled, due to alternate reports using different methods, which show a half-life of approximately 15,000 years for fluorotelomer-based polymers (Russell, Berti, Szostek & Buck, 2008; Russell, Wang, Berti, Szostek & Buck, 2010).

The finding of a half-life of thousands of years for side-chain fluorinated polymers is of note because it contrasts with degradation times for similarly structured monomers, which have half-lives of days to years. If side-chain fluorinated polymers—which are often used as oil—and water-resistant treatment for consumer products degrade, then they could be a potential source of PFAS emissions for decades or centuries if not properly disposed and contained in landfills (Li, Liu, Hu & Wania, 2017). One study suggests that degradation of polymers could increase PFAS loading to the environment by four to eight times in coming years (Washington & Jenkins, 2015).

Intact fluoropolymers and perfluoropolyether polymers (PFPEs) are generally agreed to be inert and not bioavailable or bioaccumulative, suggesting minimal health impact (Henry, Carlin & Hammerschmidt, 2018). PFPEs have thermal, chemical, photochemical, hydrolytic, oxidative, and biological stability (Buck & Korzeniowski, 2018). Polytetrafluoroethylene (PTFE) and PFPEs are practically insoluble in water and hydrocarbons, and not subject to long-range transport. However, as identified in Appendix 1: Chemistry, Section 1.4.5, the use of non-polymer processing aids during the application of PTFE coatings has also been a source of PFAA emissions into the environment.

In addition to polymer degradation as a source of PFAS, the polymerization of PFAS polymer requires the use of monomers and, in some cases, non-polymer processing aids. These may be a source of PFAS emissions into the environment. In the past, PFOA was used as a processing aid in fluoropolymer manufacture (Prevedouros, Cousins, Buck & Korzeniowski, 2006; Hopkins, Sun, DeWitt & Knappe, 2018). Manufacturers have since switched to chemicals thought of as safer, such as or ammonium 4,8-dioxa-3H-perfluorononanoate (ADONA) and hexafluoropropylene oxide (HFPO) dimer acid (GenX) (Gordon, 2011). Although these substitutes are not used in the final polymer, they have been detected numerous times

worldwide, including in drinking water in North Carolina and the Netherlands (Gebbink, Van Asseldonk & Van Leeuwen, 2017; Pan, Zhang & Cui, 2018; Song, Vestergren, Shi, Huang & Cai, 2018).

4.3 Emission sources

PFAS can be released into the environment in their manufactured form, as ambient air emissions, in aqueous solution to water sinks, or in solid form which can later be subject to degradation. Emissions can result from:

- Location of PFAS manufacturing.
- Locations where PFAS are used in manufacturing other products.
- Use of the products containing PFAS.
- Locations where wastes containing PFAS are stored.
- Degradation of PFAS released to the environment.
- Inadvertent releases to the environment via uncontrolled spills, improper burial, or dumping.

Properties of an individual PFAS will affect its solubility in water, adsorption to soil, or ability to exist as a gas. These attributes will affect the rate of transport when released to environmental media. Chemical changes caused by environmental exposure further complicate the rate of transport. An emitted compound may initially have more affinity for one type of media, but as time passes, it may change and be more likely to migrate and exist in another.

The subsections below address general mechanisms of environmental PFAS fate and transport. <u>Appendix 5: Environmental Occurrence</u> provides information on levels of PFAS measured in environmental media in Washington.

4.3.1 Air

During direct or secondary manufacturing, PFAS can be released to the ambient air through uncontrolled stack emissions (National Ground Water Association (NWGA), 2017). Such emissions can occur in the gaseous phase, or as an aerosol in small particles.

Anionic forms of PFAS, such as PFCAs at low pH, are more likely to be adsorbed to particulates in the air (Ahrens et al., 2012; NWGA, 2017). Once in the air, PFAS can travel large distances before deposition. Deposition occurs via settling of particulates or by transformation of gaseous phases into non-volatile compounds. Deposition can occur either by dry deposition (particles landing by themselves) or by wet deposition (precipitation contributing to deposition) (Taniyasu, Yamashita & Moon, 2013).

Short-range air transport causes PFAS distribution to be much more extensive than just water-based transport, which is the focus of most concerns from manufacturing plants and regulators. Air-based transport can cause contamination of soil, groundwater, and surface water that

otherwise would not be anticipated from merely looking at water flow. Long-range air transport is responsible for the wide distribution of PFAS across the globe, as shown by their occurrence vast distances from all manufacturing sites, including both the Arctic and Antarctic. In addition, sea spray may help re-aerosolize PFAS that have been deposited in oceans (Armitage, Macleod & Cousins, 2009; Gouin & Wania, 2007), contributing to further air-based transport.

Most PFAS are not very volatile, but those that are (like fluorotelomers, FTIs and FTOHs, and perfluoroalkane sulfonamides (FASA)) may partition from liquid to gaseous phases (Buck et al., 2011). This has been identified as an important transformation mechanism in landfills, resulting in landfill gas emissions (see Appendix 3 Sources and Uses, Section 3.4.3 Landfilled Products). PFAS off-gassing from consumer products has also been confirmed, as well as migration of PFAS-containing particulate from products into indoor air in both domestic and occupational settings (Buck et al., 2011). See Appendix 7: Health, Section 7.2.2 Populations with elevated PFAS exposure).

4.3.2 Water

Release to aqueous media

In many cases, PFAS manufacturing processes involve aqueous solutions, which are often a mixture of compounds. Environmental release to aqueous media resulting from manufacturing or secondary manufacturing activities can occur when industrial wastewater is discharged to surface water, or when liquid phase PFAS are directly released to ground or surface water without pre-treatment. Neither the state nor federal Clean Water Act (CWA) regulate PFAS in industrial wastewater discharges via numeric standards.

Certain industrial wastewater discharges are sometimes routed to WWTPs. Such discharges require pre-treatment permits, however these permits also do not regulate PFAS. WWTPs, in turn, discharge treated wastewater to surface water.

PFAS can be present in sewage as a result of products containing PFAS being used in residential, commercial, and institutional facilities, and disposed of in domestic wastewater (see <u>Appendix 3: Sources and Uses, Sections 3.3</u>). As discussed in <u>Appendix 3: Sources and Uses, Section 3.4.1</u>, PFAS has been found in both WWTP influent and effluent. This has been recognized as one of the larger emission sources for PFAS (NWGA, 2017).

Similarly, PFAS can also be present in domestic wastewater effluents released to domestic onsite wastewater systems, which typically discharge to groundwater. For example, a study conducted in Cape Cod, Massachusetts, correlated the presence of PFAS in domestic drinking wells with septic system leachate sources that contributed PFAS to local groundwater concentrations (Schaider et al., 2016).

Improper storage of base or secondary manufactured PFAS-containing products can result in leaching of PFAS when exposed to water. Legacy disposal of PFAS wastes in areas not classified

as landfills has resulted in groundwater contamination in numerous locations in the U.S. (EPA Enforcement and Compliance History [ECHO], 2020a).

Due to the high solubility of many PFAS (see <u>Appendix 1: Chemistry</u>, <u>Section 1.2.4</u>), compounds may be susceptible to leaching from landfills and contaminated biosolids, compost, and soils when exposed to water (Hamid & Grace, 2018; Kim, Li, Grace, Benskin & Ikonomou, 2015; Lang, Allred, Field, Levis & Barlaz, 2017; Lang, Allred, Peaslee, Field & Barlaz, 2016). Leachate from PFAS-contaminated landfills is estimated to contain around 600 kg per year of PFAS in the U.S. (Lang, Allred, Field, Levis & Barlaz, 2017). In Washington, leachate from some landfills is collected and either sent to a WWTP or deposited in evaporation ponds. Landfill leachate has been identified as an important source of PFAS that is redistributed into the environment, especially when leachate is sent for treatment in WWTPs (NWGA, 2017).

Other transport mechanisms include deposition of PFAS aerosols into water bodies as described above, as well as release of PFAS to deposited snow and soils as a result of wax shed from skis during ski competitions (Plassmann & Berger, 2013).

Firefighting using AFFF represents a large source of release of water-based PFAS mixtures into the environment through runoff into surface and migration to groundwater, as discussed in Appendix 3: Sources and uses, Section 3.2.

Many PFAS transport easily through groundwater and surface water due to their high solubility. Dispersion, diffusion, and advection will all affect the movement of PFAS in water, but generally, the compounds will follow the water flow. It is estimated that the oceans are the main final sink for PFAS (Armitage, et al., 2006).

Soil interactions

One important process that affects PFAS transportation and can complicate water transport is adsorption to organic compounds. Most PFAS have a fluorinated carbon tail, which is both hydrophobic and lipophobic, and a polar headgroup, which is hydrophilic. Depending on the types of tail and headgroup, properties of the compound will change. This means that different PFAS can have significantly different attraction to both water and soil. Hydrophobic, lipophobic, and electrostatic interactions will all influence the affinity for different phases. Due to the differences in the chemical and physical properties between the head and the tail, PFAS will often localize at phase interfaces, such as soil and water and water and air boundaries (Brusseau, 2018; Guelfo & Higgins, 2013).

Individual PFAS will adsorb to organic carbon in soil to varying degrees using hydrophobic interactions or electrostatic interactions with minerals.⁵⁸ Scientists have mostly studied this interaction in PFAAs, which are relatively soluble in water over a wide range of pH. Because of this solubility, they move easily through water flow, either in groundwater, surface water, or through leaching. However, water transport can be slowed by association with organic carbon in soil (Guelfo & Higgins, 2013; Higgins & Luthy, 2006).

PFSAs tend to adsorb more strongly to soil than PFCAs do (Guelfo & Higgins, 2013; Higgins & Luthy, 2006) and thus are less mobile. Longer carbon chain lengths are also generally associated with increased adsorption relative to shorter chains (Guelfo & Higgins, 2013). This indicates that a partitioning may occur during wastewater treatment. Shorter chains tend to leave the effluent and longer-chained compounds are more likely to stay in the solid fraction.

Adsorption of PFAS to soil increases as the soil's total organic carbon percentage increases. Soil type and its organic carbon content directly affects the leaching rate (or retention time) of PFAS when spilled on the ground such as during firefighting or training with AFFF. In addition, the chemical constituents of the flammable materials onto which AFFF is applied may influence transport of PFAS through soil and groundwater. Spills into coarse, poorly aggregated soils (such as drainage ditches) will likely leach PFAS faster compared to soil with good structure and high organic carbon.

The retention time of PFAS in soil is dependent upon numerous site specific variables, though there is evidence that desorption is often incomplete (Chen, Reinhard, Nguyen & Gin, 2016). Soil contaminated with PFAS may remain as a low volume source of contamination for ground and surface water for a long time, complicating hazard assessment.

4.3.3 Solids

Solid phase PFAS resulting from secondary manufacturing, domestic, commercial, and institutional product use (see Appendix 3: Sources and Uses) can be disposed of in solid waste landfills or, in the case of food packaging materials, recycled in composting facilities (Kim, Li, Grace, Benskin & Ikonomou, 2015; Choi et al., 2019). Waste containing PFAS at concentrations above 100 parts per million (ppm) designates as a state-only dangerous waste and must be disposed of as such. Such solid phase PFAS contaminants can serve as potential future sources for emission if exposed to environmental degradation conditions.

Solids contained in sanitary effluents can contain PFAS resulting from human ingestion of PFAS. Sanitary solids are disposed of in WWTP biosolid sludges, or as solid or liquid mixtures removed from on-site sanitary systems, which are then typically transferred and discharged to local WWTP or other appropriate treatment location. Biosolids resulting from treatment of sewage effluent in WWTPs are also known to contain PFAS.

Farmers often use compost as well as biosolids from WWTPs as amendments for agricultural soils. In the U.S., solid sewage sludge from WWTPs not used as biosolids is landfill disposed or incinerated. PFAS present in biosolids and compost applied to agricultural lands can leach and travel (Gottschall, 2017). PFAS that have leached can also be available for plant uptake as described in Section 4.3 below. Biosolids have been identified as a significant source of PFAS emissions (NWGA, 2017).

<u>Appendix 8: Biosolids</u> provides a more detailed discussion of biosolids application and risk assessment. See <u>Appendix 3: Sources and Uses, Section 3.4.3 Landfilled products</u> for additional discussion of landfilled solids and leachate.

4.4 Uptake by living organisms

Living organisms exposed to PFAS compounds in the ambient or built environment may ingest or otherwise absorb these chemicals via exposure to PFAS in various types of media. Due to the persistence and ability to transport large distances, animals do not need to be near sources of PFAS to show bioaccumulation (Roos et al., 2013). Plants have been shown to take up some types of PFAS from the soil (Blaine et al., 2013; Blaine, Rich & Sedlacko, 2014; Scher, 2018), an issue of concern since agricultural fields have the potential to be treated with PFAS-contaminated biosolids from WWTPs or PFAS-contaminated compost materials. Organisms in the natural food chain can also ingest PFAS directly in water they drink, or indirectly via PFAS present in their prey, with higher levels of PFAS appearing in animals higher on the food chain (Ahrens & Bundschuh, 2014; Houde, Silva & Muir, 2011). Appendix 6: PFAS Ecotoxicology, Section 6.2 Bioaccumulation, addresses bioaccumulation of PFAS in additional detail. Appendix 8: Biosolids addresses the potential for plant uptake of PFAS from contaminated soils.

Human beings can likewise ingest PFAS after handling or coming into contact with products that contain PFAS, drinking PFAS-contaminated water, or eating foods where PFAS is present. As identified above, some PFAS, especially shorter-chain PFAAs, may be taken up by food plants growing in contaminated soils, biosolids, or water. Those PFAS that do bioaccumulate will build up in livestock and fish when present in their food or water (Kowalczyk, 2013; Michigan Department of Community Health, 2012; New Jersey Department of Environmental Protection, 2018; van Asselt et al., 2013; Vestergren et al., 2013). PFAS may also migrate into food from coated food wrappers, fast food containers, microwave popcorn, and non-stick baking papers (Begley et al., 2005; European Commission, 2012; Geueke, 2016). Impacts of human exposure to PFAS are further addressed in Appendix 7: Health, Section 7.2 PFAS exposure in people.

4.5 Long term PFAS management

It is beyond the scope of this document to discuss in detail the methods available for management or mitigation of PFAS in environmental media. Mitigation goals and approaches are site-specific and tailored to address each site's PFAS mixture loading as well as financial and technological resources available to reduce PFAS risks to sensitive populations. Financial considerations include both short term capital costs and long term operation and maintenance costs for the life of the treatment system.

Prior to implementing any one technology, the context of all mitigation strategies has to be considered, including the possibility to stop or remove the PFAS source altogether, or, for example in the case of drinking water, to find alternative, non-contaminated sources. The

following summarizes the most prevalent remediation techniques available (EPA, 2019; Interstate Technology Regulatory Council (ITRC), 2020; NWGA, 2017). Often more than one technique is necessary to achieve intended remediation goals.

4.5.1 Removal of PFAS from water

Much effort has been focused on remediation of those PFAS that have been identified in drinking water systems through the third unregulated contaminant monitoring rule (UCMR3) data collection. This exercise has underlined that consideration of PFAS precursors is very important to implementation of long-term remediation solutions, as remediation efforts could cause precursor compounds to degrade to more stable PFAS, such as PFOA and PFOS (NGWA, 2017). Conventional water treatments, such as low pressure membranes, biological treatments, disinfection, oxidation, and advanced oxidation, have proven to be ineffective at removing PFAS from water (EPA, 2020b; Ozekin & Fulmer, 2019). Technologies identified to-date to remove PFAS from water include sorption onto granular activated charcoal (GAC), ion exchange (IX), and membranes.

Sorption on to GAC has proven removal efficacies to reduce PFAS concentrations down to and below EPA's Health Advisory Levels and state drinking water guidelines or established levels. GAC systems can be designed to meet the needs of various sizes of drinking water purveyors. PFAS sorb to the GAC substrates. However, as flow through GAC materials proceeds, sorption sites are used up and "breakthrough" can occur. GAC must then be changed out, which introduces an important maintenance cost for these systems. Some GAC can be regenerated for re-use. GAC are more effective for capturing long-chain PFAS—short-chain PFAS breakthrough GAC systems much more rapidly. These systems therefore need to be designed based on the entire mixture of PFAS present in the influent. Biochar, a carbon-rich, porous solid synthesized from biomass through high-temperature, low-oxygen pyrolysis is also being investigated as a substitute for GAC.

Ion exchange (IX) technologies use synthetic, polymeric sorbent media to remove PFAS from water. Similar to GAC, as the IX media sorption sites become used up, breakthrough will occur. IX media is available in both non-regenerable (single use) and regenerable (multi-use) versions. Single-use media is disposed of through landfilling for example. Multi-use media can be treated to remove the PFAS it has collected, so that the media can be placed back into service. Regeneration, however, creates a concentrated PFAS liquid waste stream. IX technologies have been used for many other types of contaminants and are scalable for various types of applications. They can be designed to capture long- and short-chain PFAS.

Reverse osmosis and nanofiltration are two high pressure membrane technologies. Both involve mechanisms to filter out large PFAS molecules from water. Although they can be very effective even on the smallest PFAAs, they have not been tested beyond bench scale. Both are expensive and result in treatment concentrate waste streams that have to be disposed of (ITRC, 2020; Ozekin & Fulmer, 2019).

Researchers continue to investigate other technologies such as precipitation or flocculation and redox manipulation (ITRC, 2020). Although these have shown promise at the bench scale, they are not fully developed technologies.

<u>Appendix 10: Economic Analysis, Section 10.1 Costs of recommended actions</u>, provides illustrative examples of costs borne by water suppliers in the state to mitigate PFAS in drinking water supplies.

4.5.2 Stabilization of PFAS in soils

As discussed above in Section 4.3.2, leaching from soil to groundwater generally decreases with increasing chain length, but depending on the specific soil conditions, longer chain compounds (such as PFOA and PFOS) can readily migrate through the unsaturated zone soils and into groundwater. Sorption and stabilization technologies reduce or remove the potential for PFAS to mobilize from soil to groundwater. These technologies involve amendments such as activated carbon and carbon nanotubes (CNTs), resins, minerals, biomaterials, and molecularly imprinted polymers that are added to soils. The amendments bind to PFAS and thus reduce their release from soil. Use of activated carbon has the potential to limit leaching of PFAS from soil to groundwater, but can be influenced by the presence of co-contaminants, chain length, and the PFAS functional group. The other methods are promising but should be evaluated using laboratory testing with site-specific contaminants and soil types before proceeding to full scale implementation.

4.5.3 Ultimate disposal

Ultimate disposal of residual PFAS or concentrated PFAS wastes can occur via long-term storage or destruction. As addressed in <u>Appendix 9: Regulations, Section 9.1.2 Washington state rules</u>, only certain wastes containing PFAS will designate as dangerous. In the case of long term storage, holding locations have to be designed for long-term protection of PFAS-containing materials or wastes from conditions which can result in future PFAS migration from the storage means of containment and the storage site. For example, if materials are landfilled, they need to include caps to protect the waste from long term water incursion to avoid leaching of the PFAS (ITRC, 2020). Likewise, liners are imperative to collect leachate which might form. Storage design solutions must consider long-term preservation, as the PFAS will outlast many human generations.

Destruction of PFAS by thermal decomposition (incineration) is currently the only other available long-term management solution. Thermal decomposition of PFAS by incineration has been proven effective if sufficiently high temperatures are employed (EPA, 2019). The decomposition involves several carbon-fluorine atomic bond breaking processes, which can gradually reduce PFAS to their elemental constituents. However, improperly controlled incineration conditions can result in the formation of smaller PFAS products or products of incomplete combustion, which may not have been studied and result in unknown long term adverse impacts. Additional research is continuing to better understand thermal PFAS

decomposition processes, byproducts formed, how they can be captured in emissions streams, and the overall efficacy of incineration (EPA, 2019; ITRC, 2020).

Researchers continue to investigate innovative destructive techniques, for example plasma technologies, electrochemical methods, or catalysts paired with ultraviolet light (Jansen, 2019; RTI International, 2020). However, these have yet to be demonstrated to reach desired destruction levels, scaling for actual applications, or technological and financial feasibility.

4.6 Data gaps and recommendations

4.6.1 Data gaps

As a whole, the study of the fate and transport of PFAS within the environment is an ongoing effort, and many data gaps remain to be filled by the scientific community. Persistence of precursors to terminal PFAS is only beginning to be mapped out and studied.

4.6.2 Recommendations

As identified above, several areas of concern regarding the presence and transport of PFAS throughout the environment have emerged, and are the subject of recommendations elsewhere in this Chemical Action Plan (CAP):

- For presence of PFAS in biosolids produced from WWTPs, and impacts resulting from their application in agriculture, see Appendix 8: Biosolids.
- For presence of PFAS in WWTP influents and effluents, and discharges of WWTP effluent to surface water, see <u>Appendix 3: Sources and Uses, Section 3.4.2</u> Wastewater treatment plants.
- For presence of PFAS in landfill leachate, see <u>Appendix 3: Sources and Uses, Section</u> 3.4.3 Landfilled products.
- For contamination of drinking water resulting from use of AFFF, see <u>Appendix 3:</u> Sources and Uses, Section 3.2 Aqueous film forming foam.

The following recommendations also result from the analyses presented in this appendix:

Recommendation 2.1 Establish PFAS cleanup levels for soil and groundwater

- Using existing authority under Model Toxics Control Act, Ecology will develop cleanup levels for PFOA and PFOS (and additional PFAS as appropriate). Ecology will use the SBOH's drinking water standards or other advisories adopted in rule to develop these cleanup levels.
- Ecology will explore methods for investigation and cleanup of PFAS contamination.
- Ecology will conduct monitoring for PFAS compounds in environmental media (soils, surface water, and sediment) and wildlife tissue to identify sources and assess exposure.

- Based on data collected, Ecology will consider developing cleanup levels for individual or mixtures of PFAS in soil, sediment, freshwater, and saltwater to protect ecological receptors.
- In this context, the following activities will be implemented to support activity under the recommendations above:
 - Trophic transfer and bioaccumulation of PFAS compounds should be further evaluated in aquatic and terrestrial food webs to further understand exposure.
 - Selected individual PFAS compounds, as well as common PFAS mixtures, should be evaluated for ecotoxicity in aquatic and terrestrial biota, using both laboratory and field methods.
 - Ecological risk assessment should be performed for PFAS compounds by detailing exposure and effects in order to estimate risks to nonhuman biota.
 - An uncertainty analysis should accompany PFAS ecorisk assessment to promote transparency in the risk assessment and communication processes and to more clearly identify data gaps.
 - Results of these risk assessments should support potential interventions (for example, species protections) and characterization of potential impacts on ecological services.
- Ecology will provide information to interested parties about cleanup efforts.

Recommendation 2.2 Partner with local organizations in communities with contaminated water or contaminated sites

Department of Health will identify local health departments or community-based organizations to address health equity related to contaminated sites in public communications. Health will coordinate with Ecology to distribute funding to those organizations selected for assistance. Health's new Community Engagement Guide¹¹³ may support this effort.

Funded organizations would:

- Address potential health equity issues through culturally and linguistically informed engagement.
- Find trusted messengers or platforms to deliver audience-tested risk communication messages to engage historically overburdened and higher risk populations.
- Support impacted populations in finding their own solutions through collective action and decision-making.
- Engage the community throughout the course of the public health response, source investigation, and site cleanup.
- Invite area residents to actively participate on advisory committees, in site information meetings, and in public decision-making about remediation.
- Aim to remove participation barriers by providing child care, reducing transportation costs, and planning for convenient meetings times at familiar locations.
- When possible, appropriately compensate community advisors for participation—particularly in areas with low-income populations.

 $^{^{113}\} https://www.doh.wa.gov/Portals/1/Documents/1000/CommEngageGuide.pdf$

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List of acronyms

General acronyms

Table 45. Acronyms found in the fate and transport appendix.

Acronym	Definition
AFFF	Aqueous film forming foam
CAP	Chemical Action Plan
CWA	Clean Water Act
ECHO	EPA Enforcement and Compliance History
EPA	United States Environmental Protection Agency
GAC	Granular activated charcoal
ITRC	Interstate Technology Regulatory Council
IX	Ion exchange
NGWA	National Ground Water Association
WWTP	Wastewater treatment plant
UCMR3	Third unregulated contaminant monitoring rule

Chemical names

Table 46. Chemical name acronyms found in the fate and transport appendix, excluding the acronyms listed in the table above.

Acronym	Chemical Name
ADONA	Ammonium 4,8-dioxa-3H-perfluorononanoate
FASA	Perfluoroalkane sulfonamide
FTI	Fluorotelomer iodide
FTOH	Fluorotelomer alcohol
FTSA	Fluorotelomer sulfonate
Gen X	Hexafluoropropylene oxide dimer acid
HFPO	Hexafluoropropylene oxide
HFPO-DA(GenX)	Hexafluoropropylene oxide dimer acid
NEtFBSA	N-ethyl perfluorobutanesulfonamide
N-EtFOSE	N-ethyl perfluorooctane sulfanamido ethanol
NMeFBSE	N-methyl perfluorobutane sulfonamide ethanol
PASF	Perfluoroalkane sulfonyl fluoride
PFAA	Perfluoroalkyl acid
PFAS	Per- and polyfluoroalkyl substances
PFCA	Perfluoro-carboxylic acid
PFHxS	Perfluorohexane sulfonate

Acronym	Chemical Name
PFOA	Perfluorooctanoic acid
PFOAAmS	Perflurorooctaneamido quaternary ammonium salt
PFOS	Perfluorooctane sulfonate
PFOSA	Perfluorooctanesulfonamide
PFPE	Perfluoropolyether polymer
PFSA	Perfluoro-sulfonic acid
POP	Perfluoroacylphosphate
PTFE	Polytetrafluoroethylene

Appendix 5: Environmental Occurrence

5.0 Overview

5.0.1 Findings

In Washington, perfluoroalkyl acids (PFAAs) have been detected in surface waters, groundwater, WWTP effluent, freshwater and marine sediments, freshwater and marine fish, and osprey eggs. Other media types have not been sampled in Washington.

Environmental monitoring in the state has shown that PFAA concentrations are highest in urban surface water and surface waters receiving minimally diluted wastewater treatment plant (WWTP) effluent. Monitoring also suggests that stormwater, WWTP effluent, and aqueous film-forming foam (AFFF) for firefighting use are primary ways that PFAAs are delivered to water bodies.

Perfluorooctane sulfonate (PFOS), and to a lesser extent perfluorodecanoic acid (PFDA), perfluorododecanoic acid (PFDA), perfluoroundecanoate (PFUnA), and perfluorooctanesulfonamide (PFOSA), were widespread in freshwater fish tissue of water bodies in the state. PFOS was measured in urban lake fish tissue at levels that may trigger consumption advisories to protect human health. PFOS and other long-chain PFAAs are also detected in osprey eggs, at concentrations lower than would affect offspring survival, but potentially high enough to reduce hatchability in samples from sites affected by urban sources and WWTP inputs. PFOS and PFOA were the most dominant compounds detected in marine fish, but currently below the concentrations that may trigger consumption advisories.

Environmental concentrations of PFAAs in Washington surface waters, WWTP effluent, and freshwater fish tissue sampled in 2016 were consistent with PFAS levels in other parts of the U.S. not impacted by PFAS manufacturing facilities. Additional sampling in 2018 confirmed that PFAS concentrations in freshwater fish collected from Washington urban lakes are consistent with other urban water bodies in North America. Osprey egg PFAS concentrations measured in 2016 were similar to recent findings in rural osprey eggs collected in Sweden, with the exception of higher concentrations found in the Washington samples near urban or WWTP sources. Compared to freshwater species, concentrations in marine biota from Puget Sound are generally lower than concentrations measured in marine species in other countries.

Environmental monitoring in 2016 suggested that PFAA levels in surface waters and WWTP effluent had decreased since the last round of sampling in 2008. A general shift in PFAA constituents was evident in WWTP effluent samples, with short-chain PFAAs replacing perfluoroctanoic acid (PFOA) as the most dominant compounds in effluent. PFAS concentrations (primarily made up of PFOS) in freshwater fish tissue and osprey eggs have remained unchanged between 2008 and 2016. Insufficient data are available to assess temporal

changes in PFAS in marine and anadromous species. PFOS continues to be a ubiquitous contaminant in Washington aquatic biota.

Data gaps in our understanding of PFAS contamination in Washington's environment include a lack of monitoring ambient groundwater and landfill leachate, assessing sources of PFAS in urban water bodies, and testing PFAS compounds beyond PFAAs.

5.0.2 Introduction

This appendix summarizes the available PFAS data on environmental media collected in Washington state. Relatively few studies have been conducted on PFAS in Washington. PFAS analyses in Washington have generally been limited to the PFAA included in EPA Method 537 (refer to Appendix 2: Analytical Methods for more discussion on PFAS analytical methods). Additional PFAS compounds, including precursors that have known potential to break down into PFAAs, were analyzed in surface water and WWTP effluent samples collected in 2016. Ecology studies discussed below have been conducted following the data quality and acceptance limits included in EPA Method 537. Discussions of data quality can be found in individual references.

As discussed in Appendices 3: Sources and Uses and 4: Fate and Transport, PFAS can be released to the environment during manufacturing, use, and disposal of consumer and industrial products containing PFAS. Currently, the relative importance of different environmental pathways for PFAS transport is not well characterized for Washington state. However, environmental monitoring in Washington shows that PFAA concentrations are highest in water bodies located in urban settings and where WWTP effluent makes up a significant portion of the flow, or hydrologic dilution is minimal. This suggests WWTP effluent, stormwater, and AFFF use are important pathways. Monitoring in the state has focused on releases of PFAS to surface water and the aquatic food chain—the ambient concentrations of PFAS in soils, groundwater, or air have not been investigated.

5.1 PFAS in Washington's environment

5.1.1 Air

Ecology did not identify any studies or analyses of PFAS compounds in Washington's air.

5.1.2 Soil

In 2014, one soil sample from the Moses Lake Port Aircraft Rescue and Firefighting School facility was analyzed for PFAAs and PFOSA, following a release of fire suppressant (Ecology, 2016a). Ecology's Environmental Information Management Database¹¹⁴ includes the results of measured PFAS concentrations in this study. This soil sample had an elevated concentration of

¹¹⁴ https://ecology.wa.gov/Research-Data/Data-resources/Environmental-Information-Management-database

PFO (12,000 micrograms (μg)/gram (g)), perfluoroundecanoic acid (PFUnDA) (1,100 $\mu g/g$), perfluorononanoic acid (PFNA) (120 $\mu g/g$), and perfluorodecane sulfonate (PFDS) (110 $\mu g/g$). Other perfluoroalkyl acids were detected at levels less than 100 $\mu g/g$. Following this sampling event, the impacted soils were excavated and removed from the site for proper disposal.

5.1.3 Groundwater

Ecology did not identify any ambient groundwater monitoring for PFAS in Washington. However, PFAS have been detected in groundwater wells used for drinking water in several areas. <u>Appendix 7: Health, Section 7.4 Known areas of PFAS contamination</u>, describes this sampling in more detail.

5.1.4 Surface water

Ecology's 2008 statewide study

In 2008, Ecology carried out a study measuring PFAAs in a variety of environmental media throughout the state to determine their occurrence in fresh water systems (Ecology, 2010). This study collected fresh water from 14 water bodies in the spring and fall for analysis of 11 PFAAs.

All spring samples contained measurable concentrations of at least one of the target PFAAs, ranging in total perfluoroalkyl acids (summed concentration of PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, PFOS, and PFDS) (T-PFAAs) from 1.11 - 185 ng/L (nanograms per liter) (median = 7.5 ng/L). Fall samples contained detected PFAAs in all but two samples, ranging in T-PFAAs from less than 0.9 - 170 ng/L (median = 3.6 ng/L).

The highest concentrations were found in sites receiving wastewater treatment plant effluent with limited dilution (West Medical Lake and South Fork Palouse River), followed by an urban lake (Lake Washington). The rest of the sites—mid-sized rivers draining a variety of land-use types—had T-PFAA concentrations of 1.0-10 ng/L.

Ecology's 2016 statewide study

Ecology conducted a second statewide study in 2016 to assess changes in concentrations and compound make-up following the 2008 survey (Ecology, 2017). Surface waters from 15 water bodies were collected in the spring and fall for analysis of 12 PFAAs and 13 known or potential precursors to PFAAs. Precursors analyzed included polyfluorinated sulfonamides, fluorotelomer carboxylates (saturated and unsaturated), and fluorotelomer sulfonates. Fewer than half of the surface water samples contained PFAS compounds, with 7 out of 15 sites containing at least one sample with PFAS detections. T-PFAA concentrations (sum of PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, perfluorododecanoic acid [PFDoDA], PFBS, PFHxS, and PFOS) ranged from less than 2 – 153 ng/L (median = <2 ng/L) in the spring and less than 2 – 170 ng/L (median = <2 ng/L) in the fall. Only heavily impacted water bodies had detections—those with significant WWTP inputs or in urban areas. Detection frequencies and total concentrations were generally lower than those of surface water samples collected in 2008 at the same sites.

With the exception of West Medical Lake and South Fork Palouse River samples, surface water PFAA concentrations in 2016 were very similar to PFAA concentrations recently measured in other water bodies lacking point sources collected throughout Michigan, Rhode Island, and New York (MIDEQ, 2015; Zang et al., 2016). All surface water samples were one to two orders of magnitude lower than levels found in surface water impacted by AFFF use or manufacturing facilities in the U.S. (Anderson et al., 2016; MIDEQ, 2015; Newton et al., 2017).

Perfluoroalkyl acids were the primary compound type found in the surface waters. In addition to the PFAAs analyzed, 13 precursors that potentially break down into PFAAs were analyzed in the 30 surface water samples. The only precursor PFAS compounds detected were 8:2 fluorotelomer unsaturated carboxylic acid (8:2 FTUCA), 4:2 fluorotelomer sulfonate (4:2 FTS), and 6:2 fluorotelomer sulfonate (6:2 FTS), which were all detected only once at 1.02, 11.3, and 6.87 ng/L, making up 12%, 100%, and 100% of the total PFAS concentration, respectively.

Figure 27 shows the relative percent contribution of individual PFAS observed in surface water samples. In the water bodies impacted by WWTP effluent (West Medical Lake and South Fork Palouse River), perfluoropentanoic acid (PFPeA), PFOA, and PFHxA were the most dominant compounds, each contributing an average of 24% – 28% of the total PFAS concentration. The urban lakes (Angle, Meridian, and Washington Lakes) were dominated by PFOS first, and then by the compounds seen in the WWTP-impacted sites.

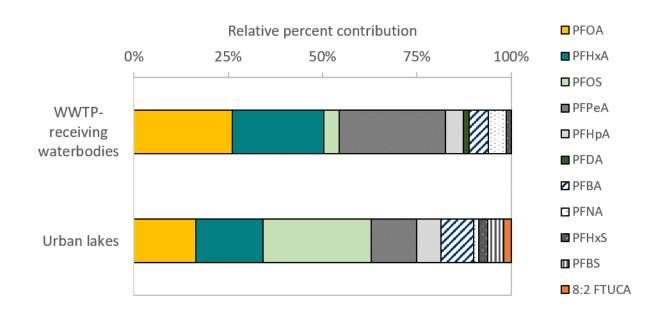


Figure 27. Average PFAS compound profiles in two types of surface waters collected from Washington state water bodies in 2016.

Local source control monitoring

Ecology (2018) analyzed 12 PFAAs in stormwater of urban and industrial catchments in 2017 as part of a larger study to support Ecology's Local Source Control actions. Stormwater was collected twice from seven commercial drainages in Clark County following spring storm events of greater than 0.2 inches (in.) of rain. All 12 PFAAs were detected at nearly every site in the study. Stormwater T-PFAA (sum of detected compounds: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS) concentrations ranged from 31.9 – 114 ng/L. PFOS was measured in the highest concentrations (range: 3.8 – 71 ng/L), followed by perfluorohexane sulfonate (PFHxS) (range: 0.4 – 16.1 ng/L) and PFOA (range: 2.89 – 11.9 ng/L).

Puget Sound study

Dinglasan-Panlilio et al. (2014) measured 14 PFAA compounds in surface water from seven sites in the Puget Sound area, as well as six sites in the nearby Clayoquot and Barkley Sounds in British Columbia, Canada. Samples were collected in spring, summer, and fall of 2009 and 2010, as well as winter 2011. At least one PFAA compound was detected in all samples analyzed. T-PFAA (sum of detected compounds: PFBA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFBS, PFHxS, PFOS, and PFDS) concentrations ranged from 1.5 – 41 ng/L. The highest concentrations were found in two urbanized sites draining to Puget Sound (First Creek in Tacoma and Portage Bay in Seattle). T-PFAA concentrations in marine waters of the Puget Sound were lower than the urban freshwater sites and comparable to levels measured in the more remote sampling locations in Clayoquot and Barkley Sounds.

Perfluoroheptanoic acid (PFHpA), PFOA, and PFOS were the most frequently detected compounds in the samples. Individual compound concentrations were not reported.

5.1.5 WWTP effluent

Statewide study, 2008

Ecology's 2008 PFAS survey analyzed 11 PFAAs in effluent of four WWTPs during the spring and fall (Ecology, 2010). All samples contained multiple compounds, with T-PFAAs (sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, PFOS, and PFDS) ranging from 61 – 418 ng/L (median = 218 ng/L) in the spring and 73 – 188 ng/L (median = 140 ng/L) in the fall. PFOA, the dominant compound detected, contributed an average of 36% in the spring and 32% in the fall to the T-PFAA concentration. In spring samples, perfluorohexanoic acid (PFHxA) was the next most dominant compound (average of 28% contribution to T-PFAA concentration), followed by PFPeA (average of 10%). PFHxA and PFPeA had similar percent contributions in the fall samples (16 – 17% of the total).

Statewide study, 2016

Ecology collected effluent from five WWTPs during the spring and fall of 2016 for analysis of 35 PFAS compounds (12 PFAAs and 23 known or potential precursor compounds) (Ecology, 2016b, 2017). Precursors analyzed included polyfluorinated sulfonamides, fluorotelomer carboxylates (saturated and unsaturated), fluorotelomer sulfonates, perfluoroalkyl phosphonates, and polyfluoroalkyl phosphates. Figure 28 shows PFAS concentrations of the individual effluent samples analyzed. PFAS were detected in all WWTP effluent samples analyzed. Spring T-PFAA (sum of detected compounds: PFBA, PFPAA, PFHXA, PFHDA, PFOA, PFNA, PFDA, PFUNDA, PFDoDA, PFBS, PFHXS, and PFOS) concentrations ranged from 42.1 – 107 ng/L, with a median of 68.9 ng/L. Fall concentrations were similar, ranging in T-PFAAs from 41.8 – 125 ng/L, with a median of 71.4 ng/L. The PFAA concentrations from all WWTPs sampled were within the range found in other recent reports of municipal WWTP effluent in the U.S., but much lower than concentrations found in effluent samples that treat wastewater containing AFFF (Appleman et al., 2014; Houtz et al., 2016).

PFAAs were the primary PFAS compound type found in the effluent samples. Only four of the precursor compounds were detected: PFOSA, bis(perfluorohexyl) phosphinate (6:6 PFPi), bis(1H,1H,2H,2H-perfluorooctyl) phosphate (6:2 diPAP), and bis(1H,1H,2H,2H-perfluorodecyl) phosphate (8:2 diPAP), which ranged in concentration from 2.8 – 19.3 ng/L, contributing 6 – 26% of the total PFAS concentration in individual samples. Overall, PFHxA was the most dominant compound in effluent samples (average contribution of 27%), followed by PFPeA (average of 22%), and PFOA (average of 16%). The low detection frequencies of precursor compounds seen in the Washington WWTP effluent was similar to the low number of precursors detected in effluent collected in California (Appleman et al., 2014).

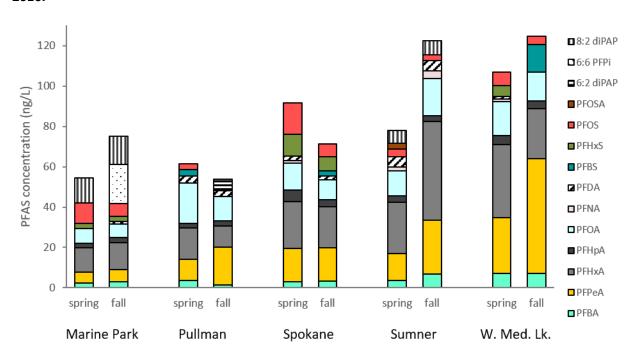


Figure 28. PFAS concentrations (ng/L) in wastewater treatment plant effluent samples collected in 2016.

Note: Results below quantitation limits are excluded from figure.

Figure 29 displays T-PFAA concentrations of WWTP effluent samples collected from the same facility in 2016 compared to 2008. T-PFAA concentrations in effluent samples collected in 2016 were consistently lower than T-PFAA concentrations measured in 2008 by Ecology (2010) at the same WWTPs. A general shift in the composition of PFAS compounds was evident in the WWTP effluent samples as well, with the percent contribution of PFOA decreased in all samples, while the percent contribution of short-chain compounds increased: PFHxA, PFPeA, and perfluorobutanoic acid (PFBA).

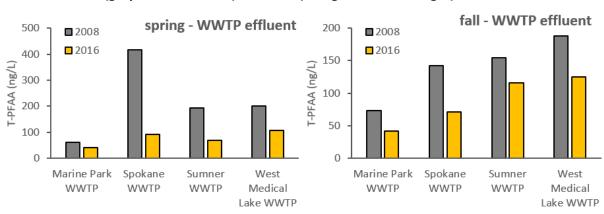


Figure 29. Total perfluoroalkyl acid (T-PFAA) concentrations in wastewater treatment plant effluent collected in 2008 (grey bars on the left) and 2016 (orange bars on the right).

Control of toxic chemicals in Puget Sound study, phase 3

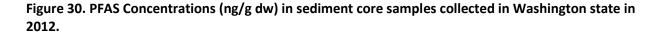
Ecology and Herrera (2010) conducted a study to provide estimates of contaminant loadings to the Puget Sound. The study analyzed 12 PFAAs and PFOSA in effluent from ten WWTPs during the winter and summer of 2009. All ten WWTPs discharged treated effluent to Puget Sound tributaries. Six to ten of the PFAA compounds were detected in every sample. T-PFAA (sum of detected compounds: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS) concentrations ranged from 35.3 – 194 ng/L (median = 73.5 ng/L) in the winter and from 46.3 – 146 ng/L (median = 93 ng/L) in the summer.

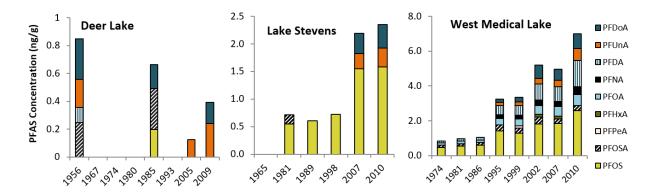
PFHxA, PFNA, PFOS, and PFOA were present in the greatest concentrations and were detected in all samples. Loading estimates for T-PFAAs in the effluents were higher than estimated loadings of T-polychlorinated biphenyls, T-polybrominated diphenyls, and T-polycyclic aromatic hydrocarbons.

5.1.6 Sediments

2012 Sediment cores

In 2012, Ecology collected sediment cores from three freshwater lakes for analysis of 12 PFAAs and PFOSA (Ecology, 2013). Figure 30 displays PFAS concentrations for each dated sediment layer. PFOS and long-chain PFAAs were the dominant compounds measured. T-PFAA (sum of detected compounds: PFBA, PFPAA, PFHXA, PFHDA, PFDA, PFDA, PFDA, PFUNDA, PFDODA, PFBS, PFHXS, and PFOS) concentrations in the surface sediments ranged from 0.392 ng/g dry weight (dw) at the remote Deer Lake to 7.0 ng/g dw in West Medical Lake, which is impacted by WWTP effluent. The urban lake—Lake Stevens—had a surface T-PFAA concentration of 2.35 ng/g dw. T-PFAA concentrations increased from the 1980s to present in the West Medical Lake and Lake Stevens cores. Concentrations and detections were erratic in the rural Deer Lake core.





Local source control monitoring

As part of Local Source Control monitoring, Ecology (2018) collected catch basin sediments over three sampling events in spring and early summer of 2017 for analysis of a suite of parameters that included 12 PFAAs. Sediments were collected from seven urban and industrial catchments during dry-weather events. PFAAs were detected in all sediment samples analyzed. Sediment T-PFAA (sum of detected compounds: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS) concentrations ranged from 0.6 – 17.4 ng/g dw. The maximum concentration measured was of PFDoDA (10.5 ng/g dw) and the maximum PFOS concentration was 9.68 ng/g dw. All other individual compound results were less than 5 ng/g dw.

Marine sediment

Ecology's Marine Sediment Monitoring Program collected sediments from Puget Sound urban bays in 2013 (Elliott Bay), 2014 (Commencement Bay), and 2015 (Bainbridge Basin) for analysis of 12 PFAAs and PFOSA. Ecology's Environmental Information Management Database 115 includes the results measured through this monitoring program. T-PFAA values were not reported. In Elliott Bay, PFOS was detected in seven out of 30 stations, with detected concentrations ranging from 0.24 – 0.48 ng/g dw (Ecology, 2014). PFDA and PFUnDA were detected in one Elliott Bay station, at slightly lower levels. In Commencement Bay sediments, PFOA, PFOS, PFOSA, and PFDoDA were detected at concentrations ranging from 0.11 – 0.57 ng/g dw. Six out of 30 Commencement Bay stations (20%) contained one or more PFAS. In Bainbridge Basin, PFOS was detected in about half of the stations monitored (17 out of 33) and one station also contained detections of PFOSA and PFUnDA. Detected concentrations ranged from 0.11 – 1.6 ng/g dw. The highest concentration of PFOS (1.6 ng/g dw) was found in a sediment sample collected from Sinclair Inlet.

5.1.7 Freshwater fish

Statewide study, 2008

Ecology collected freshwater fish from seven water bodies throughout the state in 2008 for analysis of ten PFAAs (Ecology, 2010). A total of 11 different species were collected and analyzed as a total of 15 composite fillet samples and 15 composite liver samples. Of the PFAAs analyzed, only PFOS, PFDA, PFUnDA, and PFDoDA were detected and quantified. Quantitation limits were fairly high, ranging from 5-25 ng/g. PFOS was detected in 67% of the liver samples (10 out of 15) and 40% of fillet samples (6 out of 15). Concentrations of PFOS in liver samples ranged from less than 10-527 ng/g wet weight (ww) (median = 47.5 ng/g ww). Fillet samples had PFOS concentrations of less than 10-75.5 ng/g ww (median = under 10 ng/g ww). PFDoDA, PFUnDA, and PFDA were each detected once at concentrations of 21.0-46.1 ng/g ww in liver tissue and 5.5-7.5 ng/g ww in fillet tissue.

¹¹⁵ https://ecology.wa.gov/Research-Data/Data-resources/Environmental-Information-Management-database

PBT screening study, 2011

In 2011, Ecology collected common carp and large-scale suckers from Lake Washington, lower Columbia River, Lake Spokane, and the lower Yakima River as part of a screening survey for PBTs (Ecology, 2012). All samples contained PFOS, at concentrations ranging from 2.1 – 19.8 ng/g wet weight (ww) in common carp fillet tissue and from 2.9 – 45.7 ng/g ww in whole body large-scale suckers. PFDA, PFUnDA, and PFDoDA were detected in approximately 80% of the samples, at lower concentrations than PFOS. Other PFAAs were detected infrequently or not at all. T-PFAA (sum of detected compounds: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS) concentrations across both species and sample types ranged from 2.1 – 91.9 ng/g ww, with the highest concentration in the Lake Washington large-scale sucker whole body sample.

Statewide study, 2016

Ecology collected freshwater fish of various species from 11 water bodies in Washington in 2016 (Ecology, 2017) as part of the follow-up study to the 2008 sampling (Ecology, 2010). A total of 22 composite samples of freshwater fish fillet tissue and 22 composite liver tissue samples were analyzed for 12 PFAAs and PFOSA. Eighty-six percent of fillet samples contained at least one PFAS, while the detection frequency for liver samples was 100%. Fillet T-PFAA (sum of detected compounds: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS) concentrations ranged from less than 1 – 87.3 ng/g ww (median = 3.92 ng/g ww) and liver T-PFAS concentrations ranged from 5.12 – 399 ng/g ww (median = 19.3 ng/g ww).

PFOS was the dominant compound in all fillet samples, making up 62 – 100% of the total concentration. PFAA concentrations in the Washington fish were generally much lower than concentrations found near point sources by recent U.S. and Canadian studies, and within the range seen in other water bodies lacking point sources (Gewurtz et al., 2014; Lanza et al., 2017; MIDEQ, 2015).

PFOS concentrations in six of the fillet samples were above the Washington Department of Health's (Health) provisional general population screening level for PFOS in edible fish tissue at the time of publication (23 ng/g). All six fillet samples above the provisional screening level were collected from urban lakes in Western Washington. Seven fillet samples were above Health's provisional high consumer population screening level for PFOS in edible fish tissue (8 ng/g). Only one sample was above the provisional high consumer population screening level, but below the provisional general population screening level. This data was evaluated by Health, but determined to have insufficient sample sizes for a fish advisory assessment.

2018 follow-up study

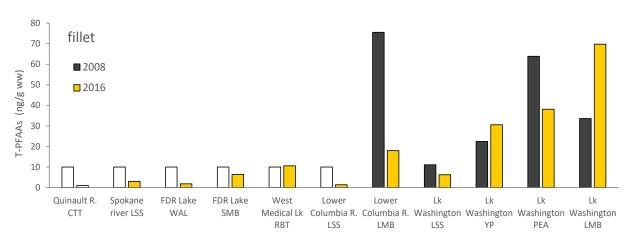
Ecology collected a large dataset of freshwater fish tissue samples from three urban lakes in 2018 to characterize species-specific PFAS concentrations and provide data for Health fish consumption advisory assessments (Ecology, in prep.). A total of 76 composite samples (328)

individual fish) were collected from Lake Meridian, Lake Sammamish, and Lake Washington. PFOS was the dominant compound in all samples analyzed. PFCAs with chain lengths of 9-14 were detected frequently at low concentrations.

Species-specific concentrations were similar for all three lakes. Across all sites, PFOS concentrations of 19.1-50 ng/g ww (largemouth bass), 4.1-19.8 ng/g ww (yellow perch), and 0.5-4.8 ng/g ww (brown bullhead). Smallmouth bass samples were collected from two of the sites and contained the highest PFOS concentrations of the study: 60-99.9 ng/g ww. Cutthroat trout and kokanee were also collected at a subset of the sites and contained PFOS concentrations of 23.9-44.1 ng/g ww (cutthroat trout) and 6.4-7.9 ng/g ww (kokanee). Health is currently updating its screening levels for PFOS and will evaluate this data when screening levels are finalized.

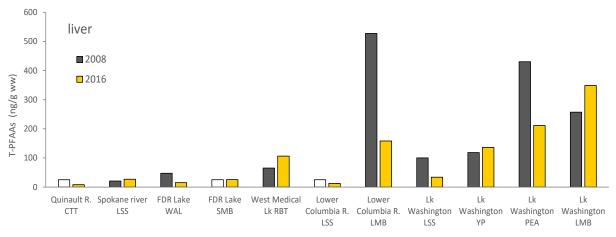
Eleven freshwater fish tissue samples analyzed for PFAS in 2016 had paired species and water body data from 2008. Figure 31 shows PFAS concentrations of fillet composites analyzed in 2016 compared to 2008 and a comparison of liver PFAS concentrations is shown in Figure 32. Of the eleven samples, a difference in quantitation limits hampered comparison in five paired fillet samples and three paired liver samples. The direction of change was mixed for fillet samples greater than the limit of quantitation (LOQ), showing no overall apparent pattern. No consistent increase or decrease over the time period was evident with liver samples either, despite higher detection frequencies.

Figure 31. Total perfluoroalkyl acid (T-PFAA) concentrations in freshwater fish fillet tissue collected in 2008 (grey bars on the left) and 2016 (yellow bars on the right).



Note: White bars indicate PFAS were not detected and the height of the bar represents the limit of quantitation (Quinault River, Spokane River LSS, FDR Lake WAL, FDR Lake SMB, West Medical Lake, and Lower Columbia River LSS).

Figure 32. Total perfluoroalkyl acid (T-PFAA) concentrations in freshwater fish liver tissue collected in 2008 (grey bars on the left) and 2016 (yellow bars on the right).



Note: White bars indicate PFAS were not detected at that concentration (Quinault River and FDR Lake SMB).

5.1.8 Osprey

Statewide study, 2008

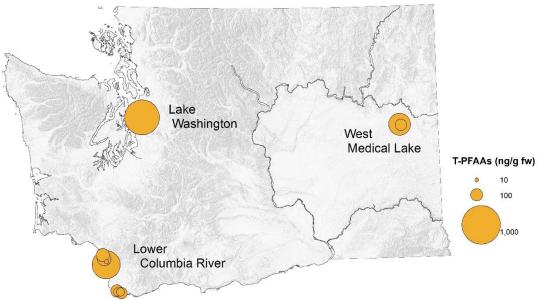
Ecology collected eleven osprey eggs in 2008 from the Lower Columbia River and tested the inner contents (whole egg without shell) for 13 PFAAs (Ecology, 2010). Egg homogenates contained T-PFAA (sum of detected compounds: PFBA, PFPAA, PFHxA, PFHpA, PFOA, PFNA,

PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, PFOS, and PFDS) concentrations ranging from 38-910 ng/g fresh weight (fw) (Ecology, 2010). Similar to fish tissue, PFOS was the dominant compound (range = 23.5-884 ng/g fw; median = 69.0 ng/g fw), followed by PFUnDA (range = 3.5-12.6 ng/g fw; median = 7.8) and PFDA (range = 2.0-10.2 ng/g fw; median = 5.8 ng/g fw). Other acids were detected less frequently and at low concentrations.

Statewide study, 2016

In 2016, Ecology collected osprey eggs from the Lower Columbia River, Lake Washington, and West Medical Lake (Ecology, 2017). A total of 11 osprey eggs were analyzed for 12 PFAAs and PFOSA. All eggs contained at least four PFAA compounds. T-PFAA (sum of detected compounds: PFBA, PFPAA, PFHAA, PFHPA, PFOA, PFNA, PFDA, PFUNDA, PFDODA, PFBS, PFHxS, PFOS, and PFOSA) concentrations ranged from 11.7 – 820 ng/g fw (median = 99.8 ng/g fw). Figure 33 displays the T-PFAA concentration in osprey eggs collected throughout the state. The highest concentration was found in an osprey egg collected from Lake Washington. Two other elevated concentrations were measured in samples collected near WWTP inputs—along the Lower Columbia River and at West Medical Lake. Osprey egg concentrations were similar to recent findings in rural osprey eggs collected in Sweden (Eriksson et al., 2016), with the exception of higher concentrations found in the three Washington samples near urban or WWTP inputs.

Figure 33. Total perfluoroalkyl acid concentrations (ng/g fw) measured in osprey eggs collected in 2016.



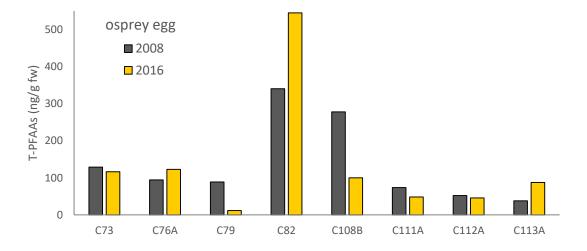
PFOS made up 69-94% of the PFAA burden in the osprey eggs (median concentration = 92.5 ng/g fw; range = 9.08-675 ng/g fw). PFDA, PFDoDA, and PFUnDA were also detected in every sample, each making up less than 10% of the total PFAS concentration. Almost all of the PFAS contamination in osprey eggs was from long-chain compounds, but the short-chain PFPeA was

detected in three samples—all from Lower Columbia River nests. However, PFPeA concentrations were quite low, at 0.45 – 1.83 ng/g fw, and made up less than 2% of the total.

No Effects Concentration of 1,000 ng/g for offspring survival in a top avian predator (Newsted et al., 2005). PFOS concentrations in five of the samples were above a lowest observable adverse effect level (LOAEL) level of 100 ng/g ww for reduced hatchability based on injections in chicken embryos (Molina et al., 2006). These five samples were collected from Lake Washington, West Medical Lake, and Lower Columbia River downstream of the Willamette River confluence. This LOAEL value of 100 ng/g is more conservative, as chicken embryos are more sensitive than wildlife species and another study found higher values for reduced hatchability (Peden-Adams et al., 2009).

Figure 34 displays PFAS concentrations of osprey eggs collected from the Lower Columbia River in 2016 compared to 2008. No consistent change in concentration levels or compound make up was evident between osprey eggs collected along the Lower Columbia River in 2008 and 2016.

Figure 34. Total perfluoroalkyl acid (T-PFAA) concentrations in osprey eggs collected from the Lower Columbia River in 2008 (grey bars on the right) and 2016 (yellow bars on the left).



5.1.9 Marine Biota

Transplanted bay mussels exposed to 18 near-shore locations in Puget Sound from November 2012 through January 2013 were analyzed for a large suite of toxic contaminants, including 13 PFAS, by James et al. (2020). PFAS were largely undetected in the mussel samples. PFOSA was the only compound detected, which was present near reporting limits in only one sample. The authors suggest that PFAS are poorly accumulated by mussels and that mussel tissue PFAS concentrations do not provide a good indication of environmental exposure to PFAS. Muschy et al. (2019) reported PFAS concentrations in mussels worldwide with concentrations typically in the sub ng/g range, which is below the levels of detection in the Puget Sound study.

Meador et al. (2017) analyzed PFAS in two species of fish collected from three sites in Puget Sound. Whole body composites of Chinook salmon and Pacific Staghorn sculpin contained low concentrations of PFOS, with the exception of a Chinook composite collected from Sinclair Inlet, which had a PFOS concentration of 34 ng/g ww. All other PFOS results were less than 2 ng/g ww or not detected. PFOSA was detected in Sculpin collected from the Puyallup Estuary and Sinclair Inlet at concentrations of 2.2 ng/g ww and 0.82 ng/g ww, respectively. PFDA was detected in only one sample, the Sinclair Inlet Chinook composite, at 0.78 ng/g ww.

In addition to the transplanted mussel samples reported by James et al. (2020), Washington Department of Fish and Wildlife (WDFW) analyzed 108 samples of marine fish, a subset of samples collected for their Toxics Biological Observation System (TBiOS), for chemicals of emerging concern, including 12 PFAAs and PFOSA. These samples represent a reconnaissance survey of PFAS in WDFW's monitoring indicator species, from a range of locations (urban to rural) throughout Puget Sound for each species. A total of 74 fillet samples, suitable for assessing potential impacts to human health, were collected, including 44 composite English sole samples from 2017 and 30 individual resident sub-adult Chinook salmon from 2016 and 2017. Samples suitable to assess fish health included 30 whole-body samples (15 composite samples each of juvenile Chinook salmon from 2013 and 2016 and Pacific herring from 2018), and four liver samples of Pacific herring from 2018.

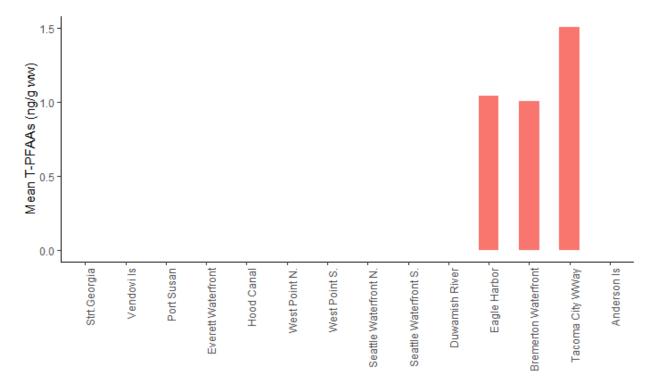
At least one PFAS was detected in 77% of whole body and 100% of the liver samples, whereas the detection frequency for fillet was only 4%. Concentrations of T-PFAA (sum of detected compounds: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, PFOS, and PFOSA) in fillet samples ranged from less than 0.9 - 1.9 ng/g ww (median = 1.0 ng/g ww) for English sole and less than 1 - 3.3 ng/g ww (median = 0.99 ng/g ww) for resident Chinook salmon. T-PFAA concentrations in whole body samples ranged from 1 - 16.1 ng/g ww (median = 0.73 ng/g ww) for juvenile Chinook salmon and less than 0.5 - 2.7 ng/g ww (median = 0.73 ng/g ww) for adult Pacific herring. Liver T-PFAS concentrations in Pacific herring ranged from 1.3 - 5.4 ng/g ww (median = 3.6 ng/g ww).

PFOS was the dominant compound detected in most marine fish samples, ranging from 38% - 100% of the T-PFAA concentrations. Concentrations of PFOS in all fillet samples (0.988 – 14.20 ng/g ww) were below Health's provisional general population screening level for PFOS in edible fish tissue at the time of publication (23 ng/g). Health is currently updating its screening levels for PFOS and will evaluate this data when screening levels are finalized. PFOSA was commonly the dominant PFAA in herring, and was the second most dominant compound detected in other species, ranging from 8% - 100% of the T-PFAA concentration. Concentrations of PFOSA ranged from 0.554 - 2.380 ng/g ww. PFUnA, PFHxA, PFNA, and PFBA were detected less frequently (1 – 5% of all samples) and at lower concentrations, ranging from 0.517 - 1.330 ng/g ww for individual compounds. All other PFAA compounds were not detected.

Overall, average T-PFAA concentrations in fillets of marine species were less than 3 ng/g ww, considerably lower than those measured in fillet of freshwater fish collected in Puget Sound

(Figures 35 and 36). Among fillet samples, PFAA were only detected in three of 14 locations for English sole (i.e., Tacoma City Waterway, Eagle Harbor, and Bremerton Waterfront; Figure 35) and two of eight sample locations for resident Chinook salmon (i.e., South Whidbey Basin/Marine Area 8.2 and South Puget Sound Basin/Marine Area 13; Figure 36).

Figure 35. Mean total perfluoroalkyl acid (T-PFAA) concentrations in English sole fillet collected in 2017.



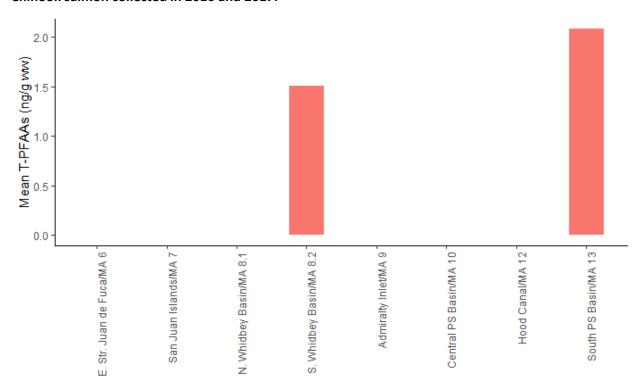
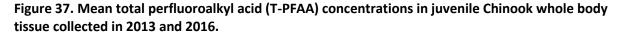


Figure 36. Mean total perfluoroalkyl acid (T-PFAA) concentrations in fillet of sub-adult resident Chinook salmon collected in 2016 and 2017.

T-PFAA concentrations in whole body samples of marine species from specific locations ranged from less than 1.0 – 16.1 ng/g ww for juvenile Chinook salmon (Figure 37) and averaged 0.6 – 1.5 ng/g ww for Pacific herring locations. PFAS were detected in 11 of 15 juvenile Chinook sampling locations, possibly associated with their proximity to freshwater sources, with generally higher T-PFAA concentrations observed in the more urbanized locations near Seattle (i.e., Duwamish River and Elliott Bay nearshore) and Everett (i.e., Port Gardner nearshore). Uniformly low average T-PFAA concentrations were detected in all five Pacific herring sampling locations. Paired whole body and liver samples were collected at four of the five Pacific herring locations, with T-PFAA concentrations in liver two to four times higher than the concentration in whole body herring from the same locations (Figure 38).

Herring liver samples measured in Puget Sound are lower than herring liver samples from the Swedish west coast, which contained PFOS and PFOSA concentrations ranging from 4.06-8.97 ng/g and 6.52-18.2 ng/g, respectively, between 1991 and 2011 (Ullah et al., 2014). Similarly, a study of PFAA concentrations in Pacific Cod fillet from the North Pacific Ocean indicated that PFAA concentrations in the Northeast Pacific Ocean were two to four times less than those in Japanese and Korean waters (Fujii et al., 2019).



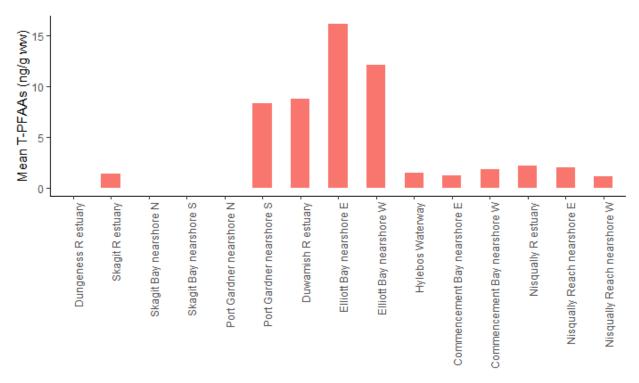
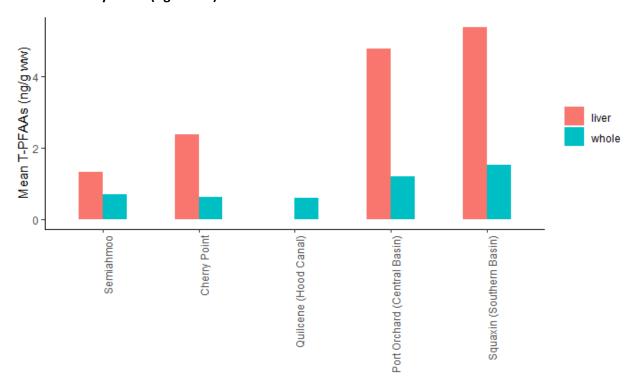


Figure 38. Mean total perfluoroalkyl acid (T-PFAA) concentrations in juvenile Chinook liver (left bars) and whole body tissue (right bars) collected in 2013 and 2016.



5.2 Wildlife studies outside of Washington

PFAS have been detected throughout the world in wildlife types that haven't been sampled in Washington, with PFOS generally detected at the highest frequency and in the greatest amounts. Giesy and Kannan (2001) were the first to report detectable levels of PFOS in a wide range of biota, including species such as bald eagles, polar bears, and seals. Their study included PFOS detections in wildlife from urbanized centers in North America to remote regions of the Arctic and North Pacific Oceans.

Literature reviews done in the mid-2000s confirmed PFOS contamination at all levels of the food chain, and particularly elevated levels in fish-eating animals living near industrialized areas (Houde et al., 2006; Lau et al., 2007). Other perfluoroalkyl sulfonates, long-chain perfluoroalkyl carboxylates, and PFOSA were detected in wildlife such as fish, amphibians, seabirds, and marine mammals (reviewed by Houde et al., 2006). A more recent review by Houde et al. (2011) concluded that PFOS and long-chain PFCAs continue to be widespread in invertebrates, fish, reptiles, aquatic birds, and marine mammals throughout the globe (Houde et al., 2011).

5.3 Data gaps and recommendations

5.3.1 Data gaps

Washington state is lacking data in some key areas for characterizing PFAS contamination in the environment, such as monitoring of ambient groundwater and landfill leachate, source assessments of PFAS in urban water bodies, and testing PFAS compounds beyond PFAAs. With the exception of drinking water wells and military base investigations, no ambient groundwater studies have been conducted in Washington. Around the U.S., PFAA compounds have been found at high concentrations in groundwater near areas of repeated AFFF use, such as airports, oil and gas sites, firefighter training areas, and military bases (Cousins, 2016), but levels of concern may be present in groundwater of other land uses as well.

Environmental monitoring identified Washington urban lakes as sites of elevated PFAA contamination relative to other water body types. The source of this contamination is not fully understood. Research on PFAA contamination in urban water bodies has suggested sources related to automobile and railway transportation may be important (Kim and Kannan, 2007; Zushi and Masunaga, 2009), as well as the transfer of indoor air PFAS loads to the outdoor environment (Gewurtz et al., 2009). An assessment of industrial users of PFAS-containing products in Washington may also contribute to our understanding of sources in the environment.

Recent research using new analytical methods has identified novel PFAS compounds—such as perfluoro-1-butane-sulfonamide (FBSA) and polyfluoroalkyl ether sulfonic acid (F-53B)—in wildlife, though levels have generally been lower than PFOS (Baygi et al., 2016; Chu et al., 2016; Shi et al., 2015). Other novel PFAS, such as cyclic perfluoroalkyl acids and fluorosurfactants,

have been found to accumulate in fish from water bodies directly impacted by AFFF use (Munoz et al., 2017; Wang et al., 2016). Recent research has identified hundreds of new PFAS, many of which have been found in the aquatic environment (Xiao, 2017). Aside from a limited list of precursor compounds measured in surface waters and WWTP effluent in 2016, none of these emerging PFAS compounds have been analyzed in Washington samples.

5.3.2 Recommendations

Filling the data gaps discussed above is recommended to further our understanding of PFAS contamination in Washington's environment. Ecology should conduct sampling in matrices not yet tested, such as ambient groundwater and landfill leachate, as well as conduct source characterization studies in areas of known PFAS contamination, like urban water bodies.

Emerging or novel PFAS which have not been tested in environmental samples should also be sampled. Filling these data gaps will help guide efforts to manage PFAS contamination in the state. Ecology should conduct investigations of areas where contamination is likely to have occurred, but where we currently lack data. These efforts would support local health departments, cities, counties, and other public entities in Washington when PFAS contamination is discovered. Initial investigation efforts could identify areas at high risk of contamination. This could include areas where training or firefighting activities used large quantities of PFAS-containing foam, or where spills released the foam.

Based on the analysis in this appendix, we make the following sub-recommendations as part of Recommendation 2.1:

Recommendation 2.1: Establish PFAS cleanup levels for soil and groundwater:

Ecology will conduct monitoring for PFAS compounds in environmental media (soils, surface waters, sediment) and in tissues of aquatic and terrestrial wildlife to identify sources and to assess exposure. Activity conducted under the following recommendations will serve to further inform our knowledge and understanding of the environmental occurrence of PFAS in the state.

- 1.2 Technical support for site characterization, source investigation, and mitigation at contaminated sites: Investigation of PFAS contamination in groundwater and surface water. See Appendix 7: Health, Section 7.6.2 Recommendations.
- 2.1 Establish PFAS cleanup levels for soil and groundwater: Investigation of PFAS contamination. See Appendix 6: PFAS Ecotoxicology.
- 4.1 Evaluate PFAS in wastewater treatment: Investigation of PFAS in WWTP influent and effluent. See <u>Appendix 3: Sources and Uses</u>.
- 4.2 Evaluate PFAS in landfill leachate and gaseous emissions: Investigation of landfillrelated PFAS emissions. See <u>Appendix 3: Sources and Uses</u>.

• 4.3 Evaluate Washington biosolids management: Investigation of PFAS in biosolids and land application sites. See Appendix 8: Biosolids.

5.4 Washington environmental concentrations data

Notes for Tables 47 through 57:

- Accessed from <u>Ecology's Environmental Information Management Database</u>. 116
- Median concentrations included in parentheses, when available.
- NR = not reported.
- LOQ = limit of quantitation.
- ND = not detected.
- Mult. sp. = multiple species.
- CC = common carp.
- LSS = largescale sucker.
- n = sample size.

Table 47. PFAS concentration ranges in Washington state surface water (ng/L).

Collection year	n	# PFAA compounds analyzed	T-PFAAs range (median)	PFBA range	PFPeA range	PFHxA range	PFHpA range	PFOA range	PFNA range	Reference
Spring	14	11	1.1 – 185	<0.1	<0.1 -	<1.0 -	<1.0 -	<1.0	<0.1	Ecology,
(2008)			(7.5)	- 3.6	26.5	10.5	28	- 96	- 17	2010
Fall	14	11	<0.9 –170	< 0.1	<0.5 –	<0.1 -	<0.9 –	<0.5	<0.5	Ecology,
(2008)			(3.6)	- 5.5	32	37	22	- 48	- 7.0	2010
Four season	13	14	1.5 – 40	NR	-	NR	NR	NR	NR	Dinglasan- Panlilio et
mean										al., 2014
(2009 – 2010)										
Spring	15	12	<2 – 153	<1.0	<1.0 -	<1.0 -	<1.0 -	<1.0	<1.0	Ecology,
(2016)			(<2)	- 13	29	33	13	_	- 5.2	2017
, ,			, ,					42.5		
Fall	15	12	<2 – 170	<1.0	<1.0 -	<1.0 -	<1.0 -	<1.0	<1.0	Ecology,
(2016)			(<2)	- 12	39	32.5	13	– 55	- 5.8	2017

¹¹⁶ https://ecology.wa.gov/Research-Data/Data-resources/Environmental-Information-Management-database

Table 48. PFAS concentration ranges in Washington state WWTP effluent (ng/L).

Collection year	n	# PFAA compounds analyzed	T-PFAAs range (median)	PFBA range	PFPeA range	PFHxA range	PFHpA range	PFOA range	PFNA range	Reference
Spring	4	11	61 – 418	0.7 –	3.8 –	14.5 –	4.1 –	16.5	3.6 –	Ecology,
(2008)			(218)	3.3	31	141	35	- 128	18	2010
Fall	4	11	73 – 188	1.9 –	13 –	11 –	<3.5 –	22 –	5.7 –	Ecology,
(2008)			(140)	5.4	47	30	13	63	14	2010
Summer	10	12	46 – 146	<1.0	<1.0 -	9.6 –	3.4 –	11 –	3.3 –	Ecology &
(2009)			(93)	- 4.9	18	44	9.7	52.5	29	Herrera,
										2010
Winter	10	12	35 – 194	<1.0	<1.5 -	11 –	2.1 –	11 –	1.4 –	Ecology &
(2009)			(73.5)	- 3.6	16	52	10	70	134	Herrera,
										2010
Spring	5	12	42 – 107	2.2 –	5.5 –	12 –	2.2 –	7.2 –	<1.0	Ecology,
(2016)			(69)	7.1	28	36	5.5	20	- 1.9	2017
Fall	5	12	42 – 125	1.6 -	6.1 –	10.5 –	2.6 –	6.6 –	<1.0	Ecology,
(2016)			(71)	7.1	57	49	3.7	18	- 4.0	2017

Table 49. PFAS concentration ranges in Washington state sediment (ng/g dw) (Ecology, 2010; EIM database).

Sample type (collection year)	n	# PFAA compounds analyzed	T-PFAAs range (median)	PFBA range	PFPeA range	PFHxA range	PFHpA range	PFOA range	PFNA range
Fresh water (0 – 2 cm) (2013)	3	12	0.4 – 7.0 (2.35)	<0.2	<0.2	<0.2	<0.2	<0.1 – 0.6	<0.1 – 0.4
Marine (0 - 3 cm) (2013 - 2015)	101	12	П	<0.1	<0.1	<0.1	<0.1	<0.1 – 0.21	<0.1

Table 50. PFAS concentration ranges in Washington state freshwater fish (ng/g ww) (Ecology 2010, 2012, 2017, in prep.).

Sample type (collection year)	n	# PFAA compounds analyzed	T-PFAAs range (median)	PFBA range	PFPeA range	PFHxA range	PFHpA range	PFOA range	PFNA range
Fillet – mult. sp. (2008)	15	10	<10 – 76 (<10)	ı	ı	<5.0	<5.0	<5.0	<5.0
Liver – mult. sp. (2008)	15	10	<25 – 527 (48)	_	_	<10.0	<10.0	<10.0	<10.0
Fillet – CC (2011)	4	12	2.1 – 21.5 (12)	<loq< td=""><td><loq< td=""><td><loq< td=""><td><0.3</td><td><0.25</td><td><0.3</td></loq<></td></loq<></td></loq<>	<loq< td=""><td><loq< td=""><td><0.3</td><td><0.25</td><td><0.3</td></loq<></td></loq<>	<loq< td=""><td><0.3</td><td><0.25</td><td><0.3</td></loq<>	<0.3	<0.25	<0.3
Whole body – LSS (2011)	4	12	3.3 – 92 (23)	<loq< td=""><td><loq< td=""><td><loq< td=""><td><0.2 – 0.6</td><td><0.2 - 0.8</td><td><0.2 – 1.6</td></loq<></td></loq<></td></loq<>	<loq< td=""><td><loq< td=""><td><0.2 – 0.6</td><td><0.2 - 0.8</td><td><0.2 – 1.6</td></loq<></td></loq<>	<loq< td=""><td><0.2 – 0.6</td><td><0.2 - 0.8</td><td><0.2 – 1.6</td></loq<>	<0.2 – 0.6	<0.2 - 0.8	<0.2 – 1.6
Fillet – mult. sp. (2016)	22	12	<1 – 87 (3.9)	<0.5	<0.5- 1.8	<0.5	<0.5	<0.5	<0.5 – 0.9
Liver – mult. sp. (2016)	22	12	5.1 – 394 (20)	<1.0	<0.5 – <2.9	<0.5 – 2.5	<0.5 – 1.1	<0.7	<0.5 – 7.3
Fillet – mult. sp. (2018)	76	13	0.9 – 132 (20)	_	_	<0.5	<0.5	<0.5	ND – 0.5

Table 51. PFAS concentration ranges in Washington state Osprey eggs (ng/g fw) (Ecology, 2010, 2017).

Collection year	n	# PFAA compounds analyzed	T-PFAAs range (median)	PFBA range	PFPeA range	PFHxA range	PFHpA range	PFOA range	PFNA range
2008	11	13	37.5 –	<0.5	<0.5	<0.5 –	<0.5 –	<0.2 -	<0.5 –
			910 (91)			0.8	0.8	<1.0	6.4
2016	11	12	12 – 820	<0.5	<0.4 -	<0.5	<0.5	<0.5	<0.5 –
			(100)		1.8				5.7

Table 52. PFAS concentration ranges in Washington state surface water (ng/L).

Sample type (collection year)	n	PFDA range	PFUnA range	PFDoA range	PFBS range	PFHxS range	PFOS range	PFDS range	PFOSA range	Reference
Spring	14	<0.1	_	_	<0.1	<1.0 -	<0.1	<1.0	_	Ecology,
(2008)		– 4.9			- 0.6	3.3	- 6.5			2010
Fall (2008)	14	<0.1	_	_	<0.1	<0.5 –	<0.5	<0.1 -	_	Ecology,
		- 3.8			- 2.0	4.5	- 7.6	1.3		2010
Four season mean (2009 – 2010)	13	NR	NR	NR	NR	NR	NR	NR	-	Dinglasan- Panlilio et al., 2014
Spring	15	<1.0	<1.0 -	<1.0 -	<2.0	<2.0 -	<2.0	_	<1.0 -	Ecology,
(2016)		- 1.9	<1.2	<1.2	- 2.1	5.3	- 9.2		<2.6	2017
Fall (2016)	15	<1.0	<1.0 -	<1.0 -	<2.0	<2.0 -	<2.0	_	<1.0 -	Ecology,
		- 3.2	<1.1	<1.1	- 13	3.0	_		<1.2	2017
							12.5			

Note for Table 52:

• In each sample, 13 PFAA compounds were analyzed.

Table 53. PFAS concentration ranges in Washington state WWTP effluent (ng/L).

Collection	n	PFDA	PFUnA	PFDoA	PFBS	PFHxS	PFOS	PFDS	PFOSA	Reference
year		range	range	range	range	range	range	range	range	
Spring	4	3.6 –	_	_	<0.1	1.3 –	3.9 –	<0.1	_	Ecology,
(2008)		13			- 1.5	16	31			2010
Fall (2008)	4	3.7 –	_	_	<0.5	2.2 –	9.4 –	<0.1 -	_	Ecology,
		13			- 6.6	12	18	<0.5		2010
Summer	10	1.5 –	<1.0	<1.0	<2.0	<2.0 -	<2.0 -	_	<2.5	Ecology
(2009)		10			- 18	8.3	55			and
										Herrera,
										2010
Winter	10	1.4 -	<1.0	<1.0	<2.0	<1.9 -	<2.0 -	_	<1.0 -	Ecology
(2009)		7.9				6.9	19.5		2.0	and
										Herrera,
										2010
Spring	5	<1.0	<1.0	<1.0	<2 –	<2.0 -	2.6 –	_	<2.5 -	Ecology,
(2016)		- 4.9			3.4	11	16		2.8	2017
Fall (2016)	5	<1.0	<1.0	<1.0	<2.0	<2.0 -	<2.0 -	_	<1.0	Ecology,
		- 5.0			- 14	7.1	6.5			2017

Note for Table 53:

• In each sample, 13 PFAA compounds were analyzed.

Table 54. PFAS concentration ranges in Washington state sediment (ng/g dw) (Ecology, 2010; EIM database).

Sample type (collection year)	n	PFDA range	PFUnA range	PFDoA range	PFBS range	PFHxS range	PFOS range	PFDS range	PFOSA range
Fresh	3	<0.1 –	0.2 - 0.7	0.2 – 0.9	<0.4	<0.4	<0.2 –	-	<0.1 -
water (0 –		1.5					2.6		0.3
2 cm)									
(2013)									
Marine (0	101	<0.1 -	<0.1 -	<0.1 -	<0.2	<0.2	<0.2 -	_	<0.1 –
– 3 cm)		0.14	0.2	0.2			1.6		0.3
(2013 –									
2015)									

Note for Table 54:

• In each sample, 13 PFAA compounds were analyzed.

Table 55. PFAS concentration ranges in Washington state freshwater fish (ng/g ww) (Ecology 2010, 2012, 2017, in prep.).

Sample type (collection year)	n	PFDA range	PFUnA range	PFDoA range	PFBS range	PFHxS range	PFOS range	PFDS range	PFOSA range
Fillet – mult. sp. (2008)	15	<5.0 – 7.5	<5.0 – 7.2	<5	<5.0	<5.0	<10.0 - 76		_
Liver – mult. sp. (2008)	15	<21.0 – 25	<10.0 – 46	<10.0 – 21	<10.0	<10.0	<10.0 - 527	_	_
Fillet – CC (2011)	4	<0.25 – 1.2	<0.25 – 1.3	<0.3 – 1.8	<loq< td=""><td><loq< td=""><td>2.1 – 20</td><td>_</td><td><0.4</td></loq<></td></loq<>	<loq< td=""><td>2.1 – 20</td><td>_</td><td><0.4</td></loq<>	2.1 – 20	_	<0.4
Whole body – LSS (2011)	4	0.3 – 10	<0.24 – 20	<0.2 – 9.5	<loq< td=""><td><loq< td=""><td>2.9 – 46</td><td>_</td><td><0.3 – 3.4</td></loq<></td></loq<>	<loq< td=""><td>2.9 – 46</td><td>_</td><td><0.3 – 3.4</td></loq<>	2.9 – 46	_	<0.3 – 3.4
Fillet – mult. sp. (2016)	22	<0.5 – 5.5	<0.5 – 5.5	<0.5 – 6.0	<1.0	<1.0	<1.0 – 74		<0.5
Liver – mult. sp. (2016)	22	<0.5 – 20	<0.5 – 26	<0.5 – 17	<0.9 – 6.2	<1	1.4 – 336	_	<0.5 – 4.9
Fillet – mult. sp. (2018)	76	ND – 10	0.06 – 11.3	0.11 – 10.6	<0.5	<0.5	0.5 – 99.9	_	

Note for Table 55:

• In each sample, 13 PFAA compounds were analyzed.

Table 56. PFAS concentration ranges in Washington state Osprey eggs (ng/g fw) (Ecology 2010, 2017).

Collection	n	PFDA	PFUnA	PFDoA	PFBS	PFHxS	PFOS	PFDS	PFOSA
year	=	range	range	range	range	range	range	range	range
2008	11	2.0 – 10	3.5 – 13	<5.0 -	<0.5	<0.5 –	24 – 884	<1.0 -	_
				11		1.8		5.8	
2016	11	1.0 – 47	1.1 – 45	0.6 – 47	<1.0	<1.0	9.1 – 675	_	<0.5

Note for Table 56:

• In each sample, 13 PFAA compounds were analyzed.

Table 57. PFAS Concentrations in Puget Sound free-ranging marine and anadromous fish (ng/g ww). (WDFW, in prep.).

Sample type (collection year)	n	T-PFAAs range (median)	PFOS range (median)	PFOSA range (median)	PFUnA range (median)	PFHxA range (median)	PFNA range (median)	PFBA range (median)	PFDoA range (median)
English sole fillet (2017)	44	<0.9 – 1.9 (1.0)	<1.0 - 1.1 (1.0)	<0.5	<0.5	<0.5	<0.5	<0.5 – 0.89	<0.5
Sub-adult resident Chinook salmon fillet (2016 – 2017)	30	<1 - 3.3 (1.6)	<1.0 – 2.4 (0.99)	<0.5 – 0.83 (0.67)	<0.5 – 0.52	<0.5 – 0.78	<0.5	<0.5	<0.5
Juvenile Chinook salmon whole body (2013 and 2016)	15	<1 – 16.1 (2.0)	<1.0 – 14.2 (2.0)	<0.6 – 1.1 (0.94)	<0.5 – 0.58 (0.57)	<0.5 – 0.53	<0.5 – 1.3	<1.5	<0.5 – 0.65
Pacific herring whole body (2018)	15	<0.5 – 2.7 (0.73)	<1.0 – 1.0	<0.5 – 2.1 (0.78)	<0.5	<0.5	<0.5 – 0.6	<0.5	<0.5
Pacific herring liver (2018)	4	1.3 – 5.4 (3.6)	<1.0 – 2.6	<0.6 – 2.4 (1.7)	<0.5 – 1.1	<0.6 – 1.3	<0.6	<0.6	<0.6

Notes for Table 57:

- In each sample, 13 PFAA compounds were analyzed.
- PFPeA, PFOA, PFHxS, PFHpA, PFDA and PFBS were not detected in any samples measured.

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List of acronyms

General acronyms

Table 58. Acronyms found in the environmental occurrence appendix.

Acronym	Definition
AFFF	Aqueous film-forming foam
dw	Dry weight
DOH	Washington Department of Health
EPA	United States Environmental Protection Agency
fw	Fresh weight
g	Gram
in.	Inch
L	Liter
LOAEL	Lowest observable adverse effect level
LOQ	Limit of quantitation
μg	Microgram
ng	Nanogram
ww	Wet weight
WWTP	Wastewater treatment plant

Chemical names

Table 59. Chemical name acronyms found in the environmental occurrence appendix, excluding the acronyms listed in the table above.

Acronym	Chemical Name			
4:2 FTS	4:2 fluorotelomer sulfonate			
6:2 FTS	6:2 fluorotelomer sulfonate			
8:2 FTUCA	8:2 fluorotelomer unsaturated carboxylic acid			
FBSA	Perfluoro-1-butane-sulfonamide			
PFAA	Perfluoroalkyl acid			
PFAS	Per- and poly-fluorinated alkyl substances			
PFBA	Perfluorobutanoic acid			
PFBS	Perfluorobutane sulfonate			
PFDA	Perfluorodecanoic acid			
PFDoA	Perfluorododecanoic acid			
PFDoDA	Perfluorododecanoic acid			
PFDS	Perfluorodecane sulfonate			
PFHpA	Perfluoroheptanoic acid			
PFHxA	Perfluorohexanoic acid			

Acronym	Chemical Name
PFHxS	Perfluorohexane sulfonate
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate
PFOSA	Perfluorooctanesulfonamide
PFPeA	Perfluoropentanoic acid
PFTeDA	Perfluorotetradecanoic acid
PFTrDA	Perfluorotridecanoate
PFUnA	Perfluoroundecanoate
PFUnDA	Perfluoroundecanoic acid
T-PFAA	Total perfluoroalkyl acid (summed concentration)

Appendix 6: Ecological Toxicology

6.0 Overview

6.0.1 Findings

Salient findings in our review on ecotoxicology of per- and polyfluoroalkyl substances (PFAS) include the following:

- PFAS are globally distributed in the environment and biota (e.g., plants, algae, invertebrates, mammals, birds, fish), including locally in Washington state.
- Both short- and long-chain PFAS are environmentally persistent.
- Short-chain PFAS (e.g., perfluorobutane sulfonate [PFBS], perfluorohexanoic acid [PFHxA]) are more water soluble, more volatile, and show greater mobility in the environment (transport in water and air), relative to long-chain PFAS.
- Relative to short-chain PFAS, long-chain PFAS (e.g., perfluorooctane sulfonate [PFOS], perfluorooctanoic acid [PFOA]) bioaccumulate to a greater extent within wildlife species, increasing their likelihood of eliciting adverse toxicological effects (e.g., endocrine disruption, immunotoxicity).
- Bioaccumulation factor (BAF) values in aquatic biota vary by specific PFAS compound, species, and tissue.
- PFOA and PFOS are the most widely studied of the long-chain PFAS in wildlife.
- Biomagnification is observed primarily in aquatic food webs with long-chain PFAS (e.g., marine mammals).
- Perfluoroalkyl carboxylic acid (PFCA) and perfluoroalkyl sulfonic acid (PFSA) bind protein more readily than lipid and accumulate in protein-rich tissues (e.g., liver, blood) in wildlife.
- The proteinophilic nature and apparent deficiency in metabolic biotransformation of PFAS compounds influence their toxicokinetics (e.g., tissue distribution, bioaccumulation), species sensitivity, and biological effects.
- Surrogate species are often used to evaluate PFAS compounds in wildlife (especially for birds and mammals).
- PFAS effects range from subtle alterations in genetic expression to deficits in apical endpoints (e.g., growth, survival, reproduction), which may increase ecological risk (consistent with an adverse outcome pathway (AOP) framework).

6.0.2 Introduction

This appendix summarizes the ecotoxicology of per- and polyfluoroalkyl substances (PFAS). To address this objective, a range of PFAS compounds are included in order to evaluate several bioaccumulation and toxicity endpoints with representative aquatic and terrestrial wildlife species. The ecotoxicology of PFAS includes the following considerations:

• Distribution, concentration, and persistence.

- Bioaccumulation.
- Toxicokinetics (i.e., absorption, distribution, metabolism, and excretion (ADME)).
- Toxicological effects.

Distribution, concentration, and persistence of PFAS in the environment is described in <u>Appendix 5: Environmental Occurrence</u>. As a result, the focus of this appendix is on bioaccumulation, toxicokinetics, and the resulting toxicological responses of biota to PFAS compounds.

PFAS terminology proposed by Buck et al. (2011) is used as overall guidance in this appendix. However, terminology specific to cited articles is not altered to avoid translation errors. In some cases, this conflicts with acronyms recommended by Buck et al. (2011). For example, in the wildlife study authored by Reiner and Place (2015), the PFAS acronym appears to denote perfluoroalkyl acids (PFAA), which include perfluoroalkyl carboxylic acids (PFCA) and perfluoroalkyl sulfonic acids (PFSA). Additionally, terminology presented by Kelly et al. (2009) and Houde et al. (2006a) include perfluoroalkyl contaminant (PFC) and polyfluoroalkyl substance (PFS), respectively, which both appear to denote PFAS.

6.1 PFAS chain length and representative PFAS

6.1.1 Short vs. long-chain PFAS

An early step in the assessment of evaluating the potential risk of PFAS is to group the shortand long-chain substances based on the number of associated perfluorinated carbons.

As described in <u>Appendix 1: Chemistry, Section 1.1.1 PFAS terminology</u>, short-chain PFAS contain up to five perfluorinated carbons terminating with a sulfonate group, or up to six perfluorinated carbons terminating with a carboxyl group (Buck et al., 2011). While resistant to degradation, these substances do not appear to be highly bioaccumulative or to have significant toxicological effects on ecological receptors (EPA, 2017; IMAP 2017a, 2017b, 2017c).

Additional literature suggests that 6:2 FTOH, 6:2 FTAC, and 6:2 FTMAC (considered short-chain PFAS) would not meet the criteria for persistence, bioaccumulation, or toxicity based on the Stockholm Convention on Persistent Organic Pollutants (Ramboll Environ, 2016). As summarized in Tables 60 and 61 (for perfluoroalkyl sulfonates and perfluorocarboxylates, respectively), research findings suggest that biomagnification and bioaccumulation increase as the number of fluorinated carbons also increase (Conder et al., 2008). Although short-chain PFAS are not bioaccumulative, according to regulatory criteria (Conder et al., 2008), high mobility and bioavailability lead to relatively high levels in fish tissues (Shi et al., 2018). Tables 60 and 61 refer to bioaccumulation factor (BAF) and bioconcentration factor (BCF). BAF is calculated as chemical concentration in the organism divided by chemical concentration in the surrounding medium (e.g., food, water), whereas BCF is calculated as chemical concentration in the organism divided by chemical concentration in the organism divided by chemical concentration in water. That is, a BAF accounts for uptake

from all exposure routes, whereas a BCF is limited to uptake from water only. A higher BAF indicates more contaminant accumulates within the organism, relative to environmental media.

Table 60. Perfluoroalkyl sulfonates. Example of the bioaccumulation potential of some PFAS, as related to the number of fluorinated carbons comprising each compound's molecular structure (Conder et al., 2008).

# fluorinated carbons	Compound	Frequency of detection	BAF/BCF Values (L/Kg)	Biomagnification	Bioaccumulative
4	PFBS	Not detected	< 1	No	No
6	PFHxS	Detected in some wildlife	10	No	No
8	PFOS	Detected in most wildlife	18 – 11,000	Possibly	Yes

Table 61. Perfluorocarboxylates. Example of the bioaccumulation potential of some PFAS, as related to the number of fluorinated carbons comprising each compound's molecular structure (Conder et al., 2008).

# fluorinated carbons	Compound	Frequency of detection	BAF/BCF values (L/Kg)	Biomagnification	Bioaccumulative
4-6	PFPn, PFHx, PFHp	Not detected or infrequently detected	<1	No	No
7	PFO	Detected in some wildlife	2 – 570	No	No
8 – 13	PFN, PFD, PFU, PFDo, PFTri, PFT	Detected in most wildlife	100 – 23,000	Possibly	Possibly

As discussed in Appendix 1: Chemistry, Section 1.1.1 PFAS terminology, long-chain PFAS contain seven or more perfluorinated carbons terminating with a carboxylate group, or six or more perfluorinated carbons terminating with a sulfonate group (Buck et al., 2011; OECD, 2013; Wang et al., 2017). These chemicals also resist degradation. Data in Tables 60 and 61 (Conder et al., 2008) limit bioaccumulation to PFAS with eight or more fluorinated carbons. In contrast to short-chain PFAS, recent data confirm that long-chain PFAS tend to be more bioaccumulative and produce adverse toxicological effects to both upland and aquatic ecological receptors, even at relatively low contaminant levels (IMAP, 2017d, 2017e, 2017f, 2017g).

Relative to long-chain PFAS, it is important to note that there appears to be less research available on short-chain PFAS. Although bioaccumulation may be lower, short-chain PFAS are more water soluble and show greater mobility in the environment, relative to long-chain PFAS (Guelfo & Higgins, 2013; ITRC, 2020a; Wang et al., 2015). However, the information presented above indicates that the potential risk of these short-chain PFAS substances (e.g., PFBS, PFPn, PFHx, PFHp) is generally less than that of the long-chain substances (e.g., PFOA, PFOS) to both aquatic and upland ecological receptors. As a result, this review will focus on evaluating the potential risks of long-chain PFAS on ecological receptors.

6.1.2 Representative PFAS substances

As mentioned earlier, the potential risk for ecological receptors is much greater for the general class of chemicals known as long-chain PFAS. It is important to note that most of the information presented in this review is derived from the most commonly detected long-chain PFAS (i.e., PFOA and PFOS). The rationale for using these two specific chemicals as representative of the general class of long-chain PFAS chemicals is:

- PFOA and PFOS are the most widely studied of the long-chain PFAS (e.g., OECD, 2002).
- These chemicals are structurally related, in that one of the defining characteristics that differentiates the chemicals within this class from other classes is chain length (or number of carbon atoms in the molecule).
- The carbon-fluorine bonds are among the strongest in organic chemistry, which renders them practically non-biodegradable and persistent in the environment (Key et al., 1997; Lau et al., 2007; Prescher et al., 1985), including their presence and persistence in:
 - Water.
 - Soil and sediment.
 - o Ambient air.
 - Aquatic and terrestrial wildlife.
- The toxicokinetics and toxicological response for these chemicals appears closely related, depending on species observed (Kelly et al., 2009; Lau et al., 2007; Lindstrom et al., 2011; White et al., 2011).

6.2 Bioaccumulation

Fluorine atoms are substituted for the hydrogen atoms that compose part of the hydrocarbon backbone in PFAS compounds. The fluorine-carbon bonds present in PFAS compounds confer high chemical and thermal stability, which contributes to their persistence in the environment and resistance to natural degradation. Despite commercial and industrial use for more than 50 years, only in the past 20 years have low level detections revealed that PFAS are globally distributed in the environment and biota (Li, 2009; Stahl et al., 2009). Some long-chain PFAS bioaccumulate in the environment and can also undergo biomagnification (Stahl et al., 2011).

As discussed in Appendices <u>3: Sources and Uses</u>, <u>4: Fate and Transport</u>, and <u>8: Biosolids PFAS</u> are released to the environment and transported over various pathways (e.g., soil, water, air), potentially exposing biota. For example, PFOA and PFOS are the major PFAS contaminants found in oceanic waters (Yamashita et al., 2015), and PFCAs (e.g., perfluorooctanoate [PFO], perfluorononanoate [PFN], or perfluorodecanoate [PFD])) have been detected in a variety of wildlife across the globe (Conder et al., 2008; Houde et al., 2006b; Tao et al., 2006). PFCA tissue residues confirm the widespread presence of these compounds from multiple sources (Prevedouros et al., 2006).

PFAS stability and water solubility have allowed for considerable transport through marine environments (Yamashita et al., 2005). Many studies report concentrations of PFAS in marine organisms (e.g., Giesy & Kannan, 2002; Houde et al., 2011). These studies found significant levels of several PFAS (including PFOS, PFOA, PFHxS, PFOSA) worldwide in a wide array of mammal, bird, and fish species, including grey seals, polar bears, brown pelicans, black footed albatross, bald eagles, and yellow-fin tuna. PFAS are easily accumulated throughout all trophic levels, including at the lowest levels of grazing, filtering, and shredding invertebrates (Ahrens & Bundschuh, 2014).

Available evidence shows the likely potential for bioaccumulation or biomagnification in marine or terrestrial species (EPA, 2009). Biomagnification results in greater levels of PFAS in animals higher on the food chain (e.g., seals, polar bears), relative to animals at lower trophic levels (Ahrens & Bundschuh, 2014; Houde et al., 2011). PFOS and longer chain PFCAs (> C₈) bioaccumulate and persist in protein-rich compartments of fish and birds, and in marine mammal tissues, such as carcass, blood, and liver (Conder et al., 2008). Carcass typically consists of bones, but sometimes includes head, fins, skin, or feathers, as well. PFOS has been the most frequently detected PFAA in zooplankton and other invertebrate studies (Reiner & Place, 2015). However, these authors note that while some studies have shown concentrations of PFOS and other PFAAs very close to detection limits, more recent studies have improved analytical techniques (e.g., lower detection limits, better recovery, and use of reference materials), which produce less uncertainty in analytical results.

Levels of PFSAs and PFCAs in organisms are consistently measured at higher levels compared to other PFAS, and chain lengths of eight and above predominate (Martin et al., 2013; Dassuncao et al., 2017). In fact, many studies have suggested that PFCAs and PFSAs with shorter than seven and six fluorinated carbons, respectively, would not be considered bioaccumulative according to common regulatory criteria (Conder et al., 2008; Martin et al., 2013). However, there may be exceptions to this.

As described in <u>Appendix 5: Environmental Occurrence</u>, such chemicals have also been identified in wildlife present in Washington state. For example, PFOS and other long-chain PFAAs have been detected in freshwater fish fillet and liver samples, as well as in osprey eggs (Ecology, 2017, in prep.). However, detection of chemicals in wildlife does not necessarily imply

high bioaccumulation potential for any specific chemical, but does comprise a standard element of many environmental monitoring programs.

The large number of biota samples collected that contain quantifiable amounts of PFCAs, the ongoing scientific discourse regarding the high persistence and long-range fate and transport of PFCAs, and perceived similarities with perfluorinated sulfonates (including PFOS) have prompted concerns regarding the bioaccumulation potential of PFCAs (Conder et al., 2008). For example, due to their persistence and ability to transport long distances, animals do not need to be near sources of PFAS to show bioaccumulation. In particular, one study has shown elevated levels of PFAS in Scandinavian marine animals, although there is no production of PFAS in Scandinavia (Roos et al., 2013). In addition to long-range transport, PFOS is reported to have a very low Henry's law constant (i.e., ratio of a chemical concentration in the air to its concentration in water), indicating aquatic environments may be a significant sink for PFOS with a potential for bioaccumulation in fish (Boudreau et al., 2003).

Plants have been shown to take up some types of PFAS from the soil (Ahrens et al., 2016; Blaine et al., 2014), an issue of concern, since, for example, agricultural fields have the potential to be treated with PFAS-contaminated biosolids from WWTPs. Certain types of PFAS (e.g., shorter chain PFAS) can accumulate in leaves, fruits, and roots, with levels correlating with water content of the plant (Blaine et al., 2013; Scher et al., 2018). Concentrations of PFOA and PFOS in plants vary greatly, depending on the concentrations applied to the soil and soil-to-plant uptake factors. The uptake and transfer of these substances to vegetative parts of the plants appear to be greater than the transfer to storage organs within the plants (Stahl et al., 2009).

PFAA studies of birds have benefited from having species derived from many regions of the planet, including both aquatic and terrestrial ecosystems, representing a broad range of PFAA sources. The majority of these studies focus on birds from the Arctic, North America, and Europe. However, there does appear to be limited studies from the Southern Hemisphere (Antarctica and the Southern Ocean). Although initial wildlife studies focused on PFOS and PFOA, subsequent studies have expanded to include PFSAs, PFCAs, and PFAA precursors (e.g., FT alcohols, sulfonamide ethanols, perfluorosulfonamides, FT saturated and unsaturated carboxylic acids) (Reiner & Place, 2015).

In an earthworm study focused on bioaccumulation of PFAAs, the highest BAF (139 g soil dry wt/g worm dry wt) was observed for PFHxS in a soil contaminated with firefighting foam (Rich et al., 2015). BAFs increased with chain length for PFCAs but decreased with chain length for PFSAs (Rich et al., 2015). The unexpected finding for PFSAs may relate to decreased bioavailability. Overall, results from this study indicated that PFAA bioaccumulation into earthworms depends on soil concentrations, soil characteristics, analyte, and duration of exposure, and that accumulation into earthworms may be a potential route of entry of PFAAs into terrestrial foodwebs (Rich et al., 2015).

Other studies have assessed PFAS uptake in terrestrial biota. For example, Das et al. (2015) reported BAFs, ranging from 1.2 – 13.9 in earthworms exposed to soils contaminated with aqueous film forming foams (AFFF) containing PFOS. In their study, BAFs were higher from soils with lower PFOS concentrations. In another study, biota-soil accumulation factors (BSAFs) in earthworms increased as PFAS chain length increased (Mohammadi, 2015). However, chain length had an inverse effect in zucchini plants, showing a decreased BCF with longer chain length (Mohammadi, 2015), similar to results of Rich et al. (2015) noted above for PFSAs. In addition, Mohammadi (2015) reported no significant differences in BSAF or BCF among PFAS compounds with the same chain length but different functional groups. However, Shi et al. (2018) found that functional group was a relatively more important predictor of internal distribution than chain length for PFAS in an aquatic study.

In summary, bioaccumulation is generally apparent for a variety of long-chain PFAS compounds in both terrestrial and aquatic wildlife. However, as shown in Table 62, BAF values in aquatic biota vary by specific compound, species, and tissue.

Table 62. BAF values for aquatic biota.

PFAS	Species	Tissue	BAF (L/Kg)	Reference
PFOS	Bluegill	Fillet	2,700	MPCA, 2013
PFOS	Carp	Fillet	1,237	MPCA, 2013
PFOS	Freshwater Drum	Fillet	3,077	MPCA, 2013
PFOS	Smallmouth Bass	Fillet	2,845	MPCA, 2013
PFOS	White Bass	Fillet	4,618	MPCA, 2013
PFOS	Common Shiner	Liver	6,300 – 125,000	Moody et al., 2002
PFOS	Rainbow Trout	Carcass	690	ECCC, 2017
PFOS	Rainbow Trout	Blood	3,100	ECCC, 2017
PFOS	Rainbow Trout	Liver	2,900	ECCC, 2017
PFOS	Phytoplankton	Whole body	169	Loi et al., 2011
PFOS	Lake Trout	Whole body	31,623	De Silva et al., 2011
PFOA	Phytoplankton	Whole body	292	Loi et al., 2011
PFOA	Lake Trout	Whole body	126	De Silva et al., 2011
PFOA	Rainbow Trout	Blood	27	OECD, 2008
PFOA	Rainbow Trout	Liver	8	OECD, 2008
PFOA	Rainbow Trout	Whole body	4	OECD, 2008
PFHxS	Phytoplankton	Whole body	58	Loi et al., 2011
PFNA	Phytoplankton	Whole body	1,650	Loi et al., 2011
PFDA	Phytoplankton	Whole body	765	Loi et al., 2011
PFECHS	Lake Trout	Whole body	631	De Silva et al., 2011
PFUnDA	Phytoplankton	Whole body	4,510	Loi et al., 2011

PFOS is typically a dominant PFAS compound in fish (Taniyasu et al., 2003; Yoo et al., 2009), although internal distribution and relative PFAS compound concentrations in fish depend on tissue (e.g., blood vs. liver) and their proteinophilic properties (Jeon et al., 2010). Shi et al.

(2018) observed that BAF patterns in carp are most consistent with protein-binding mechanisms, although partitioning to phospholipids may contribute to the accumulation of long-chain PFAS in specific tissues. Among several PFOS isomers, linear PFOS represented a much higher proportion of total PFOS (sum of linear and branched isomers) in zooplankton and fish (Houde et al., 2008). BAFs for linear PFOS were also greater than for branched PFOS isomers in this food web (Houde et al., 2008). Longer chain PFCAs (e.g., C_{12} , C_{14}) were associated with higher BCFs in carp (10,000 – 17,000), relative to shorter chain PFCAs (e.g., C_{8} , C_{11}) (Inoue et al., 2012). In this study, viscera contained higher PFAS concentrations than head and integument in carp. With most PFAS compounds (except PFOA), a positive correlation has been observed between BCF in blackrock fish and salinity (Jeon et al., 2010).

Due to the persistence of all PFAAs, including short-chain PFAAs, exposure will continue regardless of accumulation because bioaccumulation is not required for sustained internal exposure (Cousins et al., 2016). For this reason, the Norwegian and German environmental agencies have proposed adding a new designation to the REACH Substances of High Concern list—mobile (M) and very mobile (vM). This would allow short-chain compounds, which do not categorize as persistent, bioaccumulative, and toxic (PBT) since they are not bioaccumulative, to still be added to the list as persistent, mobile, and toxic (PMT) (Kotthoff & Bucking, 2018; Turley, 2018). The theory is that PMT compounds would be an equivalent concern to PBT compounds, since they would also have sustained exposure.

In summary, bioaccumulation within the organism appears to be dependent on chain length. PFAS that contain six or more perfluorinated carbons have the potential to bioaccumulate within ecological receptors. It is apparent that wildlife from around the world are exposed to PFAS compounds. The main compound found in most wildlife species is PFOS. However, especially in more recent studies, long-chain PFCAs are frequently detected and measured (Reiner & Place, 2015).

6.3 Toxicokinetics

The toxicokinetic properties and toxicological responses of PFOS and PFOA have been studied in some detail. These chemicals may act as endocrine disruptors, although their molecular mechanisms are still debated (Kang et al., 2019). Immunotoxicity seems to be another effect. For example, the literature demonstrates a high confidence in the association between PFOA exposure and suppressed antibody response in animals (NTP, 2016). While there appears to be more literature available for aquatic than upland biota, enough information is available for both to summarize the fate, as well as possible adverse effects, of these contaminants. In particular, animal studies with both PFOS and PFOA have shown that they are well-absorbed orally, but poorly eliminated, not metabolized, and undergo extensive re-uptake from enterohepatic circulation (Lau, et al. 2007). Enterohepatic circulation refers to the circulation of the chemical from the liver to the bile, followed by entry into the small intestine, absorption by the intestinal absorptive cells, and then transport back to the liver.

Unlike most other bioaccumulating compounds, PFAAs do not bind to lipids (fats), but instead bind to proteins (Jones et al., 2003). Because of this, they are found mostly in the liver and blood (Norden et al., 2013), which lessens the utility of using measures such as K_{ow} (octanolwater partition coefficient) to predict bioaccumulation. K_{ow} is not particularly relevant for assessing PFAS distribution in tissues. Thus, it is difficult to predict bioaccumulation of PFAS compounds, such that evaluating bioaccumulation may require experimental testing.

Conder et al (2008) also noted that the principal repository of bioaccumulated PFCA and PFSA in organisms is not lipid but protein. Although a portion of these chemicals is hydrophobic and may interact with lipids, the presence of the carboxylate or sulfonate functional group imparts high hydrophilicity, thereby making the molecule partly lipophilic and partly hydrophilic (Houde et al., 2008; Shi et al., 2018).

Several studies have suggested that PFAAs are proteinophilic. For example, PFO in both rats and humans was strongly associated with serum albumin and other cytosolic proteins, and the proteinophilic nature of this class of chemicals has been hypothesized for the longer-chain PFAS (seven to eight fluorinated carbons). In support of this hypothesis, PFD (nine fluorinated carbons) has been shown to be more potent than PFO (seven fluorinated carbons) in binding to avian and carp serum proteins (Conder et al., 2008).

Therefore, in general, studies indicate that PFAAs are proteinophilic. For example, the tissue distribution of PFOA is dictated, to some extent, by its ability to bind plasma and other proteins (Kennedy et al., 2004). In contrast to the protein-binding ability of those chemicals with longer fluorinated carbon chains, the shorter perfluorinated compounds (PFSA and PFCA with four and three fluorinated carbons, respectively) were found to be one to two orders of magnitude less proteinophilic (Conder et al., 2008).

6.4 Toxicological Effects

Comparing adverse effects among studies can be confounded by differences in species and gender, as well as experimental differences in dose regimen (e.g., spacing, magnitude, duration, and route of administration). However, if the toxic mechanism is conserved, and some measure of the tissue concentration (i.e., dosimetry) at the biological target can be determined, then it is expected that this dosimetric anchor would be conserved across studies. Careful consideration of toxicokinetics is therefore required in order to link chemical exposure to toxicity (Wambaugh, 2015). Tables 63, 64, and 65 presented later in this section illustrate effects associated with chemical concentrations in water (aquatic species) or chemical dose (upland species).

6.4.1 Aquatic biota

Several toxicological effects have been tabulated for PFOA and PFOS in aquatic biota (Tables 63 and 64). A variety of endpoints and effect concentrations are listed. As expected, gene

expression effects occur at low concentrations, relative to concentrations linked with deficits in apical endpoints (e.g., growth, survival, reproduction) (Table 63). This observation is consistent with an adverse outcome pathway (AOP) framework, ranging from a molecular initiating event to lethality (Ankley et al., 2010; Kramer et al., 2011).

Table 63. Toxicological effects of PFOA and PFOS in freshwater species.

Chemical	Species	Concentration (ųg/L)	Effect	Reference
PFOA	Blue-green algae	5,000 (LOEC)	Physiology/Membrane potential	Rodea-Palomares et al., 2015
PFOA	Atlantic salmon	100 (LOEC)	Genetics/Bone development	Spachmo & Arukwe, 2012
PFOS	African clawed frog	0.1 (LOEC)	Genetics/Up-regulation of thyroid hormone regulated genes	Cheng et al., 2011
PFOS	Water flea	312.5 (LOEC)	Reproduction	Ji et al., 2008
PFOS	Fathead minnow	3,300 (NOEC)	Survival	Drottar & Krueger, 2000

Notes:

- LOEC is lowest observed effect concentration.
- NOEC is no observed effect concentration.

Freshwater biota

It has been reported that PFAS inhibited growth and had detrimental effects on photosynthesis on green algae (P. subcapitata, S. capricornutum, and C. vulgaris) (Boudreau et al., 2003; Ding et al., 2012), as well as the floating macrophyte, L. gibba (Boudreau et al., 2003). PFOS has been shown to be moderately toxic to aquatic invertebrates with acute toxicity values (48 and 96 hr LC50) in the range of 10 - 300 mg/L, while PFOA has been shown to be only slightly toxic to aquatic invertebrates, with toxicity values in the range of 100 - 1,000 mg/L (Li, 2009).

OECD (2002) lists a 42 d NOEC (survival) of 0.3 mg/l in an early life stage test with fathead minnows, using the potassium salt of PFOS. Although no significant effects were observed in another study (Ankley et al., 2005) on survival and growth for developing fathead minnows over 24 days at this same concentration (0.3 mg/L PFOS), these authors reported a 21-day EC50 (fecundity) of 0.23 mg/L PFOS in adult fish. This highlights the influence of life stage, test duration, and endpoint on effect concentration. PFOA concentrations were observed in the following order in the tissues of rainbow trout (Oncorhynchus mykiss): blood > kidney > liver > gall bladder > gonads > adipose > muscle tissue, at average water exposure concentrations between 0.014 and 1.7 μ g/L (Martin et al., 2003). PFAAs also were detectable in the gills, suggesting that this was the site of uptake, depuration, or both, as has been determined for other xenobiotics (Martin et al., 2003).

Table 64. Toxicological effects of PFOA and PFOS in saltwater species.

Chemical	Species	Concentration (ug/L)	Effect	Reference
PFOA	Sea urchin	20,000 (LOEC)	Growth/length	Mhadhbi et al., 2012
PFOA	Mysid	7,800 (EC10)	Mortality	Mhadhbi et al., 2012
PFOS	Mysid	530 (LOEC)	Survival	Drottar & Krueger, 2000
PFOS	Algae	12,200 (EC10)	Population/growth rate	Mhadhbi et al., 2012
PFOS	Mysid	3,200 (EC10)	Mortality	Mhadhbi et al., 2012
PFOS	Sea urchin	2,000 (EC10)	Growth/length	Mhadhbi et al., 2012

Notes:

- LOEC is lowest observed effect concentration.
- EC10 is 10% effect concentration.

Marine environment

Marine diatoms are far more sensitive to PFCAs than green algae, probably because of differences in cell wall structure (Latala et al., 2009). These authors recommended that further research focus on effects of PFAS mixtures and their derivatives in aquatic systems.

Biomagnification of PFOS, along a marine food chain (Greenland and Faroe Islands), was in the order shorthorn sculpin > ringed seal > polar bear (Bossi et al., 2005). The greatest concentration of PFOS was found in the liver of polar bears (mean = 1,285 ng/g wet wt, n = 2). In a lower latitude region (southeastern U.S.), PFOS and C_8 and C_{10} PFCAs concentrations have been detected in dolphin plasma and tissue samples, and long-chain PFCAs were found to biomagnify in this dolphin food web (Houde et al., 2006a). Although dolphins in southern Australia contained relatively high PFOS concentrations in liver, adverse effects were not apparent (Gaylard, 2017).

In contrast, data presented on PFOS exposure to marine wildlife (e.g., nursing beluga whale calves) suggests risk for developmental impacts in a Canadian Arctic marine food web (Kelly et al., 2009). PFOS and $C_8 - C_{14}$ PFCAs were highly bioaccumulative in this Arctic marine food web. However, biomagnification of perfluorinated acids (e.g., PFCAs, PFSAs) was seen in airbreathers (e.g., seals, whales, polar bears) but not in water-breathers (e.g., fish). The lack of PFAS biomagnification observed in water-respiring biota may be due to high aqueous solubility of these PFAS compounds, along with their efficient respiratory elimination via gills. In contrast, resistance to metabolism and low volatility (such as slow protein or air elimination) of PFAS results in biomagnification in air-breathing wildlife (Kelly et al., 2009).

6.4.2 Terrestrial biota

Upland plants (and surrogates)

Surrogate plant species are often used to evaluate the effect of contaminants on native wild plant species (EPA, 2012). For example, effects of PFAS on growth and reproduction have been studied in lettuce, pak choi, and cucumber (Ding et al., 2012; Li et al., 2009). In these studies, there were no obvious effects on seed germination for these species. However, based on EC₁₀, EC₅₀, and NOECs, the five-day root elongation sensitivity of test plants to both PFOS and PFOA were in the order lettuce > pak choi > cucumber (Li et al., 2009). In addition, another study evaluated effects of seven PFCs (PFBA; 2,2,3,3,4,4,5,5 Octafluoro-1-pentanol; PFOA; PFNA; PFDA; PFUnA; PFDoA) in a five-day test on root elongation of lettuce (*L. sativa*) (Ding et al., 2012). This study indicated that toxic effects of the seven PFCs increased with increasing fluorinated carbon chain length. It should be noted that extrapolating effects of PFAS on these surrogate test species to upland plants introduces additional uncertainty into an assessment of wild native plants.

Upland animal wildlife (and surrogates)

Limited information is available on the toxicokinetics and toxicological properties of PFOS and PFOA on upland wildlife receptors. Because few studies have determined safe exposure levels (NOAELs) for situations in which wildlife have been exposed over an entire lifespan or several generations, chronic exposures to a particular chemical are often estimated from toxicity studies conducted on a surrogate species with standard protocols. In many cases, the only available information is from studies on a laboratory species (primarily rats and mice) (Sample et al., 1996). While not ideal, these surrogate species do provide valuable information.

For example, a study was performed exploring the induction of liver tumors in Wistar rats for several chemicals, including PFOA (Abdellatif et al., 1990). In comparison to controls, this study indicated that PFOA caused a 24-fold increase in the peroxisomal β -oxidation of fatty acids, but only about a 2-fold increase in catalase activity. These results suggest that PFOA has a promoting action on liver carcinogenesis.

In other laboratory studies, exposure to PFOA significantly increased relative liver weights in offspring in all treatment groups in a full gestation study. Offspring of PFOA-treated dams exhibited significantly stunted mammary epithelial growth, as assessed by developmental scoring (Macon et al., 2011). Evaluation of internal dosimetry in offspring revealed that PFOA concentrations remained elevated in liver and serum for up to six weeks and that brain concentrations were low and undetectable after four weeks. Additionally, in wild-type mice, concentrations of PFOA measured in the serum and liver were directly correlated with increasing dose to the animal, while the livers had ultrastructural changes induced by PFOA (Wolf et al., 2008).

In an air cell injection study of PFOS with white leghorn chicken embryos, Molina et al. (2006) report an LD50 of 4.9 μ g/g egg (embryos) and a LOAEL of 0.1 μ g/g egg (reduced hatchability). Pathological changes in the liver were observed at doses as low as 1.0 μ g/g egg, including bile duct hyperplasia, periportal inflammation, and hepatic cell necrosis. Based on reproductive studies with mallard and northern bobwhite, Molina et al. (2006) suggest that the chicken embryo is considerably more sensitive to PFOS, as compared with wild avian species. NOAELs, LOAELs, toxicity reference values (TRVs), and predicted no effects concentrations (PNECs) have been derived for dietary PFOS exposure for the mallard and northern bobwhite quail (Newsted et al., 2005). Toxicological endpoints included mortality, growth, feed consumption, and histopathology. Reproductive endpoints included egg production, fertility, hatchability and survival, and growth of offspring. Newsted et al. (2005) report a TRV of 0.021 mg/kg bw/d and a PNEC of 0.013 mg/kg bw/d for dietary PFOS intake, proposing that these benchmarks are protective of avian populations.

Reproductive and developmental effects are presented for several PFAS in terrestrial animal species in Table 65. Again, a variety of endpoints and dose levels are listed. In some cases, NOAELs and LOAELs were identified for selected endpoints from the study. Most of these data are for surrogate test animals, which imperfectly represent wildlife species.

Table 65. Reproductive and developmental effects of selected PFAS compounds in terrestrial upland and surrogate animal species (Stahl et al., 2011).

Chemical and dose	Species	Exposure period (gestation days)	Effect	NOAEL	LOAEL	Reference
PFOS 1 – 10	Rats	6 – 15	Decreased body mass and lens	5 (fetal, maternal)	10 (fetal, maternal)	Gortner, 1980
mg/kg BW/d			abnormalities			
PFOS 10 – 150 mg/kg feed	Quail	NR	Decreased viability of the 14 day old progeny; slight increase in incidences of small testes, however spermatogenesis and fertility were not affected	<10 mg/kg feed (progeny)	10 mg/kg feed (progeny)	Newsted et al., 2007

Chemical and dose	Species	Exposure period (gestation days)	Effect	NOAEL	LOAEL	Reference
PFOS 0.1 – 3.75 mg/kg BW/d	Rabbits	6 – 20	Decrease in weight gain of the maternal animal; decreased birth weight and delayed ossification	0.1 (maternal), 1 (fetal)	1 (maternal), 2.5 (fetal)	Case et al., 2001
PFOS 1 – 5 mg/kg egg	Leghorn	Before incubation	No effect on hatching rate; increased spleen mass; right wings shorter; frequent occurrence of brain asymmetry; decreased immunoglobulin; increased plasma lysozyme activity; increased liver mass; increased body length	<1 mg/kg egg	1 mg/kg egg	Peden- Adams et al., 2009
APFO 1 – 30 mg/kg	Rats	NR	Decreased body weight; increased liver and kidney mass; decreased birth weight; delayed puberty; increased mortality rate after weaning	>30 mg/kg (parent and F1 reproduction), 10 mg/kg (F1 mortality and birth weight)	1 mg/kg (parent and F1 male decreased BW and organ weight increases), 30 mg/kg (F1 increased mortality, decreased birth weight)	Butenhoff et al., 2004

Chemical and dose	Species	Exposure period (gestation days)	Effect	NOAEL	LOAEL	Reference
PFOA 1 – 40 mg/kg BW/d	Mice	During gestation	Liver enlargement; decrease in full term gestation, viable fetuses, fetus weight, and postnatal viability; growth deficit; delayed opening of eyes; accelerated sexual maturity of male progeny	10 (dam weight gain), <1 (dam liver mass), 3 (offspring survival), 1 (offspring growth)	20 (dam decreased weight gain), 1 (dam liver enlargement), 5 (lower offspring survival), 3 (offspring growth deficit)	Lau et al., 2006
PFOA 5 – 40 mg/kg egg	Chickens	Before incubation	Impaired hatching rate; high prevalence of splayed legs; chicks with partial or complete loss of yellow pigment in the down	<5 mg/kg egg (hatching success), 20 mg/kg egg (splayed legs), 5 mg/kg egg (abnormal pigmentation)	5 mg/kg egg (decreased hatching), 40 mg/kg egg (splayed legs), 20 mg/kg egg (abnormal pigmentation)	Yanai et al., 2008
PFBA 35 – 350 mg/kg BW/d	Mice	1-17	No adverse effects on survival rate of progeny or their postnatal growth; delayed opening of eyes; delayed onset of puberty; at the highest dosage, loss of complete litter	<35 (maternal weight gain, fetus weight, neonatal survival, postnatal growth)	35 (offspring delayed eye opening), 175 (offspring delayed onset of puberty), 350 (full litter loss)	Das et al., 2008
PFDA 0.25 – 32 mg/kg BW/d	Mice	10 – 13	Decrease in weight gain of maternal animal at high doses, fetal body weight reduced a low doses, no malformations observed	>32 no malformations	16 (maternal decreased wt gain), 0.5 (fetal wt reduced)	Harris & Birnbaum, 1989

Chemical and dose	Species	Exposure period (gestation days)	Effect	NOAEL	LOAEL	Reference
PFDA 0.03 – 12.8 mg/kg BW/d	Mice	6 – 15	Decrease in weight gain of maternal animal at high doses, fetal body weight reduced at low doses, no malformations observed	>12.8 no malformations	6.4 (maternal decreased weight gain), 0.1 (fetal weight reduced)	Harris & Birnbaum, 1989

Notes:

- NR is not reported.
- NOAEL is no observed adverse effect level.
- LOAEL is lowest observed adverse effect level.
- Dose, NOAEL, and LOAEL are presented in units of [mg/kg BW/d] or in units indicated.
- Exposure period presented as gestation days or as indicated.

6.5 Data gaps and recommendations

6.5.1 Data gaps

Broad data gaps exist in exposure and effects assessment for wildlife species. For example, surrogate species are often used to represent wildlife species, exposure pathways are incompletely elucidated (such as food chain exposures), and cleanup levels for ecological receptors are generally lacking. Because the literature is rapidly expanding in the areas of PFAS ecotoxicity and ecorisk, this review is not comprehensive but instead serves as a snapshot of the subject.

Recent efforts to investigate PFAS ecotoxicity and ecorisk in the U.S. include:

- EPA's PFAS Action Plan (EPA, 2019).
- A focused topic meeting organized by the Society of Environmental Toxicology and Chemistry (SETAC), held in August 2019 (SETAC, 2019).
- A document produced by the Interstate Technology and Regulatory Council (ITRC) (ITRC, 2020b).
- Research conducted by the Strategic Environmental Research and Development Program (SERDP) and the Environmental Security Technology Certification Program (ESTCP), which are DOD programs (DOD, 2019; Conder, 2020).

6.5.2 Recommendations

Organizations in the U.S. (e.g., EPA) and internationally (e.g., European Union) are pursuing research and developing guidance that will fill regulatory gaps related to PFAS and ecotoxicology. As such, Ecology could leverage some of this work to implement many of the broader, more generic recommendations listed below. Perhaps in some cases, the needed information could be generated or adapted by Ecology to unique features in Washington state (such as PFAS environmental monitoring or protection of orcas in Puget Sound), depending on agency resources like funding and staff time.

Based on the analysis in this appendix, we make the following sub-recommendations as part of Recommendation 2.1:

Recommendation 2.1: Establish PFAS cleanup levels for soil and groundwater:

- Ecology will conduct monitoring for PFAS compounds in environmental media (e.g., soils, surface waters, sediment) and in tissues of aquatic and terrestrial wildlife to identify sources and to assess exposure.
- Based on data collected, Ecology will consider development of cleanup levels for individual or mixtures of PFAS for soil, sediment, fresh water, and salt water to protect ecological receptors.
- In this context, the following activities will be implemented to support activity under the recommendations above:
 - Trophic transfer and bioaccumulation of PFAS compounds should be further evaluated in aquatic and terrestrial food webs to further understand exposure.
 - Selected individual PFAS compounds, as well as common PFAS mixtures, should be evaluated for ecotoxicity in aquatic and terrestrial biota, using both laboratory and field methods.
 - Ecological risk assessment should be performed for PFAS compounds by detailing exposure and effects in order to estimate risks to nonhuman biota.
 - An uncertainty analysis should accompany PFAS ecorisk assessment to promote transparency in the risk assessment and communication processes and to more clearly identify data gaps.
 - Results of these risk assessments should support potential interventions (for example, species protections) and characterization of potential impacts on ecological services.

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List of acronyms

General acronyms

Table 66. Acronyms found in the ecological toxicology appendix.

Acronym	Definition
BAF	Bioaccumulation factor
BCF	Bioconcentration factor
CAP	Chemical Action Plan
EC()	Percent effect concentration
Kg	Kilogram
L	Liter
LOAEL	Lowest observed adverse effects level
LOEC	Lowest observed effects concentration
NOAEL	No observed adverse effects level
NOEC	No observed effects concentration

Chemical names

Table 67. Chemical name acronyms found in the ecological toxicology appendix, excluding the general acronyms listed in the table above.

Acronym	Chemical name
6:2 FTAC	6:2 Fluorotelomer acrylate
6:2 FTMAC	6:2 Fluorotelomer methacrylate
6:2 FTOH	6:2 Fluorotelomer alcohol
APFO	Ammonium perfluorooctanoate
FT	Fluorotelomer
PASF	Perfluoroalkane sulfonyl fluoride
PFAA	Perfluoroalkyl acid
PFAS	Per- and Polyfluoroalkyl substances
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonate
PFC	Perfluoroalkyl contaminants

Acronym	Chemical name
PFCA	Perfluoroalkyl carboxylic acid
PFD	Perfluorodecanoate
PFDA	Perfluorodecanoic acid
PFDo	Perfluorododecanoate
PFDoA	Perfluorododecanoic acid
PFECHS	Perfluoroethylcyclohexanesulfonate
PFHp	Perfluoroheptanoate
PFHpS	Perfluoroheptanesulfonate
PFHx	Perfluorohexanoate
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexane sulfonate
PFN	Perfluorononanoate
PFNA	Perfluorononanoic acid
PFO	Perfluorooctanoate
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate
PFOSA	Perfluorooctane sulfonamide
PFPeS	Perfluoropetanesulfonate
PFPn	Perfluoropentanoate
PFS	Perfluoroalkyl substance
PFSA	Perfluoroalkyl sulfonic acid
PFT	Perfluorotetradecanoate
PFTri	Perfluorotridecanoate
PFU	Perfluoroundecanoate
PFUnA/PFUnDA	Perfluoroundecanoic acid

Appendix 7: Health

7.0 Overview

7.0.1 Findings

- We are still learning about potential health risks of per- and polyfluoroalkyl substances (PFAS) in humans. Much of what we know comes from toxicity testing in laboratory animals on several perfluoroalkyl acids (PFAAs). The evidence base is strongest for perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS), and is rapidly expanding for other PFAAs.
- Animal studies provide strong evidence that some PFAAs produce liver and kidney toxicity, immune toxicity, reproductive and developmental toxicity, endocrine disruption (altered thyroid and testosterone hormones), and certain tumors. The strongest evidence from epidemiological studies is for links between higher exposures to PFAAs and reduced antibody response to childhood vaccines, increased serum cholesterol and liver enzymes, and slightly reduced birth weights. More limited evidence exists for links to thyroid disease, hormone disruption, and reduced resistance to infections, cardiovascular disease, and cancer.
- Some long-chain PFAAs are strongly bioaccumulative in people. It takes years for human bodies to excrete PFOS, PFOA, perfluorononanoic acid (PFNA), perfluorohexane sulfonate (PFHxS), and other long-chain PFAS. Other PFAAs, such as perfluorobutanoic acid (PFBA), perfluorobutane sulfonic acid (PFBS), and perfluorohexanoic acid (PFHxA), are more rapidly cleared. The absorption, distribution, and clearance in humans for most PFAS have not been studied.
- PFOS, PFOA, PFHxS, and PFNA have been detected in the serum of nearly every person tested in Centers for Disease Control and Prevention (CDC) surveys of the U.S. general population since 1999. Levels have declined since phase-outs of these PFAS and their precursors from domestic production and use.
- People can be exposed to PFAS from contaminated drinking water, other dietary sources, indoor dust and air that contain PFAS from consumer products, and use of consumer products that contain PFAS. Although it has been difficult to assess which sources contribute the most to human exposure, studies identify food and drinking water as the likely main routes of non-occupational exposure.
- In Washington, PFAAs have been identified in drinking water in Issaquah and at or near four military bases: Naval Air Station (NAS) Whidbey Island, Fairchild Air Force Base, Joint Base Lewis-McChord, and Navy Base Kitsap-Bangor. In each area, the sum of PFOA and PFOS in at least one drinking water well exceeded the lifetime health advisory level (70 parts per trillion (ppt)) set by the U.S. Environmental Protection Agency (EPA). PFAS-based firefighting foam is the suspected source of contamination at all of these areas. Ongoing investigations may identify other contributing sources.
- Washington Department of Health (Health) is supporting the State Board of Health (SBOH) in developing state drinking water standards for PFAAs. In 2019, Health

published draft recommendations for state action levels for five PFAAs in drinking water: PFOS (15 ppt), PFOA (10 ppt), PFNA (14 ppt), PFHxS (70 ppt), and PFBS (1,300 ppt). The SBOH expects to complete the rulemaking process in 2021. Health is also developing fish consumption advisories for PFOS in freshwater fish based on Washington State Department of Ecology (Ecology) fish sampling data.

7.0.2 Introduction

The purpose of this appendix is to review potential human health impacts of the PFAS measured in food, water, the environment, and bodies. It describes how humans are exposed and what levels are commonly measured in the general population and in populations with higher exposure. It describes the known sites of drinking water contamination across the state and the actions taken to protect public health at each site. It reviews the public health advice being developed by Washington state for PFAS in drinking water and fish.

The appendix focuses on PFAAs as these are the best studied and are the ultimate environmental and metabolic breakdown products for a large number of PFAS (see Appendix 4: Fate and Transport).

7.1 Human Health Hazard Assessment

There is a growing base of toxicological and epidemiological evidence available to support health assessments for a small number of PFAS. Authoritative bodies that have reviewed available evidence include EPA (EPA, 2016b, 2016c, 2018), the Agency for Toxic Substances and Disease Registry (ATSDR) (ATSDR, 2015a, 2018b), the National Toxicology Program (NTP) (2016) (NTP, 2016), the C8 Science Panel (C8 Science Panel, 2012), the International Agency for Research on Cancer (IARC) (IARC, 2016), and the European Food Safety Authority (EFSA) (EFSA, 2018, 2020). Additional PFAS toxicity testing and health assessment is underway at EPA and NTP (Patlewicz et al., 2019). In 2019, Health scientists also evaluated the available science to guide action on PFAAs in public drinking water (Health, 2019).

In animal studies, some PFAAs produce reproductive and developmental toxicity, liver and kidney toxicity, altered levels of thyroid hormones, immune toxicity, endocrine disruption, and tumors (See <u>Supplement 1</u> for a summary of findings for eight PFAAs). Studies of humans with occupational, community, or background exposures to PFAAs have reported that higher exposures are associated with a number of adverse health outcomes such as reduced antibody response to vaccines, higher serum cholesterol and liver enzymes, reduced birth weights, and increased rates of thyroid disease and cancer. These are summarized briefly in <u>Supplement 1</u>.

7.1.1 Epidemiology

Epidemiological studies have been valuable in understanding the potential relevance of laboratory animal data to human populations. They have also been instrumental in understanding the much longer retention of some PFAAs in humans. A landmark

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epidemiological study in the mid-Ohio River Valley called the C8 Health Project is also described below.

Human observational studies are limited in their capacity to establish a causal relationship between PFAA exposure and human health effects. Variable findings on outcomes measured in different populations have been noted and make interpretation of the epidemiological evidence stream challenging.

Apparent inconsistencies may be due to differences in study populations (e.g., age, gender, occupational vs. community exposure), study design (e.g., cross-sectional, longitudinal), level of exposure, and variations in the mixture of PFAS present (i.e., measured and unmeasured). Other sources of variability may be unmeasured historical exposures influencing the rates of health effects observed and other risk factors for outcomes measured that may not be adequately controlled. Taken together with evidence streams from laboratory animal testing, mechanistic studies, and in vitro and gene expression assays, many scientific and government agencies have reasonably concluded that some PFAAs likely pose a health risk to humans and established exposure guidelines or limits to protect human health (Interstate Technology Regulatory Council (ITRC), 2020b). Supplement 1 shows some of their recommended limits for human exposure to protect health.

The C8 Health Project, Ohio River Valley, 2005 – 2012

The largest epidemiological study of health outcomes associated with mostly PFOA exposure in drinking water is the C8 Health Project. This study, funded by a lawsuit settlement, set out to identify which conditions or diseases were more probably than not related to area-wide contamination around Dupont's Washington Works plant in Parkersburg, West Virginia. This fluoropolymer production plant released primarily PFOA into the air, water (Ohio River), and soil around their manufacturing site between 1950 and 2004. PFOA was detected in six public water systems in the mid-Ohio Valley in 2002. PFOA exposures to the communities are believed to have peaked in the early 1990s. Investigators concluded that drinking water contaminated with PFOA was the principal route of exposure for this population (Emmett et al., 2006; Hoffman et al., 2011; Shin et al., 2011; Steenland et al., 2009; Winquist et al., 2013).

In all, the C8 Health Project recruited more than 69,000 residents who had consumed drinking water for at least one year from six water districts in West Virginia and Ohio (Frisbee et al., 2009). The very high participation rate (81%) strengthened the relevance of study results for this population. At the time of water sampling, the highest average PFOA concentrations in drinking water were found at Lubeck, West Virginia (520 μ g/L), and Tuppers Plain, Ohio (310 μ g/L) (Olsen, 2015b).

Blood samples were collected for the C8 Health Project in 2005 and 2006. Compared to the general U.S. population, average serum concentrations for the cohort were about eight times higher for PFOA, 1.4 times higher for PFNA, 1.7 times higher for PFHxS, and not elevated for PFOS (Frisbee et al., 2009). The mean serum PFOA level varied significantly across the six water districts and ranged from 16 micrograms per liter (μ g/L) for participants from the Village of Pomeroy to 228 μ g/L for the Little Hocking Water Association. Females had slightly lower average serum levels than males for all four PFAS (Frisbee et al., 2009). Women have additional pathways to clear PFAAs through their menstrual cycle, childbirth, and breastfeeding. PFOA levels in groups of workers at DuPont's facilities were much higher: mean serum ranged from 494 μ g/L to 3,210 μ g/L (ATSDR, 2015b).

Between 2005 and 2012, the C8 Health Project carried out exposure and health studies in the mid-Ohio Valley communities affected by drinking water contamination. The project science panel assessed the links between PFOA and a number of diseases, and concluded that a "probable link" existed between PFOA and high cholesterol, ulcerative colitis, thyroid disease, testicular cancer, kidney cancer, and pregnancy-induced hypertension among the population evaluated (C8 Science Panel, 2012). They also found no probable link to other conditions including heart disease, chronic liver or kidney disease, stroke, several autoimmune diseases, occurrence of common infectious diseases or respiratory disease, asthma, or birth defects.

Following the C8 Health Project, carbon filters were installed to remove PFOA from public drinking water systems. As a result, PFOA serum concentrations declined 26% between the end of 2007 and mid-2008 in the groups from Little Hocking and Lubeck water districts—indicating a serum elimination half-life of 2.3 years for highly exposed populations (Bartell et al., 2010).

7.1.2 Primary health endpoints of concern

Liver toxicity

In experimental animals, the liver is a sensitive biological target for most PFAAs tested (PFOA, PFNA, PFDA, PFHxA, PFOS, PFHxS, PFHxS, and PFBS). Specific effects observed include increased liver weight, lipid accumulation (steatosis), increased liver cell size (hypertrophy), cell proliferation (hyperplasia), and tumors. Degenerative changes in the rodent liver have been observed for PFOS, PFOA, PFNA, perfluorodecanoic acid (PFDA), and PFBS (EPA, 2016b, 2016c; NTP, 2019a, 2019b). Human liver is expected to be less sensitive than rodent liver to effects mediated by peroxisome proliferator-activated receptor alpha (PPARα), but effects like steatosis appear to occur by another mechanism (Das et al., 2017; Hall et al., 2012).

In human observational studies, modest increases in serum liver enzyme levels and other markers suggestive of liver damage have been associated with higher serum levels of PFOA, PFOS, PFNA, and PFHxS in adults in some studies (Bassler et al., 2019; Darrow et al., 2016; Gallo et al., 2012; Gleason et al., 2015; Salihovic et al., 2018). In Gallo et al. (2012), an increase in C8 study participants with high serum levels of the liver enzyme ALT was observed across low to modest exposures to PFOA and PFOS, but leveled off above about 30 ng/mL of PFOS and PFOA (Gallo et al., 2012). In children, serum levels of ALT were not positively associated with prenatal exposure (PFAS in maternal serum) or current exposure (PFAS in serum at age eight years old) in a Boston-area cohort recruited from the general population before the PFOS and PFOA phase-out (Mora et al., 2018).

Elevated PFOA exposure was not associated with an increase of clinically diagnosed liver disease in the large C8 study despite subclinical indications of liver damage in this and other studies (Bassler et al., 2019; Darrow et al., 2016). A recent study reported positive associations between serum levels of PFHxS and PFOS and severity of disease in adolescents with nonalcoholic fatty liver disease (Jin et al., 2020).

Serum cholesterol levels and cardiovascular effects

PFAAs appear to alter lipid metabolism in the rodent and monkey. In animal studies, PFAA exposure produces decreased serum cholesterol and triglyceride levels (ATSDR, 2018b).

In human populations with environmental exposures, serum PFOA, PFOS, PFNA, and PFHxS are often positively associated with increased serum levels of total cholesterol and low-density lipoprotein (LDL) cholesterol (Frisbee et al., 2010; Graber et al., 2019; Li et al., 2020; Nelson et al., 2010; Steenland et al., 2009), including in children (Mora et al., 2018; Rappazzo et al., 2017). The difference in effect direction may represent a different effect at low doses—in fact, the direction of effect was reversed at very high doses in one study in humans (Convertino et al., 2018). The difference may also be a function of diet (rodents fed a high fat western diet showed an increase in cholesterol) (Rebholz et al., 2016) or a species difference in liver response (Corton et al., 2018). There is also an emerging uncertainty about potential confounding by

enterohepatic circulation of lipids (EFSA, 2020), although this would not explain associations between higher cholesterol and PFAA exposure based on residence in a water district with contaminated drinking water as in Li et al. (2020). The C8 Science Panel concluded that there was a "probable link" between PFOA and clinically defined "high cholesterol" (C8 Science Panel, 2012). In studies of populations with lower exposure, PFHxS was inversely associated or not associated with serum cholesterol (Nelson et al., 2010; Seo et al., 2018; Starling, Engel, Whitworth, et al., 2014). There is limited data on this outcome for shorter chain PFAAs, but PFHxA, perfluoroheptanoic acid (PFHpA), and PFBA were not associated with serum cholesterol levels in a small Chinese study (Fu et al., 2014).

There is a well-recognized relationship between elevated cholesterol and cardiovascular disease. Two studies in the CDC National Health and Nutrition Examination Survey (NHANES), a representative survey of the U.S. population, reported positive associations between serum PFAAs and self-reported cardiovascular disease after controlling for other risk factors (Huang et al., 2018; Shankar et al., 2012). No association between relatively high PFOA exposure and coronary heart disease was reported in the C8 study. The C8 study confined analysis to self-report verified by medical records (C8 Science Panel, 2012).

The C8 Science Panel found a "probable link" between a hypertensive disorder of pregnancy (through self-report of pre-eclampsia) and maternal PFOA and PFOS serum level (C8 Science Panel, 2012; Savitz, Stein, Bartell, et al., 2012). No association with pre-eclampsia was found in two other studies of populations with lower PFOA and PFOS exposures, which validated cases with medical records (Huang et al., 2019; Starling, Engel, Richardson, et al., 2014).

Immune toxicity

PFOS and PFOA can suppress both adaptive and innate immunity in rodents and non-human primates (Corsini et al., 2014). Outcomes observed include lower spleen and thymus weights, reduced antibody response to antigen, reduced survival after influenza infection, and altered cytokine production. Toxicity testing data indicates that PFNA and PFDA can also adversely impact the immune system in rodents (Frawley et al., 2018; Fang et al., 2009; Fang et al., 2008).

In 2016, NTP concluded that PFOA and PFOS are "presumed immune hazards" to humans based on a systematic review of available toxicology and epidemiology. Lower antibody response to antigen observed in rodent studies and reduced antibody titers to childhood vaccines were key points of evidence (NTP, 2016). Associations between PFOA, PFOS, PFHxS, and PFDA levels in serum and reduced antibody response to childhood vaccines have been reported in different populations (Grandjean et al., 2012; Granum et al., 2013; Mogensen et al., 2015; Stein et al., 2016). Since the NTP review, additional studies have reported inverse associations between the sum of PFOS, PFOA, PFNA, and PFHxS in serum and reduced antibodies titers to childhood vaccines (Abraham et al., 2020; Grandjean, Heilmann, Weihe, Nielsen, Mogensen, & Budtz-Jorgensen, 2017; Grandjean, Heilmann, Weihe, Nielsen, Timmermann, et al., 2017).

The NTP review did not find consistent associations between PFOS or PFOA exposure and lowered resistance to infectious disease in people. Since this review, several studies (including one large prospective cohort study) reported positive associations between maternal concentrations of PFAS and measures of offspring infection (Dalsager et al., 2016; Goudarzi et al., 2017; Impinen et al., 2019; Impinen et al., 2018).

PFOA has been associated with a single autoimmune outcome (ulcerative colitis) in two highly exposed U.S. populations (C8 community cohort and occupational cohort) (Steenland et al., 2015; Steenland et al., 2013). A case-control study in the general U.S. population also reported that higher serum PFOA was associated with ulcerative colitis but not other inflammatory bowel diseases (Steenland, Kugathasan, & Barr, 2018). A recent study of a Swedish population with high levels of PFOS and PFHxS in drinking water contaminated by firefighting foam did not observe an association between exposure and ulcerative colitis, Crohn's Disease, or other inflammatory bowel disease (Xu, 2019).

NTP concluded it had a high level of confidence that PFOA increased hypersensitivity outcomes in animals but only low confidence in evidence from human studies (National Toxiciology Program, 2016). Hypersensitivity outcomes such as asthma and elevated Immunoglobulin E (IgE) have been reported to be associated with PFAS in several studies (Dong et al., 2013; Humblet et al., 2014; Kvalem et al., 2020; Zhu et al., 2016), but many other studies have looked for and not found associations. Overall the evidence for allergies and asthma is weak and unclear.

Developmental toxicity

There is evidence of profound developmental effects of PFOA and PFOS in experimental animals including fetal loss, altered fetal bone development, lower birth weight, reduced pup survival, altered behavior in offspring, and altered timing of sexual maturation in offspring at adolescence (Butenhoff et al., 2004; Lau et al., 2006; Luebker, Case, et al., 2005; Ngo et al., 2014; Thibodeaux et al., 2003). PFNA produced many similar effects in mouse studies (Das et al., 2015; Wolf et al., 2010). PFBA, PFBS and PFHxA, and PFHxS were either less potent or did not produce developmental effects (see <u>Supplement 1</u>).

The most consistent developmental effect observed in humans is slightly lower birth weights associated with higher maternal serum levels of PFOA and PFOS. Limited but similar associations are reported for PFNA, PFHxS, PFDA, and perfluoroundecanoic acid (PFUnDA). Preterm birth was associated with higher exposure to PFNA and PFDA (EPA, 2016b; Johnson et al., 2014; Maisonet et al., 2012; Marks et al., 2019; Meng et al., 2018; Wang et al., 2016; Wikstrom et al., 2019). Several meta-analyses have reported small but consistently inverse relationships between maternal PFOA level and birth weight. Confounding by maternal glomerular filtration rate appears to explain some of the association observed by studies that measured prenatal PFOA or PFOS exposure in maternal serum later in pregnancy or in cord blood (Johnson et al., 2014; Negri et al., 2017; Steenland, Barry, & Savitz, 2018; Verner et al.,

2015). However, two recent high quality studies measured maternal PFAA level early in pregnancy and reported significant inverse associations with birth weight (Meng, 2018; Wikstrom, 2019).

Slight delays in the age of puberty have been associated with serum PFOA (girls) and with serum PFOS (girls and boys) (Christensen et al., 2011; Kristensen et al., 2013; Lopez-Espinosa et al., 2011).

Reproductive toxicity and sex hormone disruption

Altered levels of reproductive hormones (serum testosterone, estradiol), altered sperm parameters, and changes in the testes have been observed in rodent studies following exposure to PFOS (Biegel et al., 2001; Lopez-Doval et al., 2014; Wan et al., 2011), PFOA (Biegel et al., 1995; Lau et al., 2007; NTP, 2019b), PFNA (Feng et al., 2009; NTP, 2019b), PFDA (NTP, 2019b), and PFDoDA (Shi et al., 2007). Inhibited development of mammary glands were observed in maternal mice exposed to PFOA during pregnancy and in their female offspring at puberty (White et al., 2009). The delays did not impair successful nursing of their young in either generation (White et al., 2011).

Eight PFAS were tested in vitro for binding affinity to the rat estrogen receptor alpha (ER α). PFOS and PFOA had the strongest affinity and showed higher affinity for human ER α than rat ER α receptor. Testing in rats confirmed that binding was functional (i.e., PFOS and PFOA increased expression of ER α in rat uterus and the levels of serum estradiol in rats) (Qiu et al., 2020).

Reproductive impairment such as reductions in rodent litter size were observed for PFOS, PFOA, and PFNA (Abbott et al., 2007; Lau et al., 2006; Singh & Singh, 2019a). No declines in fertility were evident in rodent testing for PFHxS, PFHxA, PFBA, or PFBS (Butenhoff et al., 2009; Das et al., 2008; Lieder et al., 2009; Loveless et al., 2009).

In a general population study, Vested et al. (2013) found that higher maternal serum PFOA was associated with lower sperm count in boys when they reached young adulthood (Vested et al., 2013). In a highly exposed population near a fluoropolymer plant in Italy, young adult men had higher serum PFAAs levels, reduced serum testosterone and semen quality, and shorter penis length and anogenital distance than a comparison population in an uncontaminated area (Di Nisio A, 2018). A number of other studies have not found associations between PFAS exposure, testosterone, or sperm parameters (Agency for Toxic Substances and Disease Registry (ATSDR), 2018b). Some epidemiological studies report reduced fertility associated with higher serum PFOA, PFOS, and PFHxS (Fei et al., 2009; Velez et al., 2015; Whitworth et al., 2012). Other studies have looked for and not found associations with fertility. Studies of communities with elevated exposure have looked for and generally not found associations between PFOA and birth defects or miscarriage. Other PFAAs are not as well studied.

Thyroid disease and thyroid hormone disruption

Alterations in thyroid hormone levels, particularly reductions in free thyroxine (T4) and free triidothyonine (T3) have been observed in laboratory animals exposed to long- and short-chain PFAAs (Butenhoff et al., 2002; National Toxicology Program (NTP), 2019a, 2019b; Seacat et al., 2002). Thyroid toxicity (i.e., increased organ weight, follicular cell hypertrophy, and hyperplasia) has been observed in some laboratory animal studies of PFOA, PFOS, PFHxS, and PFDA (Butenhoff, Chang, et al., 2009; J. L. Butenhoff, Ehresman, et al., 2009; NTP, 2019b, 2019c).

A systematic review of studies in pregnant women and their children (Ballesteros et al., 2017) and a meta-analysis of studies in adults (Kim et al., 2018) provide some suggestive evidence that PFOS, PFOA, and PFHxS are associated with altered hormones in people. Associations differed by gender and by PFAS, but no clear and consistent pattern of effect of PFAS on thyroid hormone levels has emerged. Sex-linked effects and non-linear dose-response curves are not uncommon in endocrine disrupting chemicals.

Two studies in the general U.S population reported that thyroid disease was associated with serum PFOA, PFOS, and PFHxS (Melzer et al., 2010; Wen et al., 2013). The C8 Science Panel concluded there was a probable link between PFOA exposure and increased risk for thyroid disease in the C8 Health Project (C8 Science Panel, 2012) based on increased hypo- and hyperthyroidism, especially in women (Winquist & Steenland, 2014), and an association between PFOA and hypothyroidism in children (Lopez-Espinosa et al., 2012). A Swedish study of a community with very high PFOS and PFHxS exposure through drinking water did not show higher risk for thyroid disease in men or women (Andersson et al., 2019).

Cancer

Chronic PFAA exposure studies in adult rats have shown increased tumors in liver (PFOA, PFOS); pancreas, testes, uterus (PFOA); and thyroid (PFOS) (Biegel et al., 2001; Butenhoff, Chang, Olsen, & Thomford, 2012; Butenhoff, Kennedy, Chang, & Olsen, 2012; Hardisty et al., 2010; NTP, 2019c; Thomford, 2002). Chronic oral exposure to hexafluoropropylene oxide dimer acid (HFPO-DA, also known as GenX) produced hepatic, Leydig cell, and pancreatic acinar cell tumors similar to PFOA (Caverly-Rae et al., 2015), whereas PFHxA did not produce evidence of tumors (Klaunig et al., 2015). The pattern of tumors observed for PFOA and GenX is consistent with other chemicals and drugs that activate PPARα in the rat, and is not considered relevant to human liver cancer risk (Klaunig et al., 2003).

No other PFAS have been studied for cancer in chronic animal assays, but some PFAS share attributes of chemical carcinogens (i.e., produce oxidative stress, are immunosuppressive, influence cell proliferation via nuclear receptors) (Temkin et al., 2020). For example, PFNA and PFDA were more potent than PFOA at promoting liver tumors in a trout model of PPARα-independent liver cancer. The gene-expression profile evident in liver of treated trout was similar to estrogen, which was used as a positive control in the study (Benninghoff et al., 2012).

Both occupational and non-occupational studies have investigated whether cancer is associated with PFOA and PFOS exposure in humans. IARC classified PFOA as possibly carcinogenic to humans (Group 2B) based on credible evidence of testicular and kidney cancer associated with PFOA exposure in animal testing and the C8 Health Project (Barry et al., 2013; IARC, 2016). The C8 Science Panel concluded that a "probable link" existed between PFOA exposure and testicular and kidney cancer, but no other types of cancer. Studies of the general population have looked for but not found associations between serum PFOA levels and a range of cancers (Bonefeld-Jorgensen et al., 2014; Eriksen et al., 2009; Hardell et al., 2014; Innes et al., 2014).

In occupationally exposed workers, associations between exposure to PFOS or PFOA and male reproductive, kidney, and bladder cancers have been reported. These associations were generally weak and not consistent across studies (ATSDR, 2015b; Kennedy, 2015; Raleigh et al., 2014; Steenland & Woskie, 2012). In addition, the sample sizes for many of these studies are small, and caution is needed in interpreting the results, as most studies did not control for other potential factors such as smoking (ATSDR, 2015a). A recent mortality study of Italian fluoropolymer workers highly exposed to PFOA reported higher mortality for liver cancer, liver cirrhosis, and diabetes than in a comparison group of workers at a nearby metalworking factory (Girardi & Merler, 2019).

PFOS serum levels were not associated with more breast, bladder, pancreatic, liver, or prostate cancers in the general Danish population (Bonefeld-Jorgensen et al., 2014; Eriksen et al., 2009; Roswall et al., 2018). A small case control study of Swedish men with prostate cancer found no overall association with PFOS except among a subgroup with a hereditary risk (i.e., first-degree relative with prostate cancer) (Hardell et al., 2014). Prostate specific antigen (PSA), a clinical marker used in screening for prostate cancer, was not associated with serum PFOS or other PFAA among greater than 25,000 men in the C8 cohort (Ducatman et al., 2015).

In 2016, EPA concluded that there was suggestive evidence of carcinogenic potential of both PFOA and PFOS in humans. For PFOA, EPA based its determination primarily on Leydig cell tumors observed in two rat studies and renal and testicular cancer in the C8 Health Project. For PFOS, EPA relied primarily on observations of liver and thyroid adenomas in chronic rat studies (EPA, 2016b, 2016c).

Sources of uncertainty in assessing hazard

We still know very little about the potential toxicity of thousands of individual PFAS. PFOA and PFOS have large evidence bases to support human health assessments. Another dozen or so PFAS have limited but growing data to support assessment. EPA and NTP are testing 150 PFAS using rapid high-throughput testing to more broadly inform hazard assessment of PFAS. This list includes PFAS from 75 different subclasses and may help regulators construct a grouped approach to managing PFAS (EPA, 2019; Patlewicz et al., 2019). Understanding how to assess PFAS mixtures is critical because most environmental samples and human biological samples

contain mixtures of PFAS, and yet we have very little data to understand how PFAAs may interact or the hazard of these cumulative exposure to PFAS mixtures.

Laboratory animals differ from humans in how rapidly they excrete a number of PFAS (serum half-lives in hours to days in rodents vs. years in humans), how their specific tissue responds (PPAR α activation in rodent vs. human liver tissue), and the adverse effects observed (serum cholesterol reductions in rodents vs. increased cholesterol in humans). Toxicokinetic models of internal dose help us extrapolate from animal results to humans, but some of the inputs—like human clearance rates for different lifestages, gender, and level of exposure—are uncertain. The mechanisms of action underlying adverse effects observed are only partly understood. This adds to uncertainty about which outcomes in test animals are most relevant to human risk assessment and which animal models are best suited to investigating outcomes observed in human studies.

The cross-sectional study design of most PFAS epidemiological studies limits their use in determining causality. In fact, researchers believe some health outcomes associated with serum levels of PFAS could be due to reverse causation. For example, earlier menopause and shorter breast-feeding duration may result in increased serum PFAS since menstruation and lactation are excretion pathways in women. Conditions like kidney disease that can reduce glomular filtration rate may lead to higher serum PFAS because the disease impairs a major excretion pathway.

Another concern is using a single serum sample to quantify PFAS exposure. Serum levels reflect exposure across recent months to years, but will not provide information on a historical peak exposure or fluctuations in serum over time. A single serum will not necessarily reflect the level in serum that preceded the onset of a disease or condition. Some studies, like the C8 Health Project, did exhaustive exposure reconstruction to overcome this limitation. A final concern is that a number of the outcomes with the most robust evidence in people—increased cholesterol, reduced birth weight, and immunosuppression—have many possible causes, which are difficult to control for in community-wide observational studies.

Epidemiological studies are limited in their ability to tease out associations between a specific PFAS and health outcomes in populations with simultaneous exposure to multiple PFAS. This is especially true when studying populations exposed through a community drinking water system, as individual PFAS will be highly correlated with each other in serum samples from that community. In addition, analytical methods have typically only measured about a dozen PFAS in drinking water and serum. Unidentified PFAS in drinking water may be contributing to community exposure and may confound associations between health outcomes and measured PFAS. For example, four new (previously unmeasured) PFAS were recently identified in the drinking water and human serum of residents in Wilmington, North Carolina (Center for Human Health and the Environment, 2018).

PFAS toxicology continues to be a very active area of research. NTP and EPA both have high priority research efforts underway to fill important data gaps. In addition, the National Institute of Environmental Health Sciences (NIEHS) is granting more than \$10 million dollars annually to universities to study PFAS. Hundreds of new peer-reviewed papers are published each year.

7.2 PFAS exposure in people

7.2.1 Trends and demographics of PFAA exposure

Widespread exposure to PFAAs in the U.S.

In national surveys, the CDC found that nearly all people tested had detectable levels of PFOS, PFOA, PFNA, and PFHxS in their blood serum. These four are the top four PFAAs measured in human serum in terms of frequency and concentration.

Figure 39 shows the average levels detected in U.S. serum over time. It's important to note that a voluntary phase-out of production of PFOS, PFOA, PFHxS, and related precursors by 3M occurred between 2000 and 2002, and a voluntary phase-out of PFOA and longer-chain perfluoro-carboxylic acids (PFCAs) occurred by eight major U.S. manufacturers between 2006 and 2015 under a stewardship agreement with EPA (ITRC, 2020a).

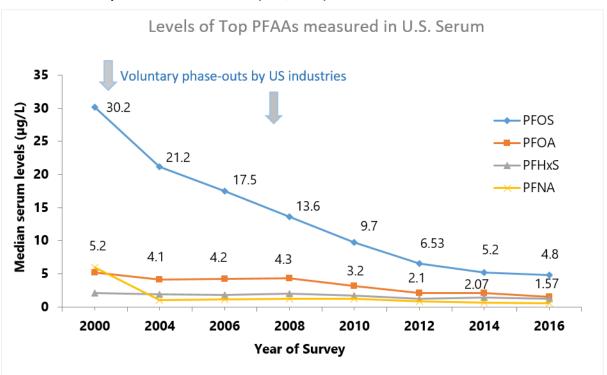


Figure 39. Time trend of median serum levels (μ g/L) of four PFAS in representative samples of U.S. residents over 12 years from the NHANES (CDC, 2019).

Biomonitoring data for the general population of Washington is limited to one study in 2004 by Olsen et al., in which seven PFAAs compounds were measured in stored blood serum of 238 elderly men and women in Seattle (Olsen et al., 2004). These levels were comparable to national levels in adults at the time (CDC – NHANES, 2017).

Serum levels of these PFAAs have declined following phase-outs in domestic production and use. Domestic production and use of these PFAS and their precursors began in the year 2000, but they have not been federally banned. Some uses were allowed to continue under EPA Significant New Use Rules. These PFAS may also be in imported materials and products (see Appendices 3: Sources and Uses, and 9: Regulations).

It is important to acknowledge that we have limited ability to measure and identify human exposures to PFAS. There are thousands of PFAS compounds, but only about a dozen have been regularly measured in blood serum of people (CDC – NHANES, 2019; Olsen et al., 2017). Individual research studies have employed expanded panels with up to 62 PFAS. PFOS, PFOA, and PFHxS still predominate among the identified PFAS in results (Miaz, 2020; Nakayama, 2020; Yeung, 2015).

Investigations of human serum with nontargeted methods, however, show that even expanded panels of PFAS can miss a considerable portion of organofluorine compounds in serum (Miaz, 2020; Yeung, 2015). In a recent Swedish study, 50% of the total extractable organofluorine content observed in archived pooled maternal serum samples collected in 1996 was

unexplained by targeted analysis of 62 PFAS. The proportion of novel PFAS appeared to be growing over time, as only 20% of the total extractable organofluorine was explained by targeted analysis in their 2017 samples (Miaz, 2020).

Some PFAAs are highly bioaccumulative in people.

Bioaccumulation occurs because the human body readily absorbs PFAAs from the digestive tract, but only slowly eliminates them. Long-chain PFAAs accumulate in human blood serum, liver, lung, bone, and other locations in the body (Koskela et al., 2017; Perez et al., 2013). Some PFAAs can cross the placenta into fetal tissue (Mamsen et al., 2019). The body excretes other PFAS—such as PFBA and PFHxA—more rapidly. These are infrequently detected in the general population serum samples above $0.1~\mu g/L$, but are detected more frequently in paired urine samples (Calafat et al., 2019).

A **half-life** is one way to measure the bioaccumulative nature of a substance. It represents the time required for the serum concentration of PFAS to drop by half after removing the source of exposure (such as contaminated drinking water). Estimates of half-life vary depending on the age and gender of the population studied, the level of PFAS exposure, the level of continuing background exposure, and the length of follow-up. Mean and median estimates that are most relevant to environmental exposures are provided below.

- PFOA: 0.3 to 3.9 years (Li et al., 2018).
- PFOS: 3.3 to 4.6 years (Li et al., 2018; Olsen et al., 2007).
- PFNA: 2.5 to 4.3 years (Zhang et al., 2013).
- PFHxS: 5.3 to 7.1 years (Li et al., 2018).
- PFHxA: 32 days (Russell et al., 2013).
- PFBS: 27 days (Olsen et al., 2009).
- PFBA: 72 hours (Chang et al., 2008).

Serum levels of individual PFAAs vary across lifestage and sex

In the general population, children's serum levels of the primary PFAAs measured are often similar to or lower than adult levels. A study of 598 children ages 2-12 years in 1994 and 1995 by Olsen et al. reported that children were comparable to adults in their PFOS and PFOA levels. However, children had substantially higher 95th percentile values of PFHxS and FOSAA (Olsen et al., 2004). The higher levels in this subset of children may have been related to child-specific patterns of exposure to household items, such as treated carpet and textiles. In a 2009 study, 1-to-2-year old children had median serum levels of PFOA, PFOS, and PFHxS lower than adults levels measured in NHANES from the same years (Schecter et al., 2012). This study reported no difference between genders among young children. A nationally representative subsample of 639 children, ages 3-11 years, in NHANES 2013 -2014 detected PFOA, PFOS, PFHxS, and PFNA in all children at levels that were similar to those of NHANES 2013 -2014 in adolescents and adults (Figure 40) (Ye et al., 2017). See Figure 40.

This is in contrast to children living in areas with contaminated drinking water who frequently have higher PFAA serum levels than adults due to their higher rate of intake of drinking water per pound of body weight and lactational transfer from mothers (if breastfed). For example, nearly 5,000 matched pairs of mothers and children aged 1-19 years in the C8 Health Project were studied to learn more about their relative serum levels following exposure to the same household drinking water. Childen aged 1-5 years had mean serum levels of PFOA that were 44% higher than their mothers, and serum levels of PFOS were 30% higher than their mothers. Children's serum remained higher until age 12 for PFOA but persisted through age 19 for PFOS (Mondal et al., 2012).

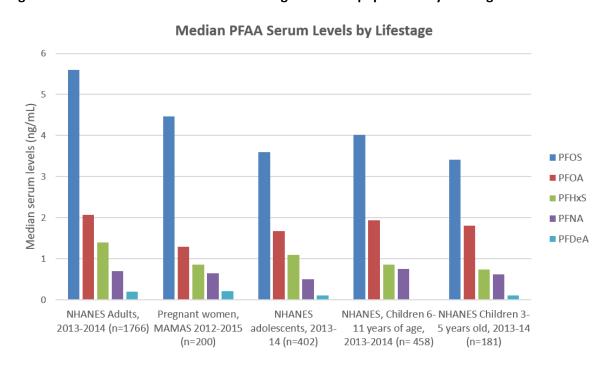


Figure 40. Median PFAAs serum levels in the general U.S. population by life stage.

Notes:

- The data on U.S. adults aged 20 and older and on adolescents aged 12 19 are from CDC NHANES (CDC – NHANES, 2017).
- Data on pregnant women are from a state of Califonia biomonitoring project called Measuring Analytes in Maternal Archived Samples (MAMAS), which is a representative sample of pregnant women statewide (California, 2017a).
- Data on children aged 3 11 years comes from a representative subset of NHANES published in Ye et al. (Ye et al., 2018).
- None of these studies were conducted in populations with known industrial sources of elevated PFAAs exposure.
- Left bar is PFOS, second bar from left is PFOA, middle bar is PFHxS, second bar from right is PFNA, right bar is perfluorodecanoic acid (PFDeA).

Breastmilk can be a major contributor to infant exposure. A recent German study of PFAA serum levels in breastfed infants analyzed archived samples from a 1990 study. At one year of age, the mean serum levels of PFOA, PFHxS, and PFNA in breastfed infants exceeded the mean maternal serum levels. Compared to formula-fed infants, the breastfed infants had 4.4 times more PFOA, 2.2 times more PFOS, 3 times more PFNA, and 1.2 times more PFHxS (Abraham et al., 2020). A study of 100 mother-infant pairs in Sweden estimated that serum levels in exclusively breastfed infants increased 30% per month for PFOA, 45% per month for PFNA, and 40% per month for PFHxS of breast feeding (Gyllenhammar et al., 2018).

Only slight differences in PFAS serum levels are seen across adult brackets although older women have higher serum levels of PFOS, PFOA, and PFNA than younger adult women (Calafat et al., 2007). This may be due to the loss of several PFAS excretion pathways: menstration, pregnancy, and lactation (Jain, 2013; Taylor et al., 2014). The levels measured in these studies likely reflect non-occupational exposures to PFAS in diet, consumer products, and homes.

There are also observed sex differences in exposure to some PFAAs. Mean serum levels are generally lower in females post-puberty than in males. Compared to males over 11 years old, the geometric mean serum level for females over 11 years old was 24.4% lower for PFOA, 16% lower for PFNA, 40.9% lower for PFOS, and 45.6% lower for PFHxS in the latest CDC survey data (2015 – 16). The other eight PFAAs measured did not have sufficent frequency of detection to calculate a mean value for males vs. females (CDC, 2019).

PFAA exposure varies by race, ethnicity, income, and education level

In 2015 – 2016 NHANES data, PFAA serum levels varied by racial and ethnic identity. Mexican-Americans had lower median serum concentrations of five PFAS than non-Hispanic whites or non-Hispanic blacks. Participants who self-identied as Asian had on average higher levels of PFOS and several other PFAAs (See Figures 41 and 42). Figure 41 shows differences in serum PFOS, which were the most pronounced. Similar demographic findings were reported in an earlier NHANES survey (Nelson et al., 2012).

In other studies, black Americans had lower PFAA levels than non-Hispanic whites. Kingsley et al. (2018) reported that non-Hispanic black women had 18% lower serum PFOA and 43% lower serum PFHxS concentrations than non-Hispanic whites in a study of pregnant women in Cincinnati, Ohio. Serum PFOS and PFNA were also lower, but the difference was not statistically significant (Kingsley et al., 2018). In Project Viva—a study of children aged 6 – 10 years born in the Boston area—children of black mothers had lower levels of PFOA, PFOS, PFHxS, and 2-(N-methyl-perfluorooctane sulfonamido) acetic acid (Me-PFOSA-AcOH), but not PFNA, compared with children of white mothers, even after adjusting for maternal concentration during pregnancy (Harris et al., 2017). It is not clear what factors underlie the differences observed, but income level, dietary habits, and use of consumer products such as stain repellents appear to contribute (Boronow et al., 2019; Kingsley et al., 2018). See Section 7.3 Pathways of exposure for more information.

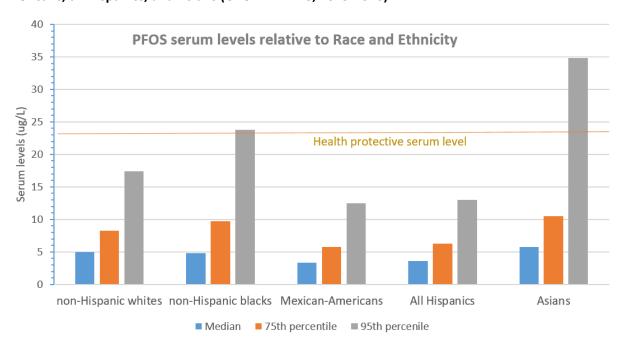


Figure 41. Demographics of PFOS serum levels in non-Hispanic whites, non-Hispanic blacks, Mexican-Americans, all Hispanics, and Asians (CDC - NHANES, 2015-2016).

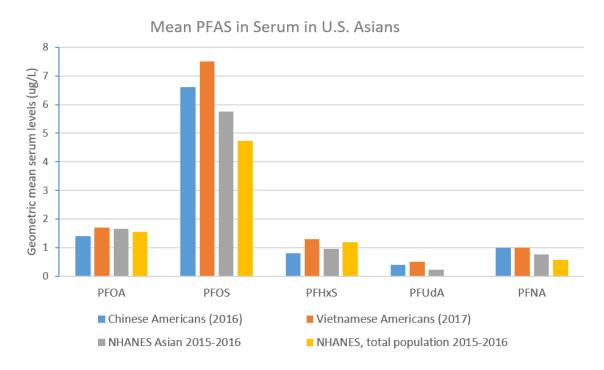
Notes:

- The orange line represents the health-protective limit for human exposure recommended by New Jersey (22.5 μg/L), New Hampshire (23.6 μg/L), and Washington (23.6 μg/L) in their recommendations for drinking water limits for PFOS (New Hampshire Department of Environmental Services, 2019; New Jersey Drinking Water Quality Institute, 2018; Health, 2019).
- Left bar is median, middle bar is 75th percentile, and right bar is 95th percentile.

The California Environmental Contaminant Biomonitoring Program investigated risk factors for elevated PFAAs in two Asian subgroups in the Asian Pacific Islander Community Exposures (ACE) Study (Biomonitoring California, 2019a, 2019b). This study recruited about 100 Chinese and 100 Vietnamese participants in San Francisco and San Jose areas in 2016 – 2017. Both subgroups in their study had even higher levels of PFOS than NHANES Asians at the mean and 95th percentile (see Figure 42).

PFAAs serum levels were significantly associated with demographic factors such as age, sex, U.S. residency, birth country, household income, and language. Specificially, higher PFAA levels were associated with a smaller portion of life spent in the U.S., presumably reflecting more time spent in countries where industrial manufacturing and emissions have continued (Xie et al., 2013). Researchers concluded that California's regional immigration and racial and ethnic patterns may contribute to differences in PFAAs observed across the statewide surveys of PFAS (Attfield, 2018).

Figure 42. Geometric mean PFAAs serum levels (μ g/L) in Asian subgroups from the ACE Study in California, compared to national norms as reported in CDC NHANES 2015 – 16. (Attfield, 2018; Biomonitoring California, 2019a, 2019b; CDC - NHANES, 2015 – 2016).



Note: Left bar is Chinese Americans (2016), second from left is Vietnamese Americans (2017), second from right is NHANES Asian (2015 - 2016), and right bar is NHANES total population (2015 - 2016).

The most recent U.S. Census estimates for Washington state demographics estimate 8.3% of state residents are Asian and 3.7% are black or African-American (U.S. Census Bureau, 2018). If the distribution of serum PFOS in the state is similar to national norms, approximately 10% of Asian and 5% of African-American residents in Washington state would have PFOS serum levels that exceed health-based protective levels recommended by New Jersey (22.5 μ g/L), New Hampshire (23.6 μ g/L) and Washington (23.6 μ g/L) (New Hampshire Department of Environmental Services, 2019; New Jersey Drinking Water Quality Institute, 2018; Health, 2019). No information was located for relative levels of PFAS in serum in U.S. or Northwest tribal populations.

Scientists don't know yet how social and economic determinants (e.g., race, ethnicity, income, education) are exactly linked with the environment, health, and well-being. Scientists believe that these associations could result from differences in socioeconomic factors, which may influence exposure and specific health outcomes.

Among a cohort of pregnant women living in eastern Massachusetts, PFAA serum levels were higher in women who were younger, less educated (but higher income), and had less educated partners (Sagiv et al., 2015). A meta-analysis confirmed that socioeconomic status (defined by

income) is an important determinant of PFAS blood levels in people. This study looked at human biomonitoring markers of PFAS exposure with education and income. The study showed consistently that a higher income is associated with higher PFAS concentrations (Buekers et al., 2018). The authors concluded that with PFAS it appears that a low socioeconomic status is not associated with an increased chemical burden.

7.2.2 Populations with elevated PFAS exposure

Occupational exposures

Manufacturing workers and those working with PFAS products

A number of occupational exposure studies of PFAAs in workers at fluorosurfactant and fluoropolymer manufacturing facilities were reviewed by ATSDR (ATSDR, 2018b). Compared to people with environmental exposures, concentrations reported in workers during the 1990s and 2000s were typically much higher (i.e., $800-10,000~\mu g/L$ for PFOA, $800-2,440~\mu g/L$ for PFOS, $200-1,850~\mu g/L$ for PFHxS).

Some professional ski waxers heat and handle PFAS-containing glide waxes during the professional competition season. Several European investigations have documented their high airborne exposure to PFAS in workspace and personal air. These studies have also shown increased serum levels of PFAS associated with this work (Freberg et al., 2010; Freberg et al., 2014; Nilsson et al., 2013; Nilsson et al., 2010; Russell et al., 2013).

Exposure information is lacking for other workers. Tanner et al. (2018) reported that in a cohort of older adults in upstate New York, those who had worked in industries known to use PFAS had higher serum PFOS and PFOA than workers who did not work in those industries (Tanner et al., 2018). Workers who may be expected to have higher exposures include those who manufacture or handle PFAS-treated paper, carpets, leather, apparel, furniture, and individuals who install carpets or provide professional carpet care services, or other retail exposures.

For example, indoor air concentrations of 14 PFAS (Fluorotelomer alcohols [FTOH]: 4:2 FTOH, 6:2 FTOH, 8:2 FTOH, 10:2 FTOH, 12:2 FTOH; fluorotelomer acrylates [FTAC]: 6:2 FTA, 8:2 FTA, 10:2 FTA; perfluorinated sulfonamido ethanols and perfluorinated sulfonamides: EtFOSA, MeFBSA, MeFOSA, N-Methyl perfluorooctane sulfonamido-ethanol [MeFOSE], MeFBSE, N-Ethyl perfluorooctane sulfonamidoethanol [EtFOSE]) were measured in various settings, with the highest concentrations in stores selling outdoor equipment, a furniture shop, and a carpet shop (Langer et al., 2010). Schlummer et al. (2013) found higher air concentrations of FTOH in carpet shops and stores selling outdoor textiles than Langer et al. (2010). Table 68 below summarizes occupational exposure estimates.

Table 68. Occupational exposure estimates (Langer et al., 2010).

Occupation	Average total PFAS air concentrations (nanogram/cubic meter) [ng/m³]	Average exposure ng/day/kg bodyweight
Retail trade workers in furniture and home furnishing stores	187	11.84
Retail trade workers in sporting goods stores	351	22.23

Firefighters

Firefighters may have contact with PFAS in class B foams (also referred to as aquous film forming foam (AFFF)) when extinguishing flammable liquid fires or during training exercises. They also have PFAS in their protective gear and may be exposed to PFAS present in smoke and dust from burning building materials. For example, serum collected from first responders after the World Trade Center collapse had 2-fold higher concentrations of PFOA and PFHxS compared to the U.S. general population. Higher exposure to PFNA, PFHxS, and PFOA was associated with work in areas that had higher exposure to dust and smoke (Tao et al., 2008).

PFAS serum levels were higher among firefighters compared to the general population in several studies (California, 2016; Dobraca et al., 2015; Jin et al., 2011; Shaw et al., 2013). A 2010 – 2011 study of 200 California firefighters showed slightly higher mean serum levels for PFOS, PFOA, and PFHxS compared to national norms for the study period (California, 2016, 2017b). A 2014 – 2015 PFAS biomonitoring study in a female cohort of firefighters and office workers reported higher exposure to PFHxS, PFUnDA, and PFNA in firefighters compared to office workers after controlling for age, race, and ethnicity. The levels of PFHxS, PFDA, and perfluorobutane sulfonic acid (PFBuS) were higher in firefighters and office workers compared to 2013 – 2014 NHANES adult women (Trowbridge et al., 2019).

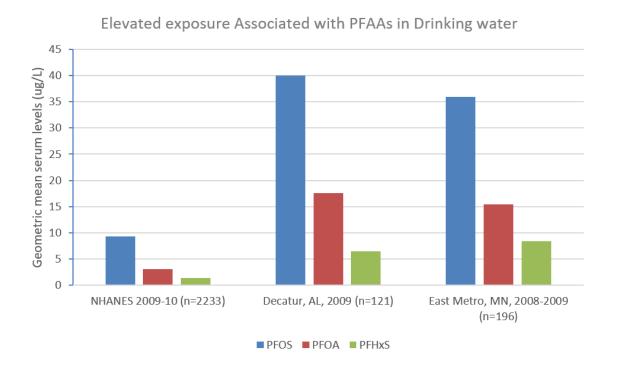
An Australian study in 149 contract firefighters who were required to train every 90 days with AFFF reported that median serum level of PFOS was ten times higher in firefighters than the general population of Australia and Canada. PFHxS was 15 times higher. PFOA, PFNA, PFHpA, and PFDA were not elevated in firefighters relative to the general population. Median serum levels in the group of firefighters studied were $66~\mu g/L$ PFOS and $25~\mu g/L$ PFHxS. Maximum detections were $391~\mu g/L$ PFOS and $277~\mu g/L$ PFHxS. Interestingly, study participants who had worked ten years or less (only after the phase out of PFOS-based AFFF) had levels of PFOS that were similar to the general population. A small study by the same research group used non-targeted analysis (quadrupole time-of-flight tandem mass spectrometry) to identify additional novel PFAS that were more frequently detected or unique to the firefighters studied (Rotander, Karrman, et al., 2015; Rotander, Toms, et al., 2015)

A small occupational exposure study in Finland analyzed 12 PFAS in the serum of eight firefighters after each of three training sessions with AFFF and simulated aircraft fires. Serum concentrations of PFHxS and PFNA were, on average, slightly higher in serum after the training sessions. Neither of these PFAS were ingredients in the foam being used, but may have been metabolites of precursor PFAS or from another part of the process (Laitinen et al., 2014).

Communities with PFAS contamination in drinking water

A large number of U.S. drinking water supply wells have tested positive for PFAS since testing began in 2013 (Environmental Working Group (EWG), 2020b). Below, we highlight three examples of communities with drinking water contamination that also had serum testing. Figure 43 shows the average community concentrations of serum PFAAs in two communities in which drinking water was impacted by industrial emissions and waste practices.

Figure 43. Elevated serum PFAAs levels ($\mu g/L$) in communities with drinking water impacted by industrial sources compared to the U.S. general population (ATSDR, 2013; CDC - NHANES, 2017; Minnesota Department of Health, 2020).



Note: Left bar is PFOS, middle bar is PFOA, and right bar is PFHxS.

In Decatur, Alabama, a PFAS manufacturer reported in 2007 that it had been unknowingly discharging PFCAs into the Decatur Utilities Dry Creek Wastewater Treatment Plant. Sewage sludge from that facility had been applied repeatedly as a soil amendment to about 5,000 acres of privately owned agricultural fields (176 fields on 35 farms). Between 2007 – 2009, EPA investigators detected PFAAs in the Decatur Utilities biosolids and in surface water,

groundwater, and drinking water at and near the site of land application. PFAS concentrations in drinking water were not fully reported. Three out of 20 private wells had PFOA and PFOS concentrations above the EPA provisional health advisory level at the time. Maximum detected levels were 365 ng/L PFOS and 2,200 ng/L PFOA. The public water system reported 155 ng/L PFOA in finished water in 2006 and 70 ng/L PFOA in finished water in 2010.

In April 2010, ATSDR tested serum samples from 153 people who lived and worked in the affected area. Median serum PFAA concentrations of 121 residents served by the public water system were 18.1 μ g/L PFOA, 39.3 μ g/L PFOS, and 7.4 μ g/L PFHxS (Figure 43). Median levels were higher in nine residents served by private wells with detectable PFAS in the water: 30.8 μ g/L PFOA, 60.8 μ g/L PFOS, and 8.3 μ g/L PFHxS (ATSDR, 2013).

The East Metro, a suburb of Saint Paul, Minnesota, is part of a larger area of Washington County, Minnesota with groundwater impacted by PFOS, PFOA, and PFHxS. This larger area is 150 square miles and affects the water supply of 140,000 Minnesotans. The sources of contamination are a large PFAS manufacting facility in Cottage Grove and disposal sites at Oakdale, Woodbury, Cottage Grove, and Lake Elmo—where the plant had disposed of wastes in the 1950s, 1960s, and 1970s (Minnesota Department of Health (MDH), 2019). Drinking water contamination in the East Metro was discovered in 2004, and water filtration to remove PFAAs was developed and installed in 2006. PFOA and PFOS levels in municipal wells ranged from nondetect to 900 ng/L. In private wells, the levels ranged from non-detect to 2,200 ng/L for PFOA and non-detect to 3,500 ng/L for PFOS (MDH, 2020).

MDH conducted a community exposure assessment in the East-Metro Area in 2008 in a random sample of residents with impacted drinking water. Mean and maximum levels detected in the 196 residents tested (Figure 43) were:

- PFOA
 - Mean 15.4 μg/L
 - Max 177 μg/L
- PFOS
 - Mean 35.9 μg/L
 - Max 448 μg/L
- PFHxS
 - Mean 8.4 μg/L
 - Max 316 μg/L

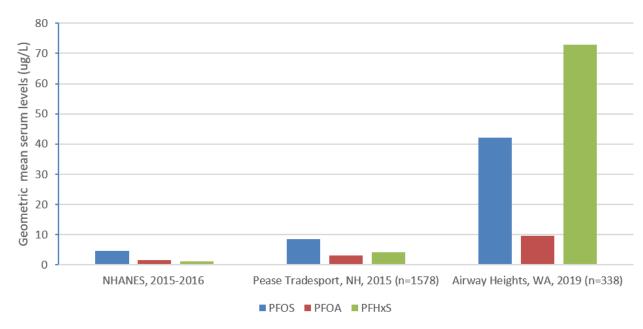
PFBA and PFBS were also detected in 28% and 23% of the samples, respectively. MDH conducted additional biomonitoring in 2010 and 2014 to confirm that water filters were working to reduce exposure. Over the six years of follow-up, average individual levels of PFOS went down by 45%, PFOA by 59%, and PFHxS by 34% (MDH, 2020).

Biomonitoring studies have also documented elevated PFAS exposure in communities where firefighting foam contaminated their residential drinking water with PFAS. Figure 44 shows the mean serum concentrations in three communities compared to national norms.

At Pease International Tradeport in Portsmouth, New Hampshire, PFAAs were reported in three production wells of a public drinking water system in May 2014. The Tradeport was developed on the site of the former Pease Air Force Base and contains more than 250 businesses, public offices, restaurants, and childcare facilities employing more than 9,500 individuals. The suspected contamination source was firefighting foam used at the former base. Levels of PFAS detected were highest in the Haven well—PFOS (2,500 ppt), PFOA (350 ppt), and PFHxS (830 ppt)—prompting the city to shut down the well. Two other wells had lower levels of these PFAS, but concentrations exceeded the 2016 EPA advisory level for PFOA and PFOS. These wells were fitted with PFAS filtration systems in 2016 (New Hampshire Department of Health and Human Services (NHDHHS), 2016).

Between April and October 2015, the NHDHHS conducted blood testing in 1,578 individuals who had lived on, worked on, or attended child care at Pease Tradeport. Three PFAAs were significantly elevated relative to national norms (Figure 44). Maximum serum levels detected were 95.6 μ g/L for PFOS, 32 μ g/L for PFOA, 116 μ g/L for PFHxS, 5.2 μ g/L for PFNA, and 5.6 μ g/L for PFDeA. Six percent of the participants reported current or past experience as a firefighter, and may have had occupational exposure as well. Firefighters had significantly higher levels of PFOS and PFHxS compared to other participants (NHDHHS, 2016).

Figure 44. Geometric serum levels (μ g/L) in samples collected from people who lived, worked or received childcare at Pease Tradeport in New Hampshire and Airway Heights, Washington. Levels are compared to national norms as measured by the CDC NHANES for the same time period.



Note: Left bar is PFOS, middle bar is PFOA, and right bar is PFHxS.

The City of Airway Heights, Washington, near Fairchild Air Force Base, was selected to participate in a national exposure assessment of eight communities known to have had PFAS in their drinking water. PFAS in firefighting foam used at the base is the key suspected source of the water contamination (see Section 7.4.3 for more information about this site). Serum levels measured in study participants, 2.5 years after the contamination was removed from community drinking water, were elevated for PFOS, PFHxS, and PFOA (See Figure 44). The study is being conducted by the Agency for Toxic Substances and Disease Registry (ATSDR, 2020).

The mean serum levels of PFHxS and PFOS reported in the Airway Heights exposure assessment are higher than those reported by other U.S. studies of communities exposed to PFAS via drinking water (see Decauter, AL; East Metro, MN; Pease Tradeport, NH above) and lower than serum levels measured in residents of Ronneby, Sweden (Andersson et al., 2020). In 2013, high levels of PFOS and PFHxS were found in the drinking water from one of the two waterworks supplying the municipality of Ronneby in Sweden (population 28,000 residents). The primary source of drinking water contamination was also firefighting foam. Drinking water concentrations were 8,000 ng/L PFOS and 1,700 ng/L PFHxS at the time the contamination was dicovered. In follow-up biomonitoring studies, median serum levels were 257 μ g/L (PFHxS), 280 μ g/L (PFOS), and 15 μ g/L (PFOA) in the group of residents exposed to contaminated water at home for at least one year during 2005 – 2013. Median levels were 257 ng/mL(PFHxS), 280 ng/mL (PFOS), and 15 ng/mL (PFOA) in the group with late exposure (contaminated water at home at least one year during 2005 – 2013).

7.3 Sources and pathways for human exposure

The primary pathways of human exposure to PFAAs are:

Non-point or diffuse sources:

- Dietary exposure to PFAS in the global environment.
- Eating foods that have been in contact with PFAS-coated food papers.
- Swallowing or inhaling indoor dust and air in homes, offices, and other buildings with PFAS-containing consumer products, such as treated carpets or furniture.
- Contact with consumer products that contain PFAS ingredients such as certain cleaning products, cosmetics, carpet treatments, car washes, waterproofing sprays, and dental floss.

Local sources around a release site:

- Drinking contaminated water.
- Eating fish and shellfish or wild game from contaminated areas.
- Eating crops or animal products (meat, eggs, milk) from farms with contaminated soils, water, or feed.

Work exposures:

- Making or processing PFAS-containing materials at on the job.
- Using PFAS-containing products on the job (e.g., firefighters).

7.3.1 Drinking water

Drinking water has been a significant source of human exposure in areas where significant PFAS contamination has occurred (see Section 7.2.2.2). Ingestion is the primary route of absorption from drinking water since the ionic forms of PFAAs found in water are not readily absorbed through skin (ATSDR, 2018b). Drinking water may be contributing to background exposures as well. Low levels of eight PFAAs (PFOS, PFOA, PFHxS, PFNA, PFHpA, perfluoro-n-pentanoic acid (PFPeA), PFUnDA, and PFDoDA) were detectable at very low levels in more than 30% of the archived U.S. tap water samples from 1990. Even these low levels of PFOA and PFNA in tap water (0.57 ng/L and 0.13 ng/L, respectively) were associated with higher plasma concentrations of PFOA and PFNA in household members. The authors estimated that tap water explained about 20% of residents' exposure for these two compounds (Hu et al., 2019).

According to Hu et al. (2016), the most significant two risk factors for detection of six PFAS in U.S. public drinking water systems in EPA's Unregulated Contaminant Monitoring Survey (UCMR3) were:

- Proximity to military fire training areas that used PFAS-containing products.
- Proximity to industrial sites that made or used PFAS.

More information about drinking water contamination is presented in <u>Appendix 4: Fate and Transport</u>.

Several large studies of long-term human exposure to PFOA in community drinking water have observed that average serum levels in the population are about 100 times the drinking water concentration (i.e., serum: drinking water ratio of 100:1) (Pitter et al., 2020; Post et al., 2012; Post et al., 2013). PFOS in community drinking water is estimated to result in average serum concentrations 172 times the chronic concentration in drinking water (Egeghy & Lorber, 2011; New Jersey Drinking Water Quality Institute, 2018).

These approximate ratios were also observed in a recent study of California teachers who lived in zip codes with detectable but modest drinking water levels of PFOS and PFOA as measured in the UCMR3 study (Hurley et al., 2016). Serum levels in any specific community are likely to relate to:

- How long the drinking water has been contaminated.
- Timing of serum sampling relative to when the exposure occurred.
- Individual consumption and use patterns of drinking water.

- Co-exposure to food.
- Consumer products.
- Other unknown sources.

7.3.2 Food

Food is another primary way most people are thought to be exposed to the PFAAs commonly detected in human serum (ATSDR, 2015; Jain, 2018; Poothong et al., 2020). In the U.S. and Canada, PFOA and PFOS were detected in some snack foods, vegetables, oils and butter, meat, dairy products, wild and farmed fish, shellfish, fast food, and microwave popcorn (Schecter et al., 2010; Tittlemier et al., 2007). In two small recent surveys by the Federal Drug Administration (FDA), few detections of 16 PFAS were reported in a wide variety of foods in the U.S. food supply. The exception was detection of PFOS (86 – 87 pg/g) in ground turkey and tilapia.

More extensive testing for PFAS in the food supply has occurred in Europe. The EFSA Panel on Contaminants in the Food Chain recently assessed more than 69,433 analytical results for 26 PFAS in common foods sampled across 16 countries of the European Union. Many samples were below the analytical detection limits for the PFAS tested. Of the samples that were positive, fish, meat, and eggs generally had the highest concentrations. Vegetables and fruits had low concentrations but frequent detections. The EFSA panel concluded that fish and other seafood, eggs, meat, and fruit were important contributors to chronic exposure of PFOS and PFOA in Europeans. Vegetables and drinking water were also important contributors to chronic PFOA exposure. The same key dietary contributors emerged when they considered combined exposure to PFOA, PFNA, PFHxS, and PFOS (EFSA, 2018, 2020).

Two large U.S. dietary studies based on CDC NHANES data found that higher fish and shellfish consumption were associated with higher serum levels of PFAAs (Christensen et al., 2017; Susmann et al., 2019). In Susmann et al. (2019), higher levels of serum PFNA, PFDA, and PFOS were associated with fish consumption reported in the past 24 hours, 7 days, or 30 days and with seafood consumption in the past 12 months. Shellfish consumption reported for these same recall periods was associated with higher serum levels of PFNA and PFDA.

This study also found that serum PFAAs were positively associated with consumption of popcorn, fast food, and pizza, and were inversely associated with the number of food meals eaten at home (Susmann et al., 2019). Similar findings were reported in another dietary exposure study of California children and adults. Positive associations were found between PFAS serum concentrations and consumption of butter and margarine, fish, meat products, and microwave popcorn (Wu et al., 2015). PFAAs in fast food wrappers, non-stick baking paper, grease-proof take-out containers, and microwave popcorn bags can migrate out of the paper into food (Begley et al., 2008; Begley et al., 2005; European Commission and Cordis, 2012; Geueke, 2016)

At contaminated sites, food raised for human consumption could have elevated PFAS. Site investigations and research studies have demonstrated that when PFAAs are present in soil, drinking water, irrigation water, or animal feed, they can transfer to livestock and food crops. Shorter chain PFCAs appear most likely to accumulate in plants (Ghisi et al., 2019). Long-chain perfluorooctane sulfonic acids (PFSAs) and PFCAs are the most likely to accumulate in animals. The degree of uptake and accumulation depends on the PFAA, the plant or animal species, and the level of contamination. For example, low levels of PFAAs in soil did not cause significant uptake in vegetables in a Minnesota garden study (Scher et al., 2018). PFAA uptake has been observed in:

- Chicken meat and eggs (Australian Government Department of Defense, 2017; Yeung et al., 2009; Yoo et al., 2009).
- Beef cattle and other livestock (Lupton et al., 2012, 2014; Numata et al., 2014).
- Dairy animals and their milk products (Kowalczyk et al., 2012; Kowalczyk et al., 2013; Maine Department of Environmental Protection, 2017).
- Fish (Hansen et al., 2016).
- Cereals (wheat, rye, oats) (Liu et al., 2019; Stahl et al., 2009).
- Vegetable crops (Bizkarguenaga et al., 2016; Lechner & Knapp, 2011; Liu et al., 2019).

See <u>Appendix 6: PFAS Ecotoxicology</u>, <u>Section 6.2 Bioaccumulation</u>, about uptake in plants, fish, birds, and other animals, and <u>Appendix 8: Biosolids</u>, <u>Section 8.6 Literature review of biosolids</u> <u>land application effects</u>, regarding uptake by plants from biosolids application, for more information.

7.3.3 Consumer products

Contact with consumer products is a potential source of human exposure to some PFAS. Residues on hands can also be absorbed if swallowed. PFAS-treated consumer products can be a source of PFAAs and their precursors in indoor dust and indoor air. People inhale and ingest contaminated air and dust, leading to human exposure to PFAS.

Carpets and carpet care treatments

Carpets have been routinely treated with PFAS-based protectants to make them resistant to stains and easier to clean (see Appendix 3: Sources and Uses, Section 3.3.2 PFAS in a typical home). Several investigations have attempted to characterize human exposure to PFAS from treated carpets and carpet care products. Karaskova et al. (2016) found that the combined concentrations of 20 PFAS on carpeted floors was higher than other floor types (Karaskova et al., 2016). Several other studies have also observed higher concentrations of various PFAS in the indoor environment in homes and offices with carpet (Fraser et al., 2013; Gewurtz et al., 2009; Kubwabo et al., 2005). Because children spend more time on or near the floor and have relatively high respiration rates and frequent hand-to-mouth activity, they have higher exposures to contaminated air and house dust, and have more direct skin and mouth contact

with carpet. Karaskova et al. (2016), Tian et al. (2016), Shoeib et al. (2011), and Trudel et al. (2008) have found that house dust is an important PFAS exposure route for toddlers.

Studies show that children with carpets in their bedrooms have higher concentrations of PFOS, PFHxS, and Me-PFOSA-AcOH in their bodies than children with other types of bedroom flooring (Harris et al., 2017). Boronow et al. (2019) found that women living in homes with treated carpet had higher exposure to PFAS, PFNA, and PFDeA.

In a Canadian study, a home where carpets had been treated approximately eight times with Scotchgard carpet protection formulations over 15 years had elevated levels of PFHxS, PFOS, and PFOA in house dust (2,780 nanograms per gram [ng/g], 1,090 ng/g, and 550 ng/g dust respectively). Serum levels of family members were also elevated (PFHxS ranged 27.5 – 423 μ g/L, PFOS ranged 15.2 – 108 μ g/L, and PFOA ranged 2.40 – 9.23 μ g/L). The authors concluded that the ingestion or inhalation of household dust was the likely pathway of their elevated exposure (Beesoon et al., 2012).

A 2016 Danish EPA (DEPA) study estimated the potential PFAS exposure to young children from five children's rugs (intended for those younger than 15 years old) that tested positive for organofluorine content in a survey of 21 rugs. PFOA and 6:2 fluorotelomer sulfonic acid (6:2 FTSA) were the primary PFAS detected in further targeted analysis. Using worst case assumptions about ingestion of carpet dust by toddlers, DEPA estimated 0.05 ng/kg-day exposure to PFOA with all other PFAS contributing another 0.04 ng/kg-day. This was well below the Danish screening value at the time (TDI of 30 nanograms per kilogram per day [ng/kg-day]) and is also below Washington state's recommended reference dose (RfD) for PFOA of 3 ng/kg-day (DEPA, 2016). It is important to note that their targeted analysis only measured a small percentage of the total organofluorine content present.

Cosmetics and personal care products

Polymeric and non-polymeric PFAS ingredients are used in sunscreens, creams and lotions, foundation and concealers, shampoos, nail polish, eye makeup, and denture cleaners (DEPA, 2018; Geueke, 2016). Although none of the cosmetic ingredients listed in two surveys were PFCAs, testing by Fujii et al. (2013) and the DEPA (2018) showed that the majority of cosmetics with PFAS ingredients did contain a range of PFCAs (C6 – C14). As identified in Appendix 1: Chemistry, Section 1.1.1 PFAS terminology, C6 represents a PFAS with a carbon chain length of 6, and so on. Presumably these occur as breakdown products of precursors or residuals from ingredient manufacturing. The listed ingredients with the highest levels of measured PFCAs were ammonium C6 – 16 perfluoroalkyl ethyl phosphate and C9 – C15 fluoroalcohol phosphate. Sunscreens and foundation had the highest measured PFCA levels.

PFOA concentrations in a couple of samples exceeded the EU limit of 25 ng/g (DEPA, 2018; Fujii et al., 2013). The Danish EPA conducted a human risk assessment based on the above survey results. DEPA used PFOA as the reference chemical because data on skin absorption and toxicity

were available. They assumed 2% absorption across skin for ionic PFOA and 70% dermal absorption for neutral PFOA. Their risk assessment did not find a likely risk to consumers from any given product, even using conservative assumptions (DEPA, 2018). However, if they had used EFSA's 2018 TDI of 6 nanograms per kilogram (ng/kg) per week for PFOA as a screening level, most of their dermal exposure estimates from single products were just below or considerably above (up to 35 times higher) that screening level.

Indoor air and dust

As certain consumer products degrade by abrasion and normal wear and tear, they may contribute to PFAS levels in indoor dust and air. Indoor air is inhaled by occupants and indoor dust is both inhaled and swallowed, especially by young children who crawl on the floor and engage in hand-to-mouth activity.

In 2000 – 2001, a number of PFAS were measured in U.S. indoor dust samples collected from 112 homes and ten day-care centers in North Carolina and Ohio. PFOA, PFOS, and PFHxA were the most commonly detected (median concentrations in indoor dust were 142, 201, and 54.2 ng/g, respectively). Some dust samples had very high concentrations of PFOS and PFHxS (up to 12,100 and 35,700 ng/g respectively) (Strynar & Lindstrom, 2008). Lower mean concentrations of PFAA were detected in indoor dust samples from 152 homes in Vancouver, Canada. PFOA, PFOS, and FOSE were prominent in dust samples. 8:2 FTOH had a mean level of 88 ng/g dust and 2.9 nanograms per cubic meter (ng/m³) in air.

Exposure estimates in this study shows that air, rather than dust, was the primary exposure pathway in adults whereas in toddlers, dust was a significant pathway of exposure (Shoeib et al., 2011). In another exposure assessment, PFOA, PFOS, and PFNA measured in serum of pregnant women in Vancouver, Canada in 2007 – 2008 correlated with precursor chemicals measured in the indoor air of participants' homes. Specifically, positive associations were discovered between airborne 10:2 FTOH and serum PFOA and PFNA, and between airborne N-methyl perfluorooctane sulfonamido ethanol (N-MeFOSE) and serum PFOS (Makey et al., 2017). The median PFOA levels in dust observed in the U.S. and Canada are higher than the levels found in European countries (D'Hollander et al., 2010). This may be due to differences in PFAS use and sources.

PFOA, PFOS, and PFHxS were also routinely detected in indoor dust from homes, offices, and vehicles in Boston, Massachusetts in 2009 (Fraser et al., 2013). Another Boston study sampled PFAS in air in 30 offices in seven buildings, and compared this to serum levels in 31 office occupants. This Boston study detected a range of newer PFAS in more than 90% of the indoor air samples of offices, and reported maximum levels of 70 ng/m³ for 8:2 FTOH, 12.6 ng/m³ for 10:2 FTOH, and 11 ng/m³ for 6:2 FTOH (Fraser et al., 2012). Collectively, FTOHs in air significantly predicted PFOA in serum of office workers (p < 0.001) and explained approximately 36% of the variation in serum PFOA concentrations. PFOS in serum was not associated with air levels of perfluorooctane sulfonamides (PFOSAs) or N-ethyl perfluorooctane sulfonamido

ethanols (N-EtFOSEs). The compounds 8:2 FTOH and 10:2 FTOH are precursors to PFOA, and represent a potential inhalation pathway.

Norwegian investigators also reported significant positive associations between household dust and air levels of certain PFAS and the serum PFAS concentrations in occupants. This study collected indoor air, dust, duplicate diet, and blood samples in 2013 – 2014 as part of a study to examine various pathways of PFAS exposure in 61 adults in the Oslo area. Air intakes of 10:2 FTOH were associated with serum PFUnDA, 8:2 FTOH inhalation correlated with serum PFNA, and MeFOSE and EtFOSE inhalation correlated with whole blood PFOSA. Air intake of 6:2 FTOH was not associated with measured serum PFCAs. Dust samples of PFOA and PFNA were also associated with serum levels. The authors estimated that PFAAs in dust contributed to 2% of median exposures and PFAAs in indoor air contributed about 3% to median exposures in study participants. Specific individuals in the study, however, had much higher estimated contributions from household dust and indoor air—up to 95% for dust intake and 72% for air intake (Poothong et al., 2020).

7.4 Known areas of PFAS contamination in drinking water aquifers in Washington state

PFAS contamination has been found in groundwater supplies used for drinking in five main areas of the state (Figure 45). Where levels exceeded the lifetime health advisory level for PFOA and PFOS of $0.07~\mu g/L$ set by EPA in 2016, water systems and the military have taken mitigation actions to meet the federal health advisory. The primary source suspected in these areas is a firefighting foam called AFFF that contained PFAS. The UCMR3 tested for six PFAS in all public drinking water systems (PWS) that serve more than 10,000 people and in a representative sample of smaller systems between 2013 and 2015. Voluntary testing by the Navy, Air Force, and Army has discovered additional drinking water contamination in private and public wells on or around four military bases between 2016 and 2020. Proactive testing by nearby public water systems has discovered additional wells that are impacted.

Additional sources of PFAS at contaminated drinking water sites may be uncovered by ongoing investigations. Available state data are presented below with a summary of actions taken in each area. In Figure 45, red dots mean that at least one private water system (PWS) well tested positive to PFAS compounds. The source of data for red dots include the Unregulated Contaminant Monitoring Survey (UCMR3), voluntary testing by the Navy, Air Force, and Army on or around four military bases between 2016 and 2020, and proactive testing by nearby PWS.

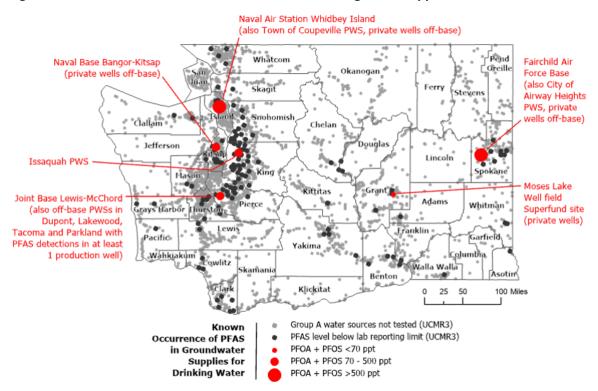


Figure 45. Known areas of PFAS contamination in drinking water supplies.

7.4.1 City of Issaquah, 2015 – 2016

As part of EPA's UCMR3 testing, the City of Issaquah discovered PFOS, PFHxS, and smaller amounts of PFOA, PFNA, PFBS, and PFHpA in one production well in their PWS. PFOS concentration in the affected well ranged from $0.4-0.6~\mu g/L$ and PFHxS ranged from $0.20-0.24~\mu g/L$. Concentrations of other PFAS were less than $0.03~\mu g/L$. Water from this well was blended in a ratio of 1:4 with a deeper adjacent well that was PFAS-free before it entered the distribution system. After blending, the water level did not exceed the 2009 provisional EPA health advisory, which was $0.4~\mu g/L$ for PFOA and $0.2~\mu g/L$ for PFOS (EPA, 2009).

In November 2015, additional sampling across the Issaquah system detected PFOS at 0.106 μ g/L at the entry point of the two blended wells, and levels ranging from 0.068 – 0.038 μ g/L in more distant areas of the distribution system. At each site, PFHxS was present at about half the PFOS concentration.

In January 2016, the city shut down the impacted well and eventually invested \$600,000 to install a granular activated carbon (GAC) treatment system in May 2016. Since June 2016, the treatment system has been effective at removing PFOA and PFOS, and is routinely tested for performance.

The city investigated the potential sources of contamination, and concluded that the likely source was the Eastside Fire and Rescue headquarters, about a mile up gradient. Soil samples in a firefighting training area at the headquarters contained PFOA and PFOS from firefighting

foam. One monitoring well and two drinking water production wells operated by nearby Sammamish Plateau Water system were also found to contain PFOA and PFOS at trace levels (Sammamish Plateau Water, 2016, 2018). These wells continue to be monitored.

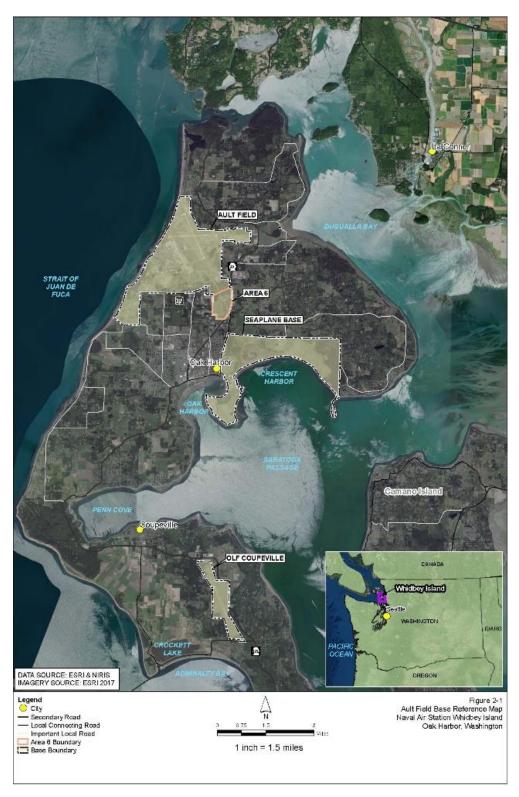
7.4.2 NAS Whidbey Island, 2016 - 2019

In 2016, the NAS Whidbey Island began offering PFAS water testing in off-base drinking water wells located within one mile from potential or known release sites of AFFF on the base. Water results were compared to the 2016 EPA health advisory level for PFOA and PFOS. Subsequent sampling rounds have "stepped out" in a half-mile radius from exceedances of PFOS or PFOA in drinking water.

In October 2018, the Navy identified PFAS in a stormwater drain near Hangar 6 at Ault Field and in an associated stormwater drainage system that empties into Clover Valley Stream and Dugualla Bay. As a result of this new information, the Navy expanded its off-base drinking water sampling near Ault Field, specifically without one-half mile of Clover Valley Stream and Dugualla Bay. No exceedance of the EPA health advisory was identified.

As of May 2020, the Navy has tested a total of 281 private or community drinking water wells as part of this investigation. PFOS or PFOA were detected in 26 wells (9%). Sixteen of these wells exceeded the EPA health advisory level. Eight are near the Outlying Landing Field (OLF) southeast of the Town of Coupeville, two are near Ault Field in Oak Harbor, and six are near a former Navy disposal site used from the 1960s to 1990s for industrial and household wastes (referred to as Area 6). See Figure 46 for a map with these locations.

Figure 46. Arial map of NAS Whidbey Island.



While PFOS was the leading PFAS detected near Ault Field and Area 6, contamination near OLF was dominated by PFOA. The Navy continues to conduct biannual sampling of all drinking water

wells with PFAS detections. It also monitors wells adjacent to properties with wells above the EPA health advisory. The Navy provides bottled water to residents whose results for PFOA or PFOS exceed the EPA health advisory until a long-term solution is developed and implemented. Long-term solutions may include connecting homes to a nearby public water system, installing whole house filtration systems, and installing a new drinking water well.

In addition to private wells, two Group A public water systems in the area were found to have PFAS detections. The Town of Coupeville is located near OLF. Coupeville's water system blends water from multiple wells, most of which are not impacted, so tap water has remained below the EPA health advisory for PFOA and PFOS (Hinds, 2017). Water testing results from two specific wells in March 2019 showed PFOA concentrations ranged from 0.022 - 0.061 µg/L (Anatek Labs Inc., 2019). Detectable levels of PFHxS and PFHpA were also present (see Table 69). As part of the long-term solution to address PFOA above the EPA's health advisory in eight private wells, the Navy recently installed a granulated activated carbon treatment system to remove PFAS from the Town's blended water system. The treatment system is routinely tested to ensure that these PFAS are removed. The second Group A public water system with PFOA and PFOS combined above the EPA health advisory is a mobile home park near Area 6. The long-term solution being considered for this system is connection to the Oak Harbor Water System. The Navy also sampled the Admiral's Cove Water District. The water district had no detections of PFOS and PFOA at the time of sampling. At least 12 small public water systems on Whidbey Island tested their wells independently from the Navy's testing for PFAS. None reported detections.

The Navy has conducted a number of public meetings where they have presented health information and answered questions about the drinking water investigation. The Navy continues to make progress on its on-base PFAS source investigation and is implementing a policy regarding removal, disposal, and replacement of legacy AFFF. No firefighting training is occurring on base with PFAS-containing foams.

7.4.3 Fairchild Air Force Base, 2017 - 2019

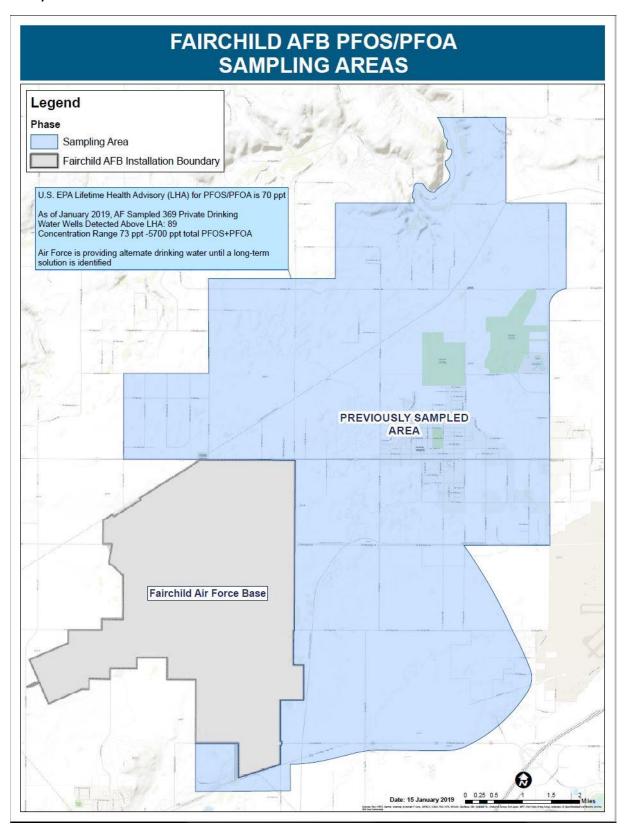
Fairchild Air Force Base (AFB) detected PFAS in groundwater monitoring wells on the base, in monitoring directed by the Department of Defense (DOD). Drinking water on the base comes from three wells located several miles east (northeast of the base near the Spokane River), and a well located on the southern tip of the base. These wells are not contaminated with PFOS or PFOA. Based on groundwater monitoring results, Fairchild AFB conducted off-base testing for PFAS in public and private drinking water wells in several phases. They detected PFAS in private wells east of the base, municipal wells for the City of Airway Heights northeast of the base, and other community and private wells to the north and northeast of the base.

As of January 2020, the Air Force has tested 372 private residential drinking water wells. One hundred and sixty nine (169) residential wells had detectable levels of PFAS and 88 of those wells currently exceed the EPA health advisory level for PFOA and PFOS (Mark Loucks, 2020).

The maximum detected level of PFOA and PFOA combined was 5,700 ppt total in a private well. The Air Force policy is to immediately notify well owners and provide bottled water if levels for PFOS and PFOA in drinking water exceed the EPA health advisory level. Bottled water is provided until a long-term solution is developed and implemented. According to a January 2020 progress report, the Fairchild AFB has installed 78 residential GAC treatment systems on impacted residential wells. Where a GAC filtration system is not feasible, the Air Force is connecting private wells to the City of Airway Heights water system (Fairchild AFB, 2018; Mark Loucks, 2020).

The Air Force sampled four municipal wells, two from the City of Medical Lake and two from the City of Airway Heights. Only the two Airway Heights wells had detectable levels of PFAS (see Table 69). The concentration of PFOS and PFOA in the Airway Heights wells were $1.1-1.2~\mu g/L$ PFOS and $0.3-0.32~\mu g/L$ PFOA in the affected wells. These levels are about 17 times higher than the EPA health advisory level for PFOS and PFOA.

Figure 47. Sampling area for private wells around the Fairchild Air Force Base (Fairchild Air Force Base, 2019).



In response to PFAS detection in April 2017, the City of Airway Heights public water system closed their contaminated wells and used an emergency intertie with the City of Spokane to flush their system with clean water. Flushing included draining reservoirs and water towers. During the flushing, the city warned residents west of Hayford Road to not drink or cook with water from city pipes, and Fairchild AFB provided bottled water to city residents. After testing throughout the water system confirmed that PFAS were sufficiently flushed, the water system resumed delivery with water from the City of Spokane. The city added another connection to the City of Spokane to supply drinking water and the Air Force installed a treatment system on municipal well #9 to supplement the Airway Heights water supply during high-demand summer and fall months. This system was operational in the Fall of 2018 (Fairchild AFB, 2018).

According to Fairchild AFB, the base has transitioned to a safer foam that is based on C6 fluorochemistry. Fairchild no longer uses AFFF during live fire training. Fire trucks on base are outfitted with a test system that prevents any foam discharge during equipment testing. AFFF use is limited to emergency responses with immediate containment requirements. The Strategic Environmental Research and Development Program (SERDP), Environmental Security Technology Certification Program (ESTCP) is funding research on new fluorine-free firefighting foam formulations that can meet the military's performance requirements (Mil-Spec), and are readily biodegradable (Ananth, 2018; Payne, 2018; Tsang, 2018).

7.4.4 Joint Base Lewis-McChord

The Army's Fort Lewis facility and the Air Force's McChord Field facility are operated as a joint military base, the Joint Base Lewis-McChord (JBLM), but have separate water systems.

Fort Lewis Water System

Fort Lewis monitored seven drinking water sources as part of the UCMR3 monitoring. PFOA was detected at 0.051 μ g/L in one well and PFHpA at 0.013 μ g/L in another. Subsequent testing in November 2016 confirmed the previous detections in those two wells and showed PFOA concentration in the Fort Lewis well 17 at 0.071 μ g/L, which is just above the health advisory level (Lynn, 2017). Well #17 was taken out of service in August 2015, but has been monitored for PFAS since. According to a Freedom of Information Act request, Army drinking water testing in 2018 detected 0.144 μ g/L total PFAS concentration at well 17 Fort Lewis with seven PFAAs detected (PFBS, PFHpA, PFHxS, PFHxA, PFNA, PFOS, PFOA) (Environmental Working Group, 2020).

The November 2016 testing also revealed additional wells with PFAS contamination. A well that serves the military golf course near DuPont had levels just above the health advisory level. Bottled water was supplied at that facility, and point-of-use treatment devices are now used to reduce exposure to PFAAs. The primary source of drinking water (Sequalitchew Springs and infiltration gallery) for the vast majority of the main base generally has $0.015-0.020~\mu g/L$ of PFOS and PFOA combined.

McChord Field

McChord Field was not involved in UCMR3 monitoring because the population served by its water system at that time was below 10,000. In the November 2016 monitoring, PFOS and small amounts of PFOA were reported in two drinking water wells serving McChord Field at combined concentrations of 0.250 and 0.216 μ g/L (Lynn, 2017). According to a Freedom of Information Act request, Army testing in 2017 detected a total PFAS concentration of 0.303 μ g/L with five PFAAs detected (PFBS, PFHpA, PFHxS, PFOS, and PFOA) (Environmental Working Group, 2020). Both wells that contained PFOS and PFOA above the advisory level were shut down in 2017. In early 2020, JBLM installed GAC filtration systems on these two wells and two other wells with lower levels of PFAS that supply drinking water to McChord Field and the housing units for McChord.

JBLM staff believes contamination came from firefighting foam used through the early 1990s for firefighter training at several locations associated with McChord Field's runway and Fort Lewis's Gray Army Airfield, as well as other potential sources such as landfills. According to JBLM staff, use of foams containing PFAS was discontinued more than 20 years ago.

Another military site managed by JBLM, the Yakima Training Center, tested drinking water onbase for PFAAs in November 2016, and there were no detections.

7.4.5 City of Lakewood

As part of the UCMR3 monitoring, the Lakewood Water District tested five of its drinking water wells drawing from three different aquifers, and no PFAS were detected at that time. Because of detections in late 2016 at McChord Field just east of Lakewood, the water district began proactively monitoring for PFAS in their water system starting in spring of 2017. Initially only trace levels were detected in the two wells at the Ponders well field. The concentrations gradually increased and Lakewood removed these wells from service in summer 2018 and installed GAC treatment at the Ponders wellfield in late 2019. Lakewood continues to monitor and is evaluating options for PFAS detected in another well field northwest of McChord Field. All six Lakewood wells with detectable levels of PFAS tap into shallow aquifers near JBLM - McChord Field. Lakewood continues to monitor the 30 wells in its system and to update its water customers about the issue (Lakewood Water District, 2019a, 2019b).

7.4.6 City of DuPont

As part of UCMR3 testing, the City of DuPont detected levels of PFOA ($\sim 0.03~\mu g/L$) in two wells in the southwest area of its distribution system. PFOA and PFOS were not detected in the three wells serving the north and east areas of the distribution system. Between October 2018 and January 2020, DuPont was proactive in conducting follow-up monitoring for PFAS. January 2020 results show PFOA concentrations of $0.010-0.015~\mu g/L$ and PFOS concentrations of $0.005-0.009~\mu g/L$ at two Bell Hill wells. October 2019 results at two Hoffman Hill wells show PFOA levels were $0.027-0.050~\mu g/L$ and PFOS levels were $0.010-0.013~\mu g/L$.

Because of blending, the combined concentration of PFOA and PFOS entering the water system from these well fields is lower: $0.014~\mu g/L$ for Bell Hill and $0.029~\mu g/L$ for Hoffman Hills. These levels are below the EPA health advisory level. The City of DuPont has hired an engineering consulting firm to investigate the hydrology of the wells and options for next steps (City of Dupont, 2020).

7.4.7 City of Tacoma

Tacoma Public Utilities tested its South Tacoma Wellfield as part of the UCMR3 monitoring and did not detect PFAS at that time. In late summer 2018, Tacoma Public Utilities tested for PFAS in some of the individual wells at the southern end of its South Tacoma Wellfield. This was a proactive effort to understand if PFAS existed in its water sources near JBLM. One of the wells sampled (Well 10C) draws from a shallow aquifer and was used exclusively as a source of nonfluoridated drinking water by customers who collected water in their own containers at the well site. Combined PFOA and PFOS levels in this well exceeded the EPA health advisory level. Tacoma notified customers and closed the well. Tacoma's Green River source, which serves all Tacoma Water customers with the vast majority of their drinking water, showed no detections of PFAS (Tacoma Public Utilities, 2018).

7.4.8 Parkland Light and Water Company

Parkland Light and Water Company tested its drinking water wells as part of the UCMR3 monitoring. No PFAS were detected at that time. Because of detections in late 2016 at McChord Field just west of Parkland, Parkland began proactively monitoring for PFAS starting in the spring of 2017. Levels of PFOS and PFOA well below EPA's Health Advisory have been detected in two of Parkland's wells located within a mile of the runway at McChord Field. These two wells draw from a very shallow but extremely productive aquifer apparently flowing mostly from the southeast. Parkland continues to monitor these wells.

7.4.9 Naval Base Kitsap-Bangor

In 2019, the Navy identified a number of historical sites where firefighting foam may have been released at Naval Base Kitsap-Bangor. The base drinking water supply was tested in the UCMR3 with no PFAS detected. Based on groundwater flow in the area, the Navy invited adjacent communities in two areas to participate in free voluntary testing for PFAS (January 2020). Results have been reported to the private well owners. Two private wells had PFOS and PFOA combined in excess of the EPA health advisory level and were immediately provided bottled water. PFAS contamination was not detected in two nearby PWS (Silverdale and Kitsap PWS) (Farley, 2020).

7.4.10 Washington state testing site summary

Table 69 reports the results outlined above from various testing sites in Washington. This testing was completed under the UCMR3, through voluntary testing by branches of the Armed Services at military sites, and via voluntary follow-up testing by PWS.

Because water from multiple wells is often blended before distribution, the PFAS results shown may not represent the water concentration delivered to taps. The results shown here also represent the levels in water detected before mitigation.

Table 69. Results of PFAS testing of drinking water in Washington state for PFAS.

Source of testing information	Year	Public drinking water systems, private wells with PFAS detections	PFOA + PFOS combined (μg/L)	Total PFAS measured (μg/L)	PFAS detected	Mitigation action
EPA UCMR3 ^a	2013 to 2015	Issaquah Water System (1 well)	0.490 ^c	0.796	PFOS PFHxS PFHpA PFOA PFNA PFBS	GAC filter installed 2016
EPA UCMR3 ^a	2013 to 2015	City of DuPont Water System (two wells)	0.030	0.030	PFOA	Follow-up monitoring
City of DuPont	2018 to 2019	City of DuPont Water system (four wells)	0.014 - 0.060 ^d	Not reported	PFOA PFOS PFHxS	Blending (2018)
EPA UCMR3 ^a	2013 to 2015	JBLM - Lewis (two wells)	0.051	0.013 – 0.051	PFOA PFHpA	Follow-up monitoring
JBLM ^b	2016 to 2018	Ft. Lewis (five wells)	0.015 – 0.071	up to 0.144	PFOA PFOS PFHxS PFHpA PFBS PFHxA PFNA	One well above health advisory level was not active and has not been returned to service

Source of testing information	Year	Public drinking water systems, private wells with PFAS detections	PFOA + PFOS combined (µg/L)	Total PFAS measured (µg/L)	PFAS detected	Mitigation action
JBLM ^b	2016 to 2018	McChord Field (four wells)	0.216 – 0.250	up to 0.303	PFOA PFOS PFHxS PFHpA PFBS	Wells removed from service until GAC filters installed (2020)
City of Lakewood	2018 to 2019	Lakewood Water District (6 wells)	0.017 – 0.063	Not reported	PFOS PFOA PFHxS	GAC filters installed (2019)
City of Tacoma	2018 to 2019	Tacoma Public Utilities (1 well)	Not reported	Not reported	Not reported	Well removed from service
Parkland Light and Water Company	2017 to 2019	Parkland Light and Water	0.007 – 0.042	Not reported	PFOS PFOA PFHxS	Follow-up monitoring
NAS Whidbey Island ^b	2016 to 2019	Town of Coupeville Evergreen Mobile Home Park 20 private, 4 Group B wells	0.006 – 7.74 ^e	0.004 – 9.9	PFOS PFOA PFHxS PFHxA PFHpA PFNA PFBS	Bottled water or PUR filter provided when tap water is above health advisory level until longterm solution installed
Town of Coupeville	Mar. 2019	Town of Coupeville water system (two wells)	0.022 – 0.061 ^e	0.035 – 0.139	PFOA PFHxS PFHpA	GAC filters installed (July 2019)
Fairchild AFB ^b	2017 to 2019	City of Airway Heights (two wells)	1.4 – 1.5	Not reported	PFOS PFHxS 6:2 FTS PFHxA PFOA	Bottled water provided until Spokane City water was provided or GAC filters installed

Source of testing information	Year	Public drinking water systems, private wells with PFAS detections	PFOA + PFOS combined (µg/L)	Total PFAS measured (μg/L)	PFAS detected	Mitigation action
Fairchild AFB ^b	2017 to 2019	88 private wells	0.073 – 5.7	Not reported	Not reported	Bottled water provided until GAC filters or other long- term solution installed
Fairchild AFB ^b	2017 to 2019	78 private wells	LOD – 0.070	Not reported	Not reported	Continued monitoring

Notes:

- a = EPA's UCMR3 directed 132 public water systems in Washington to test six perfluoroalkyl acids (PFAAs). The systems included all 113 large Group A systems that serve more than 10,000 people and 19 smaller systems. The systems tested cover 94% of Washington residents served by public water systems. PFOS, PFOA, PFNA, PFBS, PFHxS, and PFHpA were measured using EPA Method 537 with reporting limits between 0.02 and 0.04 μg/L.
- b = Military bases with fire training areas or a history of AFFF use have been testing drinking water on and off bases in response to a directive from the DOD (DOD, 2016a, 2016b; DOD Environment, 2018). This is a voluntary effort that is following the EPA lifetime health advisory for PFOS and PFOA combined of 0.070 μg/L. When a private well or public water system exceeds this level, the military has provided alternative water immediately. The military has also installed or paid for filtration of private wells and public water systems.
- c = This well was blended 1:4 with an uncontaminated well before distribution, so the concentration of PFOA and PFOS combined at the nearest businesses was closer to 0.10 μ g/L. Levels were lower still in other parts of the Issaquah water system.
- d = Because of blending, the concentration entering the water distribution system from impacted well fields is lower (0.014 0.028 μg/L for PFOS and PFOA combined).
- e = Results reported show the range of concentrations measured across all wells tested by NAS Whidbey. The maximum detections were in private wells. The maximum detections were in private wells. Town of Coupeville water results are shown above. Because of blending, water PFAS concentrations in distributed tap water were lower.

7.5 Public health advice

7.5.1 EPA health advice for PFOS and PFOA in drinking water

In 2016, EPA established a Lifetime Health Advisory of $0.070 \,\mu\text{g/L}$ for PFOA and PFOS combined in drinking water. A lifetime health advisory level is a concentration in daily drinking water considered to be without risk of adverse health effects over a lifetime of exposure, including in sensitive populations. EPA health advisories are non-regulatory and non-enforceable standards.

The EPA advisory was based on an evaluation of the studies of PFOA and PFOS in laboratory animals and considered studies of human populations exposed to PFAS. For both compounds, EPA identified developmental effects in rodents as the most sensitive endpoint in animal experiments thought to be relevant to humans. For both PFOA and PFOS there are large differences between humans and laboratory animals in how external dose (the amount of intake) translates into internal dose (the amount in blood and organs). Humans retain PFOA and PFOS much longer than laboratory rats and mice, which leads to a higher internal dose in humans given the same external dose. EPA used toxicokinetic modelling to derive equivalent human doses for exposure levels in rodents associated with minimal to no observable adverse developmental effects. Uncertainty factors were applied to derive a RfD of 20 nanograms per kilogram of body weight per day for each compound (EPA, 2016b, 2016c).

EPA assumed that significant PFOA and PFOS exposure occurs through non-drinking water sources and apportioned a default of 20% of the Reference Dose to come from drinking water. They also assumed ninetieth percentile drinking water intake rate for lactating women for their ingestion rate. For an average sized woman, this ingestion rate is 3.8 liters (L) of tap water consumed daily in beverages and foods (see Table 70).

Table 70. 2016 Lifetime Health Advisory Levels for two PFAS in drinking water established by EPA Office of Water (EPA, 2016a).

PFAS	RfD ^a (ng/kg-day)	Basis	Relative source contribution ^b	Drinking water ingestion rate	Lifetime Health Advisory Level in drinking water (µg/L)
PFOA	20	Developmental effects in mice	20%	0.054 L/kg-day ^c	0.070
PFOS	20	Developmental effects in rats	20%	0.054 L/kg-day ^c	0.070

Notes:

- a = RfD is the reference dose which is a health protective value for chronic oral consumption.
- b = Relative Source Contribution is the proportion of the RfD allocated to drinking water sources under the Safe Drinking Water Act.

• c = This ingestion rate is the 90th percentile intake by lactating women from the 2011 EPA Exposure Factors Handbook.

There are no enforceable federal drinking water standards for PFAS. In early 2020, EPA announced its proposed regulatory determination to develop maximum contaminant levels (MCLs) for PFOA and PFOS. The determination is undergoing public comment and has not been finalized. The process of establishing MCLs takes at least three to four years before regulations are adopted. Recent MCLs have taken considerably longer.

7.5.2 Washington Department of Health advice for PFAAs in drinking water

Since 2016, a large number of additional research studies have been published on the toxicity of various PFAS found in drinking water. Comprehensive federal assessments by the ATSDR, the National Toxicology Program, and several state assessments using more recent data have added significantly to the evidence base for health advisories and regulations (see Section 7.2 health concerns). In 2019, Health reviewed this newer data and recommended taking action at lower concentrations of PFOA and PFOS in drinking water (see Table 71). In all, Health recommended state action levels for five PFAS (PFOS, PFOA, PFHxS, PFNA, and PFBS). These draft recommendations support rulemaking being conducted by the SBOH to address PFAS contamination in drinking water. A draft rule and the draft state action levels were posted in November 2019 for public review and an informal comment period. Health also held three stakeholder workshops in December 2019 to engage regulated (Group A) public water systems. A proposed rule is expected 117 in 2020 with rule adoption in 2021.

Table 71. Draft Recommended State Action Levels (SALs) for Washington public drinking water systems.

PFAS	RfD/MRL ^a (ng/kg-day)	Basis	Relative source contribution ^b	Drinking water ingestion rate	SAL in drinking water ^c (μg/L)
PFOA	3	Developmental effects in mice.	50%	MDH model ^d	0.010
PFOS	3	Immune effects in mice. Also protective of developmental effects in rats.	20% Adults 50% Children	MDH model ^d	0.015
PFNA	3	Developmental effects in mice.	50%	MDH model w/ MDHHS inputs ^e	0.014

¹¹⁷ https://www.doh.wa.gov/CommunityandEnvironment/DrinkingWater/RegulationandCompliance/RuleMaking

PFAS	RfD/MRL ^a (ng/kg-day)	Basis	Relative source contribution ^b	Drinking water ingestion rate	SAL in drinking water ^c (μg/L)
PFHxS	9.7	Reduced thyroid hormone (T4) in rats (developmental concern). ^f	50%	MDH model ^d	0.070
PFBS	300	Reduced thyroid hormone (T4) in mice (developmental concern).f	20%	0.047 L/kg-day ^g	1.30

Notes:

- a = RfD is the reference dose which is a health protective value for chronic oral consumption. MRL is a minimal Risk Level which is a health screening level developed by ATSDR.
- b = RSC is the proportion of the RfD allocated to drinking water sources under the Safe Drinking Water Act.
- c = SALs are State Action Levels for Group A public water systems being developed for adoption by Washington State Board of Health.
- d = The MDH Model is the Minnesota Department of Health 2019 peer-reviewed toxicokinetic model for infant intake of bioaccumulative PFAS in drinking water. It includes age-specific drinking water ingestion rates as well as placental and lactational transfer pathways from mother to child.
- e = MDHHS inputs are from the Michigan Department of Health and Human Services 2019 recommended public health screening levels for four PFAS in drinking water.
- f = Thyroxine (T4) is a thyroid hormone.
- g = This ingestion rate is the 90th percentile intake by lactating women from the 2019 EPA Exposure Factors Handbook.

The draft public health advice reflects our best judgement for protecting Washington state residents while we continue to follow advancements in PFAS research. There is a very active research effort to understand the human health impacts of exposure to various PFAS. Health researchers continue to study health outcomes in human populations with elevated exposures. EPA is using rapid toxicity-screening tools to investigate potential biological activities of 75 compounds that are representative of the various classes of PFAS chemistry. Industry and independent scientists are publishing new findings regularly in peer-reviewed scientific literature.

7.5.3 Drinking water health advisories and MCLs set by other states

A number of states use the EPA Health Advisories for PFOS and PFOA (AS, AL, AZ, CO, KS, ME, NW, WV) to guide public health advice for drinking water. At least eight states have established independent state advisories for PFOS, PFOA, and/or other PFAAs in drinking water (CT, VT,

MA, MN, MI, CA, NC, OH). Some states issued separate advisories for specific PFAS and some—like VT, CT, and MA—have set a value for the total of five to six PFAS in drinking water (Interstate Technology Regulatory Council (ITRC), 2020c). The ITRC maintains a <u>current listing of state and international standards and guidance values</u>¹¹⁸ for PFAAs in groundwater, drinking water, and surface water/effluent wastewater. We refer readers to this resource for the most current information.

As of April 2020, only New Jersey has adopted an enforceable drinking water limit, called an MCL for a PFAS compound (PFNA). MCLs for PFOA and PFOS are in the process of adoption by New Jersey. New Hampshire adopted state MCLs in October 2019 for four PFAS, but the effective date has been stayed by a lawsuit from a PFAS manufacturer. Michigan, New Jersey, New York, and Vermont have proposed state MCLs or are in the process. California established enforceable Response Levels for public water sources with PFOS or PFOA contamination. Other states may take enforcement action on drinking water through their groundwater protection laws.

7.5.4 Washington state assessment and advice for PFAS contaminants in recreational fish

Recreational, subsistence fishers, and low-income or tribal communities that consume fish from urban waters and areas downstream of wastewater treatment plant discharges may have higher exposures to PFAS that accumulate in fish. Serum of fish and shellfish consumers who participated in NHANES in 2007 – 2014 had higher levels of several PFAS (Christensen et al., 2017). Researchers determined that consumers of fish and shellfish are at higher risk of exposure to certain PFAS than non-consumers. In Washington, PFOS was detected by Ecology surveys in Washington freshwater fish at levels up to 87 ng/g in fillets (see Appendix 5: Environmental Ocurrence, Section 5.1.7 Freshwater fish).

International studies indicate that some PFAAs—such as PFOS, PFHxS, and PFOA—can reach very high levels in serum of fishermen who eat fish from industrially impacted areas (G. W. Olsen, 2015a). A recent study also identified a number of novel PFAS in fish from the Yangtze River and Tandxun Lake, China (including six sulfonate classes, two amine classes, one carboxylate class, and one *N*-heterocycle class) (Liu et al., 2018). The discovery of these PFAS in fish demonstrates bioavailability and the potential for bioaccumulation for these compounds or their precursors, in which toxicity and environmental fate has not been studied. A study examined the association of PFAS with diet and drinking water in reproductive-aged Chinese women. Intake of freshwater fish, marine fish, shrimp, and crab was positively associated with plasma PFAS concentrations, whereas higher intake of soy products was associated with lower plasma PFAS levels (Zhou et al., 2019).

¹¹⁸ https://pfas-1.itrcweb.org/fact-sheets/

Several states with localized surface water contamination have developed fish advisories for PFAAs, including Alabama, Michigan, Minnesota, New Jersey, Oregon, and Wisconsin. Other states are considering fish advisories. In Minnesota, fish tissue with more than 800 ng/g PFOS in edible parts are listed as *do not eat*, fish with 40 – 800 ng/g have various recommended consumption restrictions, and fish with less than 40 ng/g have no suggested consumption limits. New Jersey issued a consumption advisory for 12 species of fish that were found to contain chemicals belonging to the PFAS family (Hurdle, 2018). Michigan has developed Eat Safe Fish Guidelines for PFOS across numerous water bodies (Michigan Government, 2018). These guidelines are set to be protective for everyone including children, pregnant and breastfeeding women, and people with existing health problems such as cancer or diabetes.

The Great Lakes Consortium for Fish Consumption Advisories released <u>Best Practice for Perfluorooctane Sulfonate (PFOS) Guidelines</u>¹¹⁹ in November 2019. The guidance is voluntary for states that border the Great Lakes, but is intended to promote consistency in fish consumption advice. The best practices meal advice categories for all populations are:

- PFOS in fish (microgram per kilogram [µg/kg]): ≤ 10, Meal frequency: Unrestricted.
- PFOS in fish ($\mu g/kg$): > 10 20, Meal frequency: 2 meals per week.
- PFOS in fish ($\mu g/kg$): > 20 50, Meal frequency: 1 meal per week.
- PFOS in fish (µg/kg): > 50 200, Meal frequency: 1 meal per month.
- PFOS in fish (μg/kg): > 200, Meal frequency: Do not eat.

There are currently no fish consumption advisories for PFAS in Washington. In 2018, Health determined provisional health-based screening levels for PFOS and PFOA based on the 2016 EPA RfDs. The provisional screening levels were 23 μ g/g and 8 μ g/g for the general population and high consumers, respectively. Health reviewed fish data collected by Ecology in 2008 and 2016 and found that some fillet tissue contained PFOS concentrations in excess of these values. Specifically, PFOS was detected in Washington freshwater fish at levels up to 87 ng/g in fillets (see <u>Appendix 5: Environmental Ocurrence, Section 5.1.7 Freshwater fish</u>). Health requested additional data to provide an adequate basis for a fish consumption advisory. That data was collected by Ecology and provided to Health in 2019. Health is reviewing these data and reevaluating its screening levels in consideration of the lower recommended oral intake recently recommended as part of Health's draft drinking water advice.

7.5.6 International health guidance values

German human biomonitoring commission (HBM Commission)

In 2017, the German Human Biomonitoring Commission (HBM commission) derived health-related guidance values in blood plasma for PFOA and PFOS. The "HBM I" value represents the concentration of a substance in human biological material at which, and below, there is no risk of adverse health effects, and no need for action. Based on an assessment of literature on

¹¹⁹ https://www.health.state.mn.us/communities/environment/fish/docs/consortium/bestpracticepfos.pdf

human epidemiological studies, and on animal studies, the HBM commission derived an HBM I value of 2 nanograms per milliliter (ng/mL) for PFOA and 5 ng/mL for PFOS (Apel et al., 2017).

In deriving the HBM I, the HBM commission included fertility and pregnancy, weight of newborns at birth, lipid metabolism, immunity after vaccination, hormonal development, thyroid metabolism, and onset of menopause as relevant and significantly associated with an exposure to PFOA and/or PFOS. The most recent testing from NHANES 2015 – 2016 shows that more than one quarter of the U.S. population have levels over 2.5 ng/mL for PFOA and PFOS at levels over 8.1 ng/mL (CDC, 2019).

French Agency for Food, Environmental, and Occupational Health and Safety

In 2017, the French Agency for Food, Environmental, and Occupational Health and Safety (ANSES) developed human reference doses (toxicity reference values (TRVs)) for PFBA, PFHxS, PFBS, and PFHxA based on studies conducted in laboratory animals (Table 72) (ANSES, 2017). TRVs are established for a given critical effect, and are specific to a substance, a duration of exposure (acute, subchronic or chronic), and a route of exposure (oral, inhalation, etc.).

Table 72. Toxicity reference values developed by ANSES (ANSES, 2017).

Chemical	Critical effect and study	Critical concentration	Uncertainty factor (UF)	Toxicity reference value (TRV) (mg/kg-day)
PFBA	Hepatic effects Butenhoff et al., 2012	NOAEL = 6 mg/kg-d HED = 1.764 mg/kg-d	75	0.024
PFHxS	Hepatic effects Butenhoff et al., 2012	NOAEL = 1 mg/kg-d HED = 0.289 mg/kg-d	75	0.004
PFBS	Renal effects (Hyperplasia tubular) Lieder et al., 2009b	BMD 10% = 24 mg/kg-d HED = 6.06 mg/kg-d	75	0.08
PFHxA	Renal effects (papillary necrosis & tubular degeneration) Klaunig et al., 2015	NOAEL = 30 mg/kg-d HED = 7.91 mg/kg-d	25	0.32

Notes:

- LOAEL: Lowest-observed-adverse-effect level.
- NOAEL: No-observed-adverse-effect level.
- BMD: Benchmark dose.
- HED: Human equivalent dose.

European Food Safety Authority

In 2018, the EFSA issued a provisional scientific opinion on tolerable weekly intakes of PFOA and PFOS (EFSA, 2018). EFSA used a different approach and did not derive their estimates from adverse health outcomes in controlled animal studies. Rather, they used serum measurements in human observational studies to model serum levels associated with 5% changes in adverse outcomes.

After an extensive review of epidemiological evidence, they selected the outcomes with the strongest evidence for a causal association with PFOS and PFOA. These were increased serum cholesterol, decreased antibody response to vaccines, and lower birthweight for PFOS; and increased serum cholesterol, elevated liver enzyme (ALT), and decreased birth weight for PFOA. They then used physiologically based pharmacokinetic (PBPK) modelling to estimate the dietary intake that would produce that serum level over a lifetime of continuous exposure. For children, they used maternal serum levels and models of maternal transfer during gestation and breastfeeding to target children's serum levels at five years old (EFSA, 2018).

- PFOS: Serum levels associated with a 5% change in total cholesterol or birthweight ranged from $21-25~\mu g/L$. The serum level for vaccine response was lower, 10.5 $\mu g/L$. EFSA recommended a tolerable daily intake of 1.8 2.0 ng/kg bw-day based on the cholesterol endpoint.
- PFOA: Serum levels associated with a 5% change in total cholesterol ranged from 9.2 9.4 μ g/L, for increase in liver enzyme was 21 μ g/L, and for birth weight ranged from 4.4 10.6 μ g/L. EFSA recommended a tolerable daily intake of 0.4 2.0 μ g/kg bw-day.

In February 2020, EFSA proposed a tolerable weekly intake (TWI) of 8 ng/kg bw-day for PFOS, PFOS, PFNA, and PFHxS combined (EFSA, 2020). This reflects an updated evaluation of recent scientific findings and use of new Agency guidance for assessing combined exposure to multiple chemicals (EFSA, 2019). The TWI is based on effects in the immune system seen in animals (PFOA, PFOS) and associations between the sum of PFOS, PFOA, PFHxS, and PFNA in serum, reduced titers of antibodies to diptheria and tetnus in 5-year-old children in the Faroe Islands, and reduced antibody titers against haemophilus influenza type b (Hib) in 1-year-old children in a study from Germany. The proposed EFSA opinion is undergoing public review and comment.

7.6 Data gaps and recommendations

7.6.1 Data gaps

Further information on the following would reduce uncertainty about toxicity and health effects:

 Testing PFNA and PFHxS for immune system toxicity including antibody response to antigens.

- Investigation into developmental impacts of PFOA in mouse mammary glands to include testing of other PFAS, other animal models, and prospective human studies in exposed communities.
- Mechanistic investigations into underlying activity of PFAS compounds in mammals.
- Further toxicity testing on other PFAS that occur in drinking water and human serum (especially PFHpA, PFHxA, PFBA, 6:2 FTS, and PFDA).
- Further investigation into potential interactions within mixtures of PFAS (additive, antagonistic, synergistic effects).
- Prospective and longitudinal studies in human populations.

Further information on the following would improve exposure assessment:

- Better characterization of human exposure to PFAS in environmental media such as drinking water and in human serum. Both expanded panels of targeted analysis and nontargeted analysis methods would be helpful.
- Wider and more comprehensive PFAS testing in U.S. foods and pathway investigations into plant and animal uptake of various PFAS from soil, feed, and water.
 - For example, PFOS was frequently detected in freshwater fish in the state, but we know little about PFAS in marine fish and shellfish. Biomonitoring surveys have indicated that self-reported consumption of fish and shellfish are associated with serum levels of certain PFAS in the general populations, making this an important data gap to fill.
- Additional studies on the relative contribution of sources other than food are needed, especially for PFAS, which are present in the highest concentrations in indoor air and house dust. Information needed includes:
 - The mechanisms for PFAS transfer from products to indoor air, dust and surfaces.
 - The relationship between household products containing PFAS and inhalation exposure.
 - o The levels of dermal exposure from household products containing PFAS.
 - Mitigation measures that can reduce PFAS levels in homes, house dust, and indoor air, including the availability of safer alternatives.
- Studies on half-life of PFAS in humans by gender and lifestage.
- Prospective and longitudinal studies in human populations.

7.6.2 Recommendations

Recommendation 1.0: Ensure drinking water is safe.

Protecting public health by ensuring safe drinking water is a fundamental responsibility of the Health Office of Drinking Water (ODW).

There are three types of drinking water systems in Washington:

- Group A water systems serve <u>85% of state residents</u>.¹²⁰ They service more than 15 connections or more than 25 people. ODW primarily regulates these public water systems.
- Group B public water systems are smaller and serve 1.5% of state residents. The local health department usually oversees these systems. Group B systems have few testing requirements for continued operation.
- Private wells serve 13.5% of state residents. Private wells are only regulated in their design and installation; chemical testing is not usually required.

Less than 1% of all Group A systems have been tested for PFAS. However, those that have been tested serve most water customers in the state. The percentage of Group B and private wells tested for PFAS is even lower. A water test is required to determine whether PFAS are in drinking water, because PFAS are tasteless and odorless at levels of public health concern.

Because testing and treating for PFAS in drinking water is expensive, exposures to PFAS-contaminated water may be disproportionately borne by populations who do not have the financial means to test for and remove these contaminants.

The following additional actions are recommended to ensure drinking water is safe.

Recommendation 1.1: Identify funding for PFAS drinking water mitigation.

State agencies, the Washington State Legislature, and local water systems should work together to fund PFAS drinking water mitigation. These costs should be reimbursed by responsible parties under applicable laws. Once PFAS water contaminants are officially classified as hazardous substances by the federal government or by the state of Washington, they can be addressed under the state Model Toxics Control Act (MTCA) framework.

Existing resources

Drinking Water State Revolving Fund is an EPA-funded loan program administered by Health. The loans are used to:

- Improve drinking water infrastructure.
- Finance the cost of installing treatment or other infrastructure improvements over a number of years.

Drinking Water State Revolving Fund can provide emergency loans in the event a water system is issued a "Do Not Use" order as a result of PFAS contamination. The program recently funded a reservoir project for the City of Spokane to allow Spokane to provide reliable water service to Airway Heights. Airway Heights has PFAS in their wells and is now relying on the City of Spokane for its water.

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U.S. Environmental Protection Agency (EPA) provides funding to Health Office of Drinking Water for set-aside activities and source water protections. We can use these funds in limited circumstances to defray costs of additional water testing.

Other funding programs in the state could be tapped for loans or grants to help with costs of new infrastructure in response to PFAS contamination:

- Public Works Assistance Account overseen by Public Works Board.
- Community Development Block Grant overseen by Department of Commerce.
- Rural Development loans and grants overseen by the U.S. Department of Agriculture.

Public water systems can pursue reimbursement from potentially liable parties under the state MTCA when PFAS are declared hazardous substances under MTCA. Even under MTCA, water systems may have to carry costs long-term or permanently because:

- The process of identifying responsible parties and being reimbursed can take years.
- Responsible parties may be difficult, if not impossible, to determine.
- The potentially liable party could be a local entity under the same public administration as the water utility (for example, a local fire station).

In each of these cases, the costs borne by the water system would be long-term or permanent.

Recommendation 1.2: Provide technical support for site characterization, source investigation and mitigation at contaminated sites.

Ecology and Health will continue to develop expertise and provide technical assistance and guidance to drinking water purveyors, local jurisdictions, and responsible parties to address PFAS contamination and conduct cleanup actions.

Those actions include:

- Ecology will continue to collaborate with involved parties at PFAS contamination sites in the state. These efforts will help to better understand the sources, composition, and distribution of PFAS contamination in soil and water. Identification and evaluation of appropriate cleanup actions and their costs will be informed by this work. This work is being done within Ecology's existing resources.
- Health will continue to provide water systems with advice and assistance to
 understand the mitigation options and guide voluntary action on unregulated PFAS
 until the rulemaking for PFAS in drinking water is complete. To-date, technical
 assistance has focused on public water systems near military bases with PFAS
 detections in groundwater. Department of Health continues to include local health
 departments in outreach and guidance. This work is being done within Health's
 existing resources.
- Ecology will look at targeting Safe Drinking Water Action Grants (a category of Remedial Action Grants for Local Governments) to help address PFAS-contaminated drinking water, once some of the uncertainties discussed above have been addressed.

- Ecology will investigate PFAS contamination in groundwater and surface water.
 These efforts would support local health departments, cities, counties, and other public entities in Washington when PFAS contamination is discovered. Initial investigation efforts could identify areas at high risk of contamination. This could include areas where trainings or firefighting activities used large quantities of PFAS-containing AFFF, or where spills released the foam. Ecology could prioritize funding for site-specific assessments and groundwater testing. Funding for this action is estimated below.
- Ecology will consider the number of people impacted, the concentration of the PFAAs in the drinking water, and vulnerable populations present when prioritizing mitigation and cleanup activities. Ecology will use mapping tools such as Environmental Justice (EJ) screen and the Information by Location (IBL) tool in the Washington Tracking Network (WTN) portal to characterize the demographics of the population served by impacted drinking water.
- Obtain chemical identities from products and at contaminated sites to find chemical "fingerprints" useful in identifying source locations. Analytical methods may not yet be developed to obtain all the required data.

Recommendation 1.3: Support biomonitoring and other studies to help answer important human exposure and health questions.

Health should continue to find opportunities for Washington residents to participate in exposure and health studies. These studies help answer important community and public health questions about PFAS exposure and health outcomes. For example, Health requested and supports inclusion of Airway Heights as one of eight sites in the ATSDR's PFAS Exposure Assessment study. We also applied for but were not awarded a cooperative agreement to include a Washington site in the ATSDR Multisite PFAS Health Study.

State agencies should also support investigations into sources and pathways of PFAS contamination in food, drinking water, and the indoor environment and pursue policies to mitigate and reduce these sources of human exposure over time.

Supplement 1: Summary of Primary Health Concerns by PFAA

Table 73. Health-based values for oral intake of different types PFAA chemicals according to various government agencies and authoritative bodies.

Type of PFAA chemical	Authoritative body responsible for value (year)	Health-based value for subchronic/chronic oral intake (ng/kg-day)
PFOA	EPA RfD (2016)	20
PFOA	ATSDR MRL (2018)	2
PFOA	NJ DWQI RfD (2017)	2
PFOA	NH DES RfD (2019)	6.1
PFOA	MI SAW TV (2019)	3.9
PFOA	CA OEHHA ADD (2019)	0.45
PFOA	EFSA TDI (2018)	0.8
PFOA	Health Canada TDI (2018)	21
PFOS	EPA RfD (2016)	20
PFOS	ATSDR MRL (2018)	2
PFOS	NJ DWQI RfD (2018)	1.8
PFOS	NH DES (2019)	3
PFOS	MDH RfD (2019)	3.1
PFOS	MI SAW TV (2019)	2.89
PFOS	CA OEHHA ADD (2019)	1.8
PFOS	EFSA TDI (2018)	1.8
PFOS	Health Canada TDI (2018)	60
PFNA	ATDSR MRL (2018)	3
PFNA	NJ DWQI RfD (2015)	0.74
PFNA	NH DES (2019)	4.3
PFNA	MI SAW TV (2019)	2.2
PFHxS	ATDSR MRL (2018)	20
PFHxS	MDH RfD (2019)	9.7
PFHxS	NH DES RfD (2019)	4
PFHxS	MI SAW TV (2019)	9.7
PFBS	EPA RfD (2018)	14,000
PFBS	MDH RfD (2017)	430
PFBS	MI SAW TV (2019)	300

Notes:

 EPA RfD (2016, 2018). The Environmental Protection Agency Office of Water developed RfDs for PFOA and PFOS in 2016 to support drinking water health advisories. The Agency IRIS program also released a draft RfD for PFBS in December 2019. An RfD is an EPA estimate, with uncertainty or safety factors built in, of the daily lifetime dose of a substance that is unlikely to cause harm in humans. RfDs are based on non-cancer endpoints (EPA, 2016a, 2018b).

- ATSDR MRL (2018). The Agency for Toxic Substances and Disease Registry released a
 draft Toxicological Profile for PFAS in 2018 that included Minimal Risk Levels (MRLs) for
 four PFAAs. An MRL is an ATSDR estimate of daily human exposure to a hazardous
 substance at or below which that substance is unlikely to pose a measurable risk of
 harmful noncancerous effects. PFAS MRLs are screening values for intermediate
 duration oral exposures of more than 14 days and less than a year (Agency for Toxic
 Substances and Disease Registry (ATSDR), 2018a).
- NJ DWQI RfD (2015 18). The New Jersey Drinking Water Quality Institute develops technical recommendations and supporting documentation for drinking water standards in the state of New Jersey. They developed target serum levels and RfDs for PFNA, PFOS, and PFOS (New Jersey, 2017; New Jersey Drinking Water Quality Institute, 2015; NJDWQI, 2018).
- MI SAW TV (2019). Michigan Science Advisory Workgroup (SAW) developed toxicity values (TVs) corresponding to serum concentrations expected to have minimal or no risk for humans with daily chronic exposure. The toxicity values were developed to support drinking water regulation in the state (Michigan Science Advisory Workgroup, 2019).
- CA OEHHA ADD (2019). The State of California, Office of Environmental Health Hazard
 Assessment developed Acceptable Daily Doses (ADD) for PFOA and PFOS in support of
 drinking water notification and removal levels. ADDs are developed for noncancer
 endpoints and are estimates of the maximum daily dose of a chemical that can be
 consumed by humans for an entire lifetime without toxic effects (CalEPA OEHHA, 2019).
- EFSA TDI (2018). The European Food Safety Authority published Tolerable Daily Intakes (TDI) for PFOA and PFOS in December 2018 (European Food Safety Authority (EFSA), 2018).
- Health Canada TDI (2018). Health Canada developed Tolerable Daily Intakes for PFOS and PFOA (Health Canada, 2018a, 2018b).
- MDH RfD (2017-19). Minnesota Department of Health developed RfDs for PFOS, PFHxS, PFBA and PFBS in support of state drinking water guidance. This is part of their program to develop health-based values for emerging contaminants (MDH, 2017; Minnesota Department of Health (MDH), 2017, 2019).
- NH DES RfD (2019). New Hampshire Department of Environmental Services developed target serum values and RfDs for PFOS, PFOA, PFNA, and PFHxS to support state drinking water standards (New Hampshire Department of Environmental Services, 2019).
- IARC (2017). International Agency for Research on Cancer (IARC), an agency of the World Health Organization, published their monograph on PFOA in 2017 classifying it as Group 2B possibly carcinogenic to humans (IARC, 2017).

PFOA

The primary effects observed in laboratory animals following PFOA exposure are:

 Liver toxicity (L. Biegel, Hurtt, Frame, O'Conner, & Cook, 2001; J. Butenhoff, G. Kennedy, S. Frame, J. O'Connor, & R. York, 2004; Loveless, Hoban, Sykes, Frame, & Everds, 2008; Perkins, Butenhoff, Kennedy, & Palazzolo, 2004).

- Immunotoxicity (J. DeWitt, Copeland, Strynar, & Luebke, 2008; Q Yang, Xie, & Depierre, 2000; Q. Yang, Xie, Eriksson, Nelson, & DePierre, 2001).
- Reproductive and developmental toxicity (B. D. Abbott, 2015; J. Butenhoff et al., 2004; Lau et al., 2006; Macon et al., 2011; White et al., 2011; C. Wolf et al., 2007).
- Altered thyroid hormones (National Toxicology Program (NTP), 2019b).

The strongest and most consistent associations between PFOA exposure and adverse health effects in humans are:

- Elevated serum cholesterol (Lau, 2015; New Jersey Drinking Water Quality Institute (NJDWQI) Health Effects Subcommittee, 2016).
- Reduced birth weight (Johnson et al., 2014; Meng et al., 2018).
- Reduced antibody response to vaccines (National Toxiciology Program, 2016).
- Increased serum liver enzymes (Gallo et al., 2012; C. Y. Lin et al., 2010; G. Olsen et al., 2007; Sakr et al., 2007; Yamaguchi et al., 2013).

Studies also report associations between PFOA exposure and:

- Altered development of reproductive tissue and delayed puberty (Di Nisio A, 2018; Lopez-Espinosa et al., 2011).
- Higher serum uric acid (Geiger, Xiao, & Shankar, 2013; Shankar, Xiao, & Ducatman, 2011b; Steenland, Tinker, Shankar, & Ducatman, 2010)
- Altered thyroid hormone levels and thyroid disorders (Jain, 2013; Knox, Jackson, Frisbee, Javins, & Ducatman, 2011; G. W. Olsen, Burris, Burlew, & Mandel, 2003; Wen et al., 2013).
- Pregnancy-induced hypertension and preeclampsia (Savitz, Stein, Bartell, et al., 2012; Savitz, Stein, Elston, et al., 2012; C. Stein, Savitz, & Dougan, 2009).
- Ulcerative colitis (Steenland et al., 2015; Steenland et al., 2013).

PFOA is not considered genotoxic or mutagenic, but studies in laboratory animals have reported increased incidence of tumors in liver, testicular, and pancreatic tissues as well as ovarian tubular hyperplasia (L. B. Biegel et al., 2001; J. L. Butenhoff, Kennedy, et al., 2012; Environmental Protection Agency (EPA), 2016a). PFOA exposure was positively associated with increased incidence of kidney and testicular cancers in a large epidemiological study (the C8 Health Project) (Barry et al., 2013; Vieira et al., 2013). Studies of the general population have looked for but not found associations between serum PFOA levels and a range of human cancers (Bonefeld-Jorgensen et al., 2014; Eriksen et al., 2009; Hardell et al., 2014; Innes et al., 2014).

The following cancer classifications have been applied to PFOA:

- "Suggestive evidence" of carcinogenic potential in humans (EPA, 2016).
- Group 2B, possibly carcinogenic to humans (IARC, 2017).

PFOS

The primary types of toxicity observed in experimental animals exposed to PFOS are:

- Developmental toxicity (Lau et al., 2003; Luebker, Case, et al., 2005; Luebker, York, Hansen, Moore, & Butenhoff, 2005).
- Immune suppression (G. Dong et al., 2009; G. H. Dong et al., 2011; Guruge et al., 2009; Peden-Adams et al., 2008; Zheng, Dong, Jin, & He, 2009).
- Liver and kidney toxicity (Cui, Zhou, Liao, Fu, & Jiang, 2009; A. M. Seacat et al., 2003; Xing et al., 2016).
- Disruption of thyroid and other hormones (S. C. Chang et al., 2008; L. Li et al., 2018; Wan et al., 2011; Yu et al., 2009; Zhao et al., 2014).

Limited studies in rats and mice also report:

- Degenerative changes in male reproductive tissue (Lopez-Doval et al., 2014; Wan et al., 2011; Zhao et al., 2014).
- Signs of altered neurodevelopment (changes in motor activity and habituation response) (Johansson, Fredriksson, & Eriksson, 2008; Y. Wang, Liu, Zhang, Zhao, & Quan, 2015)
- Altered glucose metabolism following prenatal exposures. (Lv et al., 2013; Wan, Zhao, Leung, & Wong, 2014).

The most consistent findings from human epidemiological studies are positive associations between serum PFOS and:

- Higher serum cholesterol (Frisbee et al., 2010; Nelson et al., 2010; G. W. Olsen et al., 2003; Steenland, Tinker, Frisbee, Ducatman, & Vaccarino, 2009).
- Reduced antibody response to vaccines (J. C. DeWitt, Blossom, & Schaider, 2019; National Toxicology Program (NTP), 2016).
- Reduced birth weight (Koustas et al., 2014).

Other endpoints of concern with less evidence include:

- Elevated uric acid (Geiger et al., 2013; Steenland et al., 2010).
- Altered energy metabolism and glucose intolerance (Domazet, Grontved, Timmermann, Nielsen, & Jensen, 2016; Lin, Chen, Lin, & Lin, 2009; Liu et al., 2018).
- Altered hormone levels (Ballesteros et al., 2017; Kim et al., 2018; Webster et al., 2016).
- Thyroid disease (Melzer et al., 2010; Webster, Venners, Mattman, & Martin, 2014; Wen et al., 2013).
- Chronic kidney disease (Shankar, Xiao, & Ducatman, 2011a; Steenland et al., 2010).

PFOS does not appear to be mutagenic or genotoxic but chronic rodent studies observed liver, thyroid and mammary gland tumors (J. L. Butenhoff, Chang, et al., 2012). Data relevant to cancer risk of PFOS are limited. In terms of cancer classifications, for PFOS, there is "suggestive evidence" of carcinogenic potential in humans (EPA, 2016).

PFNA

The general types of rodent toxicity observed with PFNA are similar to PFOA:

- Liver toxicity (Das et al., 2015; Mertens et al., 2010; National Toxicology Program (NTP), 2019b; Stump et al., 2008; J. Wang, Yan, Zhang, Zhang, & Dai, 2015).
- Male reproductive toxicity (Y. Feng et al., 2009; National Toxicology Program (NTP), 2019b; Singh & Singh, 2019a, 2019b).
- Female reproductive toxicity (Stump et al., 2008; C. J. Wolf et al., 2010).
- Developmental toxicity (Das et al., 2015; Rogers et al., 2014; Rosen, 2010; C. J. Wolf et al., 2010).
- Immunotoxicity (X. Fang et al., 2009; X. Fang, Feng, Wang, & Dai, 2010; X Fang et al., 2008; National Toxicology Program (NTP), 2019b).
- Thyroid hormone alterations (National Toxicology Program (NTP), 2019b).

Epidemiological studies relevant to PFNA provide limited evidence of an association between PFNA exposure and increased serum cholesterol, but not with other lipid alterations (HDL, LDL, triglycerides) (Agency for Toxic Substances and Disease Registry (ATSDR), 2018b; NJDWQI, 2015) and small associations between serum PFNA and some liver function biomarkers (Jain & Ducatman, 2019; Nian et al., 2019; Salihovic et al., 2018).

Investigations of PFNA and immune endpoints in humans reported associations between higher PFNA exposure and:

- Decreased antibody response to a vaccine (Granum et al., 2013; Kielsen et al., 2016).
- Higher number of reported respiratory infections or common cold in children (Granum et al., 2013; Impinen et al., 2018).
- Asthma in children (G. H. Dong et al., 2013). Asthma and allergic diseases were not associated with PFNA in a number of other studies (Granum et al., 2013; Humblet et al., 2014; Impinen et al., 2019; Impinen et al., 2018).

Other associations between PFNA exposure measures and reproductive and developmental outcomes have been reported in epidemiological studies (Meng et al., 2018; Sagiv et al., 2018) including:

- Gestational diabetes (Rahman et al., 2019).
- Increased risk of miscarriage (Jensen et al., 2015).
- Lower birth weights (Sagiv et al., 2018).
- Altered timing of puberty onset for boys and girls (Ernst et al., 2019).
- Altered bone mineral density in girls at 17 years old (Jeddy et al., 2018).
- Lower levels of sex hormones and insulin-like growth factor (IGF-1) in boys and girls six to nine years old (Lopez-Espinosa, Mondal, Armstrong, Eskenazi, & Fletcher, 2016).

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PFHxS

The liver is the primary target of PFHxS toxicity in rodent studies. Effects observed include increased liver weight, hepatocellular hypertrophy, altered lipid metabolism, steatosis, and necrosis (Bijland et al., 2011; J. L. Butenhoff, Chang, et al., 2009; S. Chang et al., 2018). Several studies have reported thyroid cell damage and reduced T4 and T3 thyroid hormone levels in rodent studies (J. L. Butenhoff, Chang, et al., 2009; NTP, 2018; Ramhoj et al., 2018).

Reproductive and developmental effects have been reported in some studies such as reduced litter size (S. Chang et al., 2018) and reduced birth weight (Ramhoj et al., 2018), but have not been consistently observed. One study reported altered spontaneous behavior and habituation in adult mice that had received a single dose of PFHxS on postnatal day ten (Viberg, Lee, & Eriksson, 2013). A key data gap is the lack of immune toxicity testing in animal studies.

According to ATSDR's 2018 draft assessment, the weight-of-evidence for epidemiological studies supports associations between PFHxS exposure and liver damage (as evidenced by increases in serum enzymes and decreases in serum bilirubin levels) and decreased antibody response to vaccines (Agency for Toxic Substances and Disease Registry (ATSDR), 2018b). There is also limited and somewhat inconsistent evidence of associations between higher PFHxS exposure and increased risk of hyperactivity in children (K Hoffman, Webster, Weisskopf, Weinberg, & Vieira, 2010; C. R. Stein & Savitz, 2011) and reduced T4 levels in pregnant women and male infants (Ballesteros et al., 2017; Preston et al., 2018).

The carcinogenicity of PFHxS has not been investigated.

Perfluoroheptanoic acid (PFHpA)

There is very limited data in laboratory animals to assess PFHpA. *In vitro* studies showed that PFHpA is as biologically active as PFOA in activating PPARα, however this activity was not evident *in vivo*, probably because PFHpA was rapidly excreted and did not accumulate in the rodent liver (Goecke-Flora & Reo, 1996; HSDB, 2016; Kudo, Bandai, Suzuki, Katakura, & Kawashima, 2000; Kudo et al., 2001; Ohmori, Kudo, Katayama, & Kawashima, 2003). People do not excrete PFHpA as rapidly as rodents. In a study of 11 professional ski waxers, it took between 31 and 123 days after exposure ceased for their individual serum level of PFHpA to drop by half. A study of Chinese adults reported a longer estimated half-life in human serum (1.5 years) (Russell, Himmelstein, & Buck, 2015; Zhang, Beesoon, Zhu, & Martin, 2013).

Studies in humans are lacking. Fu et al. (2014) reported no association between PFHpA in serum of adults and increased total cholesterol and LDL cholesterol at environmental exposure levels (Fu et al., 2014). Epidemiological studies investigating immune toxicity did not find associations between serum PFHpA levels and diphtheria or tetanus antibody levels in adults (Kudo et al., 2001), or risk of asthma diagnosis, eczema, or wheezing in children (Smit et al., 2015). Mattsson et al. (2015) reported that the risk of coronary artery disease was higher in individuals with serum PFHpA levels in the 3rd quartile of exposure, but not the 4th (highest) exposure quartile.

PFHxA

Liver effects observed in 90-day rat studies with PFHxA were generally mild and required higher doses than PFOA (Chengelis, Kirkpatrick, Radovsky, & Shinohara, 2009; DuPont, 2007; Loveless et al., 2009). As discussed in Hall et al. (2012), these liver effects may not be relevant to humans (Hall et al., 2012). Effects on kidney and tubular degeneration observed in rodents (Klaunig et al., 2015) served as the basis for Michigan's toxicity value, and are supported by a comprehensive review of available toxicology data (Luz, Anderson, Goodrum, & Durda, 2019). The health-based value for subchronic or chronic intake of PFBA from Michigan Science Advisory Workgroup (SAW) is 83,000 ng/kg-day (SAW, 2019).

Thyroid effects were observed only at very high doses in rats (DuPont, 2007; Loveless et al., 2009). In reproductive and developmental toxicity studies, PFOA-like effects were seen but at much higher doses (National Industrial Chemicals Notification and Assessment Scheme (NICNAS), 2015, 2017). A cancer study in rodents of PFHxA was negative for tumors (Klaunig et al., 2015).

Limited human evidence has been reported (Luz et al., 2019). Four cross-sectional human epidemiology studies provide some evidence of statistical associations between serum PFHxA levels and testosterone (Zhou et al., 2016), thyroid antibody markers (Li, Cheng, Xie, & Zeng, 2017), and Gilbert's syndrome (Fan, Ducatman, & Zhang, 2014).

PFBS

In rodent testing, PFBS reduced thyroid hormones and produced kidney toxicity such as hyperplasia, developmental toxicity including delayed growth and maturation, hypertrophy in liver tissue, increased serum liver enzymes, and altered lipids and hematological profiles (X. Feng et al., 2017; Lieder, York, Hakes, Chang, & Butenhoff, 2009; York, 2002, 2003; Lieder, Chang, et al., 2009; Bijland et al., 2011; 3M Company, 2001; National Toxicology Program (NTP), 2019a).

Very little information is available on potential immune toxicity or carcinogenicity of PFBS (EPA, 2018a). Using a structured literature review, EPA identified only seven epidemiologic studies that report on the association between PFBS and human health effects. Statistically significant positive associations between PFBS and asthma, serum cholesterol, and high-density lipoprotein levels were reported in at least one study (EPA, 2018a).

PFBA

Toxicity observed in laboratory mice and rats:

- Mild liver toxicity (J. L. Butenhoff, Bjork, et al., 2012; Crebelli et al., 2019; Foreman et al., 2009).
- Mild thyroid toxicity (increased thyroid weight, hyperplasia, and hypertrophy of the follicular epithelium).

 Decreased thyroid hormone (T4) levels and decreased serum cholesterol (Bjork & Wallace, 2009; J. L. Butenhoff, Bjork, et al., 2012).

Males were more sensitive than females. In a reproductive and developmental study in mice, litter loss was observed at high doses. Despite liver enlargement in dams and pups, no effects on maternal, fetal, or pup weight gain were noted. Mouse offspring exposed to PFBA prenatally had significant delays in eye opening and a slight delay in onset of puberty (Das et al., 2008). PFBA was less developmentally toxic than PFOA presumably because of its more rapid elimination from the mouse. The health-based value for subchronic or chronic intake of PFBA from Minnesota Department of Health is 2,900 ng/kg-day (MDH, 2018).

Studies of health effects of PFBA in humans are lacking.

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List of acronyms

General acronyms

Table 74. Acronyms found in the health appendix.

Acronym	Definition				
ACE	Asian Pacific Islander Community Exposures				
ADj. BMD	Adjustment benchmark dose				
AFB	Air Force Base				
AFFF	Aqueous film forming foam				
ALT	Elevated liver enzyme				
ANSES	French Agency for Food, Environmental and Occupational Health and				
	Safety				
ATSDR	Agency for Toxic Substances and Disease Registry				
BMD	Benchmark dose				
CDC	Centers for Disease Control and Prevention				
DOD	United States Department of Defense				
DEPA	Danish EPA				
Ecology	Washington State Department of Ecology				
EFSA	European Food Safety Authority				
EJ	Environmental Justice				
EPA	United States Environmental Protection Agency				
ERα	Estrogen receptor alpha				
ESTCP	Environmental Security Technology Certification Program				
FDA	United States Food and Drug Administration				
Health	Washington Department of Health				
HED	Human equivalent dose				
HBM commission	German Human Biomonitoring Commission				
Hib	Haemophilus influenza type b				
IARC	International Agency for Research on Cancer				
IBL	Information by Location, a tool used in Washington Tracking Network				
IgE	Immunoglobulin E				
ITRC	Interstate Technology Regulatory Council				
JBLM	Joint Base Lewis-McChord				
L	Liter				
LDL	low-density lipoprotein				
LOAEL	Lowest observed adverse effect level				
μg/L	Microgram per liter				
μg/kg	Microgram per kilogram				
MAMAS	Measuring Analytes in Maternal Archived Samples				
MCL	Maximum contaminant level				
MDH	Minnesota Department of Health				

Acronym	Definition			
Mil-spec	Military performance requirements			
MTCA	Model Toxics Control Act			
NAS	Naval Air Station			
ng/g	Nanogram per gram			
ng/kg	Nanogram per kilogram			
ng/kg-day	Nanogram per kilogram per day			
ng/mL	Nanogram per milliliter			
ng/m ³	Nanogram per cubic meter			
NHANES	CDC National Health and Nutrition Examination Survey			
NHDHHS	New Hampshire Department of Health and Human Services			
NIEHS	National Institute of Environmental Health Sciences			
NOAEL	No observed adverse effect level			
NTP	National Toxicology Program			
OLF	Outlying Landing Field			
PBPK	Physiologically based pharmacokinetic			
PPARα	Peroxisome proliferator-activated receptor alpha			
ppt	Parts per trillion			
PSA	Prostate specific antigen			
PWS	Public drinking water system			
RfD	Reference dose			
SBOH	State Board of Health			
SERDP	Strategic Environmental Research and Development Program			
TRV	Toxicity reference values			
TSH	Thyroid simulating hormone			
TWI	Tolerable weekly intake			
T3	Triidothyonine			
T4	Thyroxine			
UCMR3	Unregulated Contaminant Monitoring Survey			
WTN	Washington Tracking Network			

Chemical names

Table 75. Chemical name acronyms found in the health appendix, excluding the acronyms in the table above.

Acronym	Chemical name
6:2 FTS	6:2 fluorotelomer sulfonate
6:2 FTSA	6:2 fluorotelomer sulfonic acid
APFO	Ammonium perfluorooctanoate
EtFOSA	N-Ethyl perfluorooctane sulfonamide
EtFOSE	N-Ethyl perfluorooctane sulfonamidoethanol
FTAC	Fluorotelomer acrylate
FTOH	Fluorotelomer alcohol

Acronym	Chemical name			
HFPO-DA(GenX)	Hexafluoropropylene oxide dimer acid			
MeFBSA	N-Methyl perfluorobutane sulfonamide			
MeFBSE	N-Methyl perfluorobutane sulfonamide ethanol			
MeFOSE	N-Methyl perfluorooctane sulfonamido ethanol			
MeFOSA	N-Methyl perfluorooctane sulfonamide			
Me-PFOSA-AcOH	2-(N-methyl-perfluorooctane sulfonamido) acetic acid			
N-EtFOSEs	Perfluorooctane sulfonamido ethanols			
N-MeFOSE	N-Methyl perfluorooctane sulfonamido ethanol			
PFAA	Perfluorinated alkyl acid			
PFAS	Per- and polyfluoroalkyl substances			
PFBA	Perfluorobutanoic acid			
PFBS	Perfluorobutane sulfonic acid			
PFBuS	Perfluorobutane sulfonic acid			
PFCA	Perfluoro-carboxylic acid			
PFDA	Perfluorodecanoic acid			
PFDeA	Perfluorodecanoic acid			
PFDoDA	Perfluorododecanoic acid			
PFHxA	Perfluorohexanoic acid			
PFHpA	Perfluoroheptanoic acid			
PFHxS	Perfluorohexane sulfonate			
PFNA	Perfluorononanoic acid			
PFOA	Perfluorooctanoic acid			
PFOS	Perfluorooctane sulfonic acid			
PFOSA	Perfluorooctane sulfonamide			
PFPeA	Perfluoro-n-pentanoic acid			
PFSA	Perfluorooctane sulfonic acid			
PFUnDA	Perfluoroundecanoic acid			

Appendix 8: Biosolids

8.0 Overview

8.0.1 Findings

Biosolids are nutrient- and organic-rich residuals from wastewater treatment. They are land applied on agricultural fields as a soil amendment and fertilizer under a regulated program. Washington's biosolids rule incorporates federal standards, and requires permitting specific sites and approval of application rates and procedures.

An extensive risk assessment was conducted by the U.S. Environmental Protection Agency (EPA) prior to the promulgation of the federal biosolids rule. Three National Sewage Sludge Surveys have been conducted to assess contaminants in biosolids thought to pose risks to human health and the environment. Per- and polyfluoroalkyl substances (PFAS) were not evaluated under the initial risk assessment or the sewage sludge surveys, even though PFAS compounds were widely used throughout the period.

Biosolids PFAS concentrations in the scientific literature have been measured using a variety of methods, although the dense organic matrix has made accurate and precise results difficult to obtain. EPA is currently validating specific methods for PFAS analysis suitable for biosolids and soil. Completion of the validation process is expected sometime in 2020.

Scientific studies evaluating PFAS from land-applied biosolids have investigated results of extremely high application rates, biosolids contaminated by direct industrial production, or used artificial spiking of PFAS compounds. These conditions are not reflective of the rates, likely concentration, or availability of PFAS in Washington biosolids under current rules.

Worldwide, concentrations of the two most common PFAS, perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA), in biosolids have trended downward following reduced production of these congeners. Since there is no known industrial production of PFAS in Washington, biosolids exposure pathways in Washington are primarily from homes and consumer products. Secondary manufacturers may be a source of some contamination in municipal waste streams, but primary exposure is largely from consumer products. Reduction of PFAS in consumer products will lower direct PFAS exposure and the indirect concentrations that may occur in Washington biosolids. Currently, no data exists for PFAS in Washington biosolids.

Several states in the U.S. are considering setting PFAS contaminant levels in soil and biosolids. A PFAS task force in Maine developed PFAS biosolids standards of low ppb for PFOS, PFOA, and perfluoro butane sulfonate (PFBS). Leaching models used in the calculations of these limits use impractical values for parameters such as the fraction of organic carbon in soil (F_{OC}) and degree of molecular sorption (K_{OC}). This can result in calculating unrealistically low soil contaminant limits.

Adoption of extremely low regulatory limits for soil PFAS could have adverse consequences for organics and residual recycling. Such limits could interfere with established goals and benefits of recycling programs, but may not provide demonstrated risk-reduction for human health and the environment.

Risk assessment of PFAS in land-applied biosolids requires a baseline dataset for PFAS that includes:

- Assessment of concentrations in Washington biosolids.
- Measurement of soil concentrations directly attributable to land-applied biosolids under conditions that mimic current state rules.
- Evaluation of contaminant modeling that uses realistic values for input parameters.
- A review of exposure pathways.

8.0.2 Introduction

This appendix presents information regarding the presence of PFAS in biosolids produced as a result of wastewater treatment. It reviews the evolution of how state and federal regulations address PFAS in biosolids, discusses the lack of accredited analytical methods to measure PFAS in biosolids, and presents current knowledge regarding biosolids PFAS concentration trends. Impacts of PFAS-contaminated biosolids application are reviewed and placed into the context of typical biosolids recycling and use in the state.

8.1 Background

Amendments to the Federal Water Pollution Control Act of 1948, now known as the Clean Water Act (CWA), set the creation of wastewater treatment across the U.S. in motion. Large scale construction of wastewater treatment plants (WWTPs) that included secondary treatment was initiated in 1972 when these facilities were nationally funded under a grant program administered by EPA.

One of the primary functions of wastewater treatment is to remove solids from the influent. Treatment plants utilize a variety of engineering designs, but most employ some sort of biological treatment whereby aquatic bacteria consume (i.e., digest) the organic constituents in the influent. The biological and organic floc—along with mineral and some chemical constituents—is settled out of the wastewater prior to discharge of effluent. These solids are typically high in organic matter and mechanically dewatered. Some facilities in arid climates airdry the solids as a primary method of dewatering or in addition to a mechanical process.

In Washington, biosolids are land applied for their nutrient and soil amending properties. Land application of biosolids is conducted primarily in conjunction with commercial farming operations across the state. Washington State Department of Ecology (Ecology) approves individual biosolids applications on an agronomic basis—matching nitrogen needs of the crop with that supplied by biosolids. Analysis of both soil and biosolids is required by rule to calculate site-specific rates on permitted fields in advance of application.

8.2 Federal and state regulations

EPA administers the federal biosolids rule (40 Code of Federal Regulations (CFR) Part 503), under which specific sampling, analysis, and management is required of WWTP residuals. Requirements under the federal rule were developed during extensive scientific review and risk analysis conducted by EPA over a multi-year period preceding the adoption of the federal rule in February 1993. Under federal rules, the solids generated by wastewater treatment are called "sewage sludge."

Washington regulation (Chapter <u>173-308</u>¹²¹ Washington Administrative Code (WAC)—Biosolids Management) differentiates between wastewater solids that meet the regulatory standards to allow land application, classified as "biosolids," and solids not meeting the standards, defined as "sewage sludge." Washington law requires that biosolids are land applied to the greatest extent possible, but that sewage sludge be disposed in a landfill. Based on etimates derived from Annual Report data compiled by Ecology (2017), currently about 85 – 90% of biosolids generated in Washington are land applied.

Washington's biosolids rule adopts all the standards in the federal rule regarding sampling and analysis of WWTP solids, but analysis for PFAS is not required. The Washington rule imposes additional management criteria related to:

- Land application, site evaluation, and permitting.
- Development of management plans that govern the land application procedures.
- Ongoing oversight and approval of application rates and operations.

8.3 Biosolids risk assessment: Rule development, national surveys, and National Research Council

EPA developed the federal rule after undertaking a substantive nine-year evaluation of sewage sludge land application. The process included an "extensive multi-pathway risk assessment for evaluating and setting limits to manage pollutants in biosolids" (EPA, 1995). It involved making a list of pollutants, developing risk-assessment methodologies, determining pollutant limits, defining management practices, and issuing the rule.

In 1984, EPA identified a list of 200 potential pollutants in wastewater residuals for evaluation. Included in this list were a range of toxic organics such as dioxins, furans, polyaromatic hydrocarbons, pesticides, and herbicides. A scientific panel reviewed this list and made a recommendation that approximately 50 of these pollutants be evaluated for further study (EPA, 1995). The evaluation considered toxicity, occurrence, and fate and effects of the pollutants, with a focus on pathways of exposure.

¹²¹ https://apps.leg.wa.gov/wac/default.aspx?cite=173-308

In 1988, EPA conducted the first National Sewage Sludge Survey (NSSS) (EPA, 1992b) to develop a reliable database in support of the final Part 503 biosolids regulation. Samples were collected from 180 Publically Owned Treatment Works (POTWs). These samples were analyzed for more than 400 pollutants according to analytical protocols adapted specifically for the matrix of biosolids. EPA also reviewed the operational practices of 462 POTWs that utilized secondary treatment.

Following the initial survey, two subsequent NSSS have been conducted. In 2007, the survey prioritized an evaluation of dioxins (EPA, 2007), and a Targeted NSSS published in 2009 focused on pharmaceuticals and personal care products (EPA, 2009). Although in wide use at the time, neither PFAS as a chemical class, nor the specific congeners PFOS and PFOA were evaluated.

The National Research Council (NRC) has twice reviewed the federal rule, 40 CFR Part 503. In 1996 the NRC released "Use of Reclaimed Water and Sludge in Food Crop Production" and in 2002 reviewed the science and methodology underlying the health and environmental standards entitled "Biosolids Applied to Land: Advancing Standards and Practices." Both studies concluded that the federal rule was protective of human health and the environment, but PFAS substances were not specifically part of these evaluations. The 2002 NRC review stated that "there is no documented scientific evidence that the Part 503 rule has failed to protect public health."

Pursuant to the CWA Section 405(d), EPA must review the biosolids regulations every two years. They are directed to identify additional toxic pollutants that show sufficient evidence of harm and establish management practices protective of human health and the environment. An international study in 2011 stated that research on organic contaminants in biosolids has been undertaken for more than 30 years, and the increasing body of evidence demonstrates that the majority of compounds studied do not place human health at risk when biosolids are land applied on farmland (Clarke & Smith, 2011). The study cautions that "continued vigilance in assessing 'emerging' organic contaminants in sludge is necessary to support and ensure the long-term sustainability and security of the beneficial agricultural route for biosolids management."

8.4 PFAS analysis methods for biosolids

The required analytical methods and analytes for WWTP residuals in the U.S. are specified by EPA in the federal rule (40 CFR Part 503) and incorporated into the Washington state rule (Chapter 173-308¹²² WAC). Municipalities are required to test their biosolids for a range of chemical parameters including nutrients and regulated metals, but PFAS is not a required analyte. The frequency of testing is determined by the quantity of biosolids the facility generates, with larger facilities required to conduct more frequent testing.

¹²² https://apps.leg.wa.gov/wac/default.aspx?cite=173-308

Regulatory analysis of biosolids in Washington state is required to be conducted by Ecology-accredited laboratories. WWTP residuals in Washington state are considered sewage sludge until they are analyzed by accredited labs using specified methods with the results meeting minimum standards. Residuals meeting the standards in the Washington rule are deemed biosolids and are required to be beneficially used.

Wastewater residuals are a dense organic matrix and have proved difficult to accurately analyze. EPA spent considerable time developing the appropriate methodologies required in the federal rule in order to achieve accurate and consistent results. Laboratories often commit the analytical error of conducting biosolids analyses using methods developed for water and wastewater (EPA, 1992a).

As identified in Appendix 2: Analytical Methods, Section 2.1, EPA has developed Method 537 for analyzing PFOS, PFOA, and 12 other PFAS in drinking water (EPA, 2018a). Some U.S. labs are analyzing biosolids using modified procedures based on EPA Method 537, but guidelines are inconsistent and results have not been validated (Personal communication, Elizabeth Resek, Biosolids Program Lead at EPA, 2018). Ecology's lab accreditation unit at Manchester Environmental Laboratory has performed a technical review of one laboratory in Washington for analysis of a limited number of PFAS compounds in solids and chemical materials using a modified 537 method. Manchester has also recognized the accreditation of the National Environmental Laboratory Accreditation Program (NELAP) for a few other Washington laboratories for PFAS analysis in the solids and chemical materials matrix using modified 537 methods (Personal Communication, Rebecca Wood, Unit Supervisor of Laboratory Accreditation Unit, Manchester Environmental Laboratory, Washington Department of Ecology, 2018). Such accreditation ensures that the procedures are being appropriately followed, not that the method provides accurate and consistent results when analyzing biosolids.

EPA is in the process of validating a different procedure for analyzing biosolids and soil for PFAS—SW-846. Phase I was carried out in the winter of 2017 for 24 PFAS in various media. Phase II of this process was conducted in the fall of 2018 and several external labs are in the process of validating these procedures for public review (EPA, 2018a). It is unlikely that EPA will have finalized its approval of method(s) for PFAS analysis in biosolids before 2020 (Beecher & Brown, 2018; Personal communication, Elizabeth Resek, Biosolids Program Lead at EPA, 2018).

Separate from the SW-846 analysis procedures being reviewed by EPA is another analysis method for PFAS—Total Oxidizable Precursor (TOP) assay. This method uses a chemical oxidation pretreatment. While a number of commercial labs offer the TOP assay, the oxidation can be more or less aggressive depending on the details of the procedure (Masunaga, 2017). Also, there is no currently settled methodology or agreed-upon best approach. TOP is not an EPA method, but is the only commercialized screening tool to measure the presence of PFAA (perfluoroalkyl acid) precursors.

8.5 PFAS concentration and trends in biosolids

The concentrations of PFOS and PFOA in biosolids have been reported from a variety of sources outside of Washington state (Arvaniti & Stasinakis, 2015; Loganathan et al., 2007; Sepulvado et al., 2011; Ulrich et al., 2016; Venkatesan & Halden, 2013; Zareitalabad et al., 2013). The literature reports PFOA concentrations from 7 – 219 nanograms per gram (ng/g) and PFOS from less than 2.5 – 990 ng/g. PFOS is often reported as the most abundant among the PFAS compounds (Arvaniti & Stasinakis, 2015; Sepulvado et al., 2011). Four WWTPs in Washington had effluent analysis for PFOS and PFOA, but this review did not include an analysis of biosolids for these compounds (Furl & Meredith, 2010).

In general, the chemistry of biosolids is reflective of the chemistry of people's daily lives, as is the dust in homes (Haug et al., 2011; Hundal, Lakhwinder, Kumar, & Basta, 2011). Washington residents are exposed to PFAS from carpets, food packaging, personal care products and cosmetics, surface coatings on textiles, paints, lubricants, waterproof fabric, ski wax, and a wide variety of other sources. Therefore, it would not be surprising if there were trace amounts in Washington biosolids.

Industrial sources of perfluoroalkylates can influence concentrations of these compounds in biosolids when a WWTP receives influent directly from industries that work with fluorotelomer compounds. A WWTP in Decatur, Alabama received discharge from industries that conducted electrochemical fluorination and worked with a variety of fluorotelomer compounds and perfluoroalkylates (Washington et al., 2010). The data for PFOA concentrations from Decatur sewage sludge are fragmentary, but show high levels in 2005 and 2006: 528 ng/g and 683 ng/g in 2005, and 1,875 ng/g in 2006. Subsequent to significant reduction from industrial discharges, the concentration of PFOA in the Decatur biosolids decreased markedly. The reported PFOA concentrations in biosolids were 50 ng/g and 128 ng/g in 2007, and 27 ng/g and 32 ng/g in 2008 (Washington et al., 2010).

Washington state does not have commercial production of PFAS compounds, although secondary manufacturing using PFAS chemicals may occur within the state (see <u>Appendix 1: Chemistry</u>, <u>Section 1.3</u>). This suggests that there may be some industrial discharge, but the vast majority of perfluorinated compounds in Washington municipal wastewater would originate from domestic sources—our homes and consumer products. Contamination such as that identified in Alabama biosolids is highly unlikely to occur in Washington.

A trend of decreasing concentrations of PFOA and PFOS is observed across a broad spectrum of data characterizing biosolids using a variety of analytical methods. A review of sewage sludge in Germany evaluated perfluoroalkyl acids (PFAAs) concentration from 4,981 samples from 1,165 WWTPs collected between 2008 and 2013 (Ulrich et al., 2016). Seventy-one WWTPs had samples exceeding a European Union (EU) precautionary level of 125 ng/g, but this occurred with decreasing frequency over time. The exceedances decreased from 6% in 2008 to 0.8% in 2013. During the same timeframe, WWTPs uncontaminated with PFOS and PFOA increased by

32%. In the samples evaluated, PFOS was found in 41% and PFOA in 7%. Forty-seven percent of WWTPs showed clear decreases over time and 16% showed an increasing trend. The total load of PFAAs in sewage sludge was reduced by more than 90% during this time period. These reductions are likely the result of the decreased production of PFOS and PFOA.

In 2013, archived samples of biosolids from the NSSS from 2001 were combined into five composite samples and analyzed for PFOA and PFOS. The average concentration of PFOS was 276 – 430 ng/g, and PFOA was 12 – 56 ng/g. (Venkatesan & Halden, 2013). These archived NSSS samples represented 94 WWTPs in 32 states, but did not include Washington state. A summary of PFAS compounds in sewage sludge from 2005 to 2015 monitoring data worldwide was compiled by carbon chain length at concentrations of ng/g (Arvaniti & Stasinakis, 2015). With few exceptions, these more recent samples are lower for PFOS and PFOA than the composite results from samples in the EPA NSSS of 2001. A reduction in PFOS and PFOA levels in human blood (Calafat et al., 2007) was also observed in data compiled from the National Health and Nutrition Examination Surveys (NHANES). They conclude this is "most likely related to discontinuation in 2002 of industrial production of PFOS and related perfluorooctanesulfonyl fluoride compounds."

8.6 Literature review of biosolids land application effects

In the case from Decatur, Alabama referenced above, biosolids were land applied to about 2,000 hectares (ha) of agricultural fields for more than a decade (Washington et al., 2010). The elevated levels of PFAS in the biosolids generated concern that land application may constitute a pathway to contaminate surface and groundwater. In order to evaluate this risk, EPA collected some initial soil samples in 2007 from Decatur land application sites and from nearby background fields that did not receive biosolids. Results indicated the presence of high concentrations of several fluorotelomer alcohols (FTOHs) and PFAS in soil. After collection of these initial soil samples and public drinking water samples, EPA collected an expanded set of soil samples in 2009 to more accurately characterize the extent of contamination in and around the land application sites. These land application activities and the subsequent EPA review have received widespread coverage in news reports and have been noted in a variety of websites (Fluoride Action Network, 2009; Northwestern University, n.d.; Renner, 2009).

The soil from sludge-applied fields in Alabama had PFAS concentrations that were higher than the background field samples. The highest PFOA concentrations from sludge-applied fields were less than or equal to 320 ng/g, and PFOS were less than or equal to 410 ng/g (Washington et al., 2010). Annual application rates of Decatur biosolids for a five-year period between 2002 and 2006 averaged approximately 32 Megagram per hectare (Mg/ha) ranging from 14.9 – 60 Mg/ha. These amounts are well above Washington's mean application rate of 6.95 Mg/ha calculated from 809 regulatory approvals for land application of biosolids for Alfalfa or grass hay, barley, canola, corn, hops, sunflowers, triticale, and wheat over the years 2010 – 2017, for which data are available (Severtson, 2017). The minimum Decatur rate exceeds all but six of the

809 Washington approvals. The six higher land application rates approved in Washington were for lagoon biosolids that contained significant amount of mineral material (sand) and low nitrogen (N) content. From the perspective of an agronomic evaluation, application rates used for the Decatur biosolids would have likely resulted in excessive N accumulations and leaching of nitrate. Such rates would be unlikely to receive regulatory approval in Washington.

Sepulvado et al. (2011) land applied municipal biosolids from Chicago to investigate questions about the fate of perfluorochemicals (Sepulvado et al., 2011). This investigation indicated four significant results:

- Concentrations of PFAS in soil increased linearly with increasing biosolids loading rate (PFOS = 2 483 ng/g).
- Desorption experiments suggested the leaching potential of perfluorochemicals decreased with increasing carbon chain length.
- Previously derived organic carbon partition coefficients may not be accurate predictors of the desorption of long-chain PFAS compounds.
- Trace levels of short-chain PFAS were detected in soil cores below the level of incorporation.

The Chicago biosolids in the Sepulvado et al. (2011) study were land applied at very high cumulative loading rates. Their long-term plots had applications over 32 years amounting to 553 Mg/ha (low rate), 1,109 Mg/ha (medium rate), and 2,218 Mg/ha (high rate) (Sepulvado et al., 2011). Although the PFAS soil concentrations were linearly correlated with application rates, the loading rates in the Sepulvado et al. (2011) study were significantly higher than the mean Washington agronomic rate of 6.95 Mg/ha. It would require 79, 159, and 319 years of annual applications respectively at Washington's average application rate to achieve similar cumulative loading.

However, most fields in Washington do not have biosolids applied annually. On a wheat-fallow, rotation applications are made every other year at most, and commonly every four years. Applications on alternate years would require a minimum of 158 years to achieve the lowest cumulative biosolids loading in the Sepulvado et al. (2011) study. These were biosolids likely to have higher levels of PFOS and PFOA than Washington biosolids, due to the dates when they were produced and potential industrial contamination.

In a spiked soil study, Stahl et al. (2009) reported bioaccumulation and that PFOA and PFOS at very high concentrations can result in diminished growth of spring wheat. However, spiked-soil studies are known to create results not seen in field investigations with typical agronomic application rates. Studies by Blaine et al. (2013) and Pannu et al. (2012) showed that biosolids amended spiked-soil studies can show artificially greater accumulation in plants and earthworms as compared to samples grown in field soils (Blaine et al., 2013; Pannu, O'Connor, & Toor, 2012). Similarly, regarding metal uptake, Brown et al. (1998) demonstrated that "significantly less cadmium (Cd) was taken up by lettuce grown on biosolids-amended soil than lettuce grown on soil amended with equivalent rates of a Cd salt."

Negative impacts on crop growth are not representative of yield data from biosolids applications in Washington state. Results from a long-term Washington State University study evaluating biosolids applications to winter wheat and canola have shown significant long-term yield increases from biosolids applications compared with the control or mineral fertilizer additions (Cogger et al., 2013). This is despite the fact that there are likely to be trace amounts of PFAS in the biosolids. However, there are no PFAS soil concentration data from this site nor has EPA addressed agricultural soil concentration limits to date.

In a widely distributed greenhouse and field study of plant uptake of perfluoroalkyl acids (PFAAs) from biosolids, Blaine et al. (2013) looked at PFAA concentrations in lettuce (*Lactuca sativa*) and tomato (*Lycopersicon lycopersicum*) grown in biosolids-amended soils. The greenhouse portion of the study used industrially impacted biosolids, biosolids from a long-term application site, as well as "clean" soil. They calculated bioaccumulation factors (BAFs), looking at concentrations in soil relative to plant concentrations primarily from the greenhouse portion of the trial. They conclude that the "study confirms that the bioaccumulation of PFAAs from biosolids-amended soils depends strongly on PFAA concentrations, soil properties, the type of crop, and analyte."

In the Blaine et al. (2013) field scale trial using lettuce and tomato, and a full-scale field study with corn, the plant concentrations were below the level of quantification (LOQ) for all treatments except the 4x agronomic rate (100 Mg/ha), which is 14 times the average application rate in Washington. Given the results of the Blaine et al. (2013) study, it is highly unlikely that grain would exhibit concentrations above the LOQ as a result of agronomic applications in Washington, where PFOS and PFOA concentrations are likely to be very low due to lack of industrial production.

The Blaine et al. (2013) greenhouse study shows how small-scale investigations into bioaccumulation can differ significantly from regulated, field-scale applications in Washington. The study used three types of soil: control, "industrially impacted," and "municipal." The industrial soil was created by adding a 10% (dry weight) biosolids compost and the municipal soil originated from cumulative applications of municipal biosolids that totaled 1,654 Mg/ha. The industrially impacted soil had PFOA concentrations of 78.5 ng/g and PFOS concentrations of 49.7 ng/g. The municipal soil had PFOA concentrations of 14.9 ng/g and PFOS concentrations of 319.5 ng/g.

In both cases, soil concentrations are orders of magnitude higher than would realistically result in Washington state from agronomic biosolids applications. Indeed, it would be impossible to reach the PFAS soil concentrations seen in the Blaine et al. (2013) study if initial biosolids concentrations are significantly lower than those modeled. Tables 76 and 77 show the time necessary to reach such concentrations—using biosolids PFAS concentration data from the literature, combined with the mean Washington biosolids application rate. Actual PFAS concentrations in Washington biosolids are likely to be significantly lower than these values.

Table 76. Estimates of the time needed to reach Blaine et al. (2013) municipal soil concentrations from annual applications of biosolids using various concentration estimates and mean Washington application rate in megagrams per hectare (6.95 Mg/ha).

Biosolids PFOS/PFOA concentration (ug/kg)	Biosolids application rate ⁽¹⁸⁾ (Mg/ha)	PFOS/PFOA applied per application- dry weight (ųg)	Soil depth (cm)	Soil weight (kg/ha)	Calculated soil conc. (ųg/kg)	Blaine et al. (2013) municipal soil conc. (ug/kg)	Years to reach soil levels*
PFOS: 19	6.95	PFOS:	15	2,000,000	PFOS:	PFOS:	PFOS:
PFOA: 10		132,050			0.066	319.5	4,841
(Ulrich, 2016)		PFOA:			PFOA:	PFOA:	PFOA:
		69,500			0.035	14.9	426
PFOA: 32	6.95	220,400	15	2,000,000	PFOA:	PFOA:	PFOS:
(Washington et al., 2010)					0.110	14.9	135
PFOS: 403	6.95	PFOS:	15	2,000,000	PFOS:	PFOS:	PFOS:
PFOA: 34		2,800,850			1.400	319.5	228
(EPA, 2001)		PFOA:			PFOA:	PFOA:	PFOA:
		236,300			0.118	14.9	126

Note: * = One application annually, summed empirical amounts only (no degradation or leaching of PFOS or PFOA).

Table 77. Estimates of the time needed to reach Blaine et al. (2013) industrial soil concentrations from annual applications of biosolids using various concentration estimates and mean Washington application rate in megagrams per hectare (6.95 Mg/ha).

Biosolids PFOS/PFOA concentration (ug/kg)	Biosolids application rate ⁽¹⁸⁾ (Mg/ha)	PFOS/PFOA applied per application- dry weight (ųg)	Soil depth (cm)	Soil weight (kg/ha)	Calculated soil conc. (ųg/kg)	Blaine et al. (2013) industrial soil conc. (ug/kg)	Years to reach soil levels*
PFOS: 19	6.95	PFOS:	15 cm	2,000,000	PFOS:	PFOS:	PFOS:
PFOA: 10		132,050			0.066	49.7	753
(Ulrich, 2016)		PFOA:			PFOA:	PFOA:	PFOA:
		69,500			0.035	78.5	2,243
PFOA: 32	6.95	220,400	15 cm	2,000,000	0.111	78.5	PFOA:
(Washington							707
et al., 2010)							
PFOS: 403	6.95	PFOS:	15 cm	2,000,000	PFOS:	PFOS:	PFOS:
PFOA: 34		2,800,850			1.400	49.7	36
(EPA, 2001)		PFOA:			PFOA:	PFOA:	PFOA:
		236,300			0.118	78.5	665

Note: * = One application annually, summed empirical amounts only (no degradation or leaching of PFOS or PFOA).

8.7 Factors influencing risk assessment of PFAS in Washington biosolids

8.7.1 PFAS concentration data

PFOS was voluntarily phased-out of production in the U.S. between 2000 and 2002 by its primary producer, 3M Company (EPA, 2016). Since 2006, eight global manufacturers participated in a voluntary phase-out of PFOA by 2015 (EPA, 2018b). Reduced production of PFOS and PFOA is likely the reason for the lower reported concentrations of these chemicals in sewage sludge and biosolids in recent years. The biosolids PFOS and PFOA data in Germany (Ulrich et al., 2016) and locations worldwide using a variety of analytical methods suggest that concentrations of these chemicals are trending downward.

There is currently no PFAS data from biosolids generated in Washington. In June 2018, regulators and officials from major producers of biosolids across the state discussed the issue of PFAS data. There were a number of unresolved issues regarding how samples would be collected, what analysis method(s) would be used, who would pay for any analysis, data use and evaluation, and public dissemination of proprietary analysis results.

PFAS data in the literature from biosolids outside of Washington has been obtained using a variety of analytical methods. The accuracy and precision of these data is uncertain considering that EPA has not concluded its validation of analysis methods in a biosolids or soil matrix. It is "virtually impossible" to correlate PFAS soil data gathered across different states that have used various sample collection procedures and non-validated analysis methods (Personal communication, Ned Beecher, Northest Biosolids Association, 2019).

Some commercial laboratories are claiming they can measure PFAS in solid matrices (biosolids and soil) at reporting limits as low as 0.2 ng/g (ppb). Claims of such precision in analysis results are suspect, because actual lab results often show reporting limits in the range of 2-5 ppb. Different methods also show wide variation in results. For example, Vermont DEC conducted split sample tests comparing a DOD-preferred isotope dilution method (MLA 110) with a modified Method 537 (Weston & Sampson, 2018). When analyzing wastewater, they found differences in results ranging from 10% - 200%. When analyzing wastewater solids, the range of difference between the methods exceeded 300%.

8.7.2 Modeling Data

New York and Maine have made attempts at groundwater migration models to estimate leaching of PFAS in soil. The models used to derive soil screening values have not been field-verified for the PFAS chemicals, and there is insufficient published research on soil leaching of biosolids-sourced PFAS to allow for robust understanding of the potential leaching risks.

The sorption of perfluorinated compounds to soil influences their fate and distribution in the environment after land application. There is evidence that PFOS and PFOA persistence in soil is

related to carbon-chain length, with longer carbon-chains being more persistent and less mobile in soil (Vanketasan and Halden, 2013; Calafat et al., 2007). Laboratory determined organic-carbon partitioning coefficients (K_{OC}) are often used to predict potential mobility of organic contaminants in the environment. The K_{OC} values can vary based on the method used for calculation (Snyder, O'Connor, & McAvoy, 2010), and derived values appear to differ from gross distribution of PFAS compounds in the environment. Lab-based Log K_{OC} values may overestimate PFOS and PFOA concentrations in water and underestimate soil residence time (Zareitalabad et al., 2013).

Determining K_{OC} values that are reflective of the environmental fate of biosolids-sourced PFAS compounds has proven difficult. It is likely that the database for K_{OC} values for the range of perfluorinated compounds is incomplete and may not provide adequate information to accurately model movement in a soil system.

Given that biosolids are settled out of an aqueous media, PFAS congeners with lower sorption are likely to leave wastewater treatment in the effluent. This may reduce overall PFAS amounts and provide an inherent bias for higher sorption congeners (higher K_{OC}) to remain in biosolids. This may result in reduced mobility of biosolids-sourced PFAS relative to the suite of PFAS congeners found in the WWTP influent. Thus, field-scale studies investigating the transfer or leaching of biosolids-sourced PFAS in natural soil systems are important to evaluate actual mobility and risk from biosolids land application.

Leaching models use a number of factors including the Fraction of Organic Carbon (F_{OC}) in soil and K_{OC} . Small changes in these factors directly affect model outcomes. Alaska Department of Environmental Conservation (ADEC) is currently evaluating soil standards based on a leaching model where the F_{OC} is assumed to be 0.1%. This is an unrealistic value associated with land applied biosolids where PFAS compounds would be land applied in a dense, organic-matter matrix, and likely be applied to soils with significantly higher organic matter content. The Alaska model also used EPA's lab-based log K_{OC} values, which are not field verified.

ADEC's online calculator run with more realistic inputs for organic content and partitioning coefficients resulted in significantly higher calculated soil PFAS limits (Lono-Batura, Maile, Beecher, Ned, Franciosi, & Riggs, 2018). The State of Maine PFAS task force recently developed screening concentrations of 2.5 ng/g for PFOA, 5.2 ng/g for PFOS, and 1,900 ng/g for PFBS for biosolids that may be land applied. The levels were developed using SESOIL and AT123D models, which are primarily based on the leaching from soil to groundwater pathway (Maine Department of Environmental Protection, 2018). Maine also required all Maine utilities to test the biosolids for three PFAS compounds before any land application can occur. Following the setting of screening levels, representatives from various wastewater utilities wrote a letter to the PFAS task force and requested the screening levels be based on real-world research and not on fate and transport models.

Data and modeling uncertainties inhibit accurate assessment of risk to human health and the environment from biosolids-sourced PFAS land applied at agronomic rates in Washington. Biosolids are applied to less than 1% of state farmland on an annual basis. Applications occur only on permitted fields with rates based on crop type, yield, biosolids nutrient content, and site-specific soil analysis. PFAS in Washington biosolids result primarily from domestic sources and concentrations are likely to be very low. Mean application rates result in soil dilution exceeding 1,000-fold in the top two feet. State regulations regarding site restrictions also limit the pathways of exposure. These conditions, combined with available data, mean we can reasonably expect that health risks directly attributable to biosolids-sourced PFAS from land application in Washington are likely low.

8.8 Biosolids policy discussion

Biosolids management entities include private business, public utilities, and regulatory agencies. Issues regarding contaminants of emerging concern have occurred numerous times. The focus of concern has included a variety of categories such as pesticides, dioxins, PBDEs, antimicrobial compounds, and pharmaceuticals. Typically, the concentrations of these chemicals in biosolids have been very small, and until recent years, analytical techniques did not afford consistent identification and quantification. Current analytical reports on PFAS indicate identifiable concentrations in parts per billion and parts per trillion in a variety of media, drinking water in particular. This has renewed concern over the presence of contaminants in biosolids.

PFAS are nearly ubiquitous in modern society and Washington biosolids will likely show trace concentrations of PFAS. However, trends in the literature regarding PFOS and PFOA concentrations have shown decreases in biosolids, primarily attributed to reduced production. If PFAS concentrations in Washington biosolids largely reflect the domestic exposure people receive from consumer products in their homes, we would expect these concentrations to be low. As such, upstream source reduction—reduced use of products containing these compounds—will be the direct means of lowering PFAS exposure from biosolids for Washingtonians.

Use of hypothetical leaching models with unrealistic input parameters may calculate unachievable soil contaminant concentration limits. Several states are currently considering a variety of PFAS threshold values for soil based on such modeling. Some of these values for PFAS concentrations in soil may exceed local background levels making them unrealistic and to implement as a regulatory standard. Setting unrealistic (and potentially unenforceable) contaminant thresholds undermines public support for regulation.

Very low regulatory limits for PFAS soil concentrations that are widely applicable could have adverse impacts to businesses and municipalities managing biosolids and other residuals. The economic and management impacts would extend to a variety of end-users of biosolids and

compost products. This may result in hesitancy by generators to publicly release their proprietary analytical results that are not required by rule. As such, without field verification data, Ecology will be cautious of implementing low calculated contaminant thresholds (such as the soil levels being considered in Alaska).

Risk assessment of biosolids land application requires appropriate analytical methods, modeling of biosolids-related contaminant transfer to soil and groundwater, and toxicological data on identified pathways of exposure for Washington residents.

8.9 Data gaps and recommendations

8.9.1 Data gaps

The following data gaps are identified with respect to PFAS in biosolids:

- Washington state has not conducted any biosolids sampling for PFAS with accredited methods, so there is no accredited data on PFAS concentrations in Washington biosolids.
- Background levels of PFAS in regulated land application zones are unknown.
- Soil PFAS concentrations in areas of direct biosolids land application are unknown.
- The source and amount of industrial discharge containing PFAS to municipal WWTPs is unknown.

8.9.2 Recommendations

The information gaps regarding biosolids are significant and currently prevent assessment of risk from PFAS in biosolids land applied in Washington. Any regulatory changes should be founded on defensible data and science-based risk assessments. If scientific modeling is used by Ecology to assess potential PFAS transfer from biosolids to soil or groundwater, realistic model parameters must be used.

Washington biosolids regulation in the near term should ensure sound agronomic land application practices on permitted sites where human exposure is limited. It is premature to add or change regulatory limits given the absence of data from Washington biosolids and problems identified with models and their input parameters.

Based on the analysis conducted in this appendix, we make the following recommendation.

Recommendation 4.3: Evaluate Washington biosolids management.

We recommend the following key steps to address the current data gaps:

- Establish biosolids and soil sample collection and handling methods for PFAS analysis.
- Accredit Washington labs for EPA-validated analysis method(s).
- Use EPA-validated analysis methods for biosolids and soils.

- Conduct credentialed third-party review of raw mass spectrometer PFAS data.
- Investigate land application sites where procedures mimic rates and practices under current state rule (Chapter <u>173-308</u>¹²³ WAC).
- Evaluate realistic exposure pathways.
- Evaluate risk modeling with use of realistic input values.
- Collaborate with stakeholders to get accurate and precise biosolids data. Initial results should remain anonymous.
- Compile analysis data with statistical review.

As part of implementation of these steps, Ecology would:

- Allocate staff resources and funding to support biosolids PFAS data collection (sampling and analysis).
- Establish sampling methods and accreditation of laboratories.
- Collaborate with Northwest Biosolids Association, research institutions, and other stakeholders to establish the protocols and procedures.
- Work collaboratively with a variety of generators to analyze biosolids for PFAS using accredited methods and laboratories.
- Collect and analyze preliminary soil samples from biosolids application sites with known histories that mimic current Washington rules.
- Emphasize triplicate sample analysis in order to evaluate the precision of results for all sampling and analysis recommended in this appendix.
- Evaluate the basis of contaminant limits set in other states and Canada. Such an evaluation would include a review of baseline biosolids data, contaminant models and their parameters, pathways of exposure, and level of uncertainty.

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¹²³ https://apps.leg.wa.gov/wac/default.aspx?cite=173-308

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List of acronyms

General acronyms

Table 78. Acronyms found in the biosolids appendix.

Acronym	Definition
ADEC	Alaska Department of Environmental Conservation
BAF	Bioaccumulation factor
Cd	Cadmium
CFR	Code of Federal Regulations
cm	Centimeter
CWA	Clean Water Act
Ecology	Washington State Department of Ecology
EPA	United States Environmental Protection Agency
EU	European Union
Foc	Fraction of organic carbon in soil or Fraction of organic matter
ha	Hectare
Kg	Kilogram
Koc	Degree of molecular sorption or Organic-carbon partitioning coefficient
LOQ	Level of quantification
Mg	Megagram
ng/g	Nanograms per gram
ųg	Microgram
ųg/kg	Microgram per kilogram
NHANES	National Health and Nutrition Examination Surveys
NRC	National Research Council
NSSS	National Sewage Sludge Survey
POTW	Publicly owned treatment works
ТОР	Total oxidizable precursor
U.S.	United States
WAC	Washington Administrative Code
WWTP	Wastewater treatment plant

Chemical names

Table 79. Chemical name acronyms found in the biosolids appendix, excluding the acronyms found in the table above.

Acronym	Chemical Name
11Cl-PF3OUdS	11-chloroeicosafluoro-3-oxaundecane-1-sulfonic acid
FTOH	Fluorotelomer alcohol
PFAA	Perfluoroalkyl acid
PFAS	Per- and polyfluorinated alkyl substances
PFBS	Perfluoro butane sulfonate
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate

Appendix 9: Regulations

9.0 Overview

9.0.1 Findings

Washington state currently implements the following laws and regulations to reduce per- and polyfluoroalkyl substances (PFAS):

- Chapter <u>70.75A</u>¹²⁴ Revised Code of Washington (RCW): Firefighting Agents and Equipment Toxic Chemical Use Law.
- Chapter 70.95G¹²⁵ RCW: Packages Containing Metals and Toxic Chemicals Law.
- Chapter <u>70.105</u>¹²⁶ RCW and Chapter <u>173-303</u>¹²⁷ Washington Administrative Code (WAC): Dangerous waste regulations.
- Chapter <u>173-333</u>¹²⁸ WAC: Persistent Bioaccumulative Toxics (PBT) Chemical Action Plans (CAPs).
- Chapter <u>70.240¹²⁹</u> RCW and Chapter <u>173-334¹³⁰</u> WAC: Children's Safe Products Act.
- Chapter 70.365¹³¹ RCW: Pollution Prevention for Healthy People and Puget Sound Act.

The following regulatory activities are in progress to address PFAS risk:

• Chapter <u>246-290</u>¹³² WAC: Considering setting drinking water standards for five PFAS in Group A Public Water Supplies.

Regulatory actions at the federal level include:

- Adopting voluntary phase out and stewardship programs to eliminate perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) from commerce in the U.S.
- Developing recommendations for addressing contaminated groundwater and establishing screening levels for PFOS and PFOA.
- Establishing Lifetime Drinking Water Health Advisory levels for PFOS and PFOA.
- Removing two PFAS from the list of approved substances for oil and water repellants for paper and paperboard for use in contact with food.
- Requiring reporting for certain PFAS under Toxics Release Inventory (TRI).
- Implementing various provisions related to military use of products containing PFAS.

¹²⁴ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

¹²⁵ https://app.leg.wa.gov/RCW/default.aspx?cite=70.95G&full=true

¹²⁶ https://app.leg.wa.gov/RCW/default.aspx?cite=70.105

¹²⁷ https://apps.leg.wa.gov/wac/default.aspx?cite=173-303

¹²⁸ https://apps.leg.wa.gov/wac/default.aspx?cite=173-333

¹²⁹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.240

¹³⁰ https://apps.leg.wa.gov/wac/default.aspx?cite=173-334

¹³¹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365

¹³² https://apps.leg.wa.gov/wac/default.aspx?cite=246-290

9.0.2 Introduction

This appendix summarizes Washington state laws and regulations that currently apply to PFAS. The Washington State Department of Ecology (Ecology) administers state and federal laws designed to protect Washington's land, air, and water. Additional discussion is provided of drinking water rulemaking currently underway by the Washington State Department of Health (Health), and two executive orders which also address reducing PFAS exposure and risks. Federal regulations and guidance are also discussed. Finally, resources for understanding PFAS regulations elsewhere in the U.S. and other countries are provided.

Regulatory activity surrounding certain PFAS is developing rapidly. It is outside the scope of this CAP to identify all requirements beyond those applicable in Washington state.

9.1 Washington state laws and regulations

Several Washington state laws, regulations, and executive orders apply to specific PFAS or to PFAS as a class, as summarized in Table 80.

Table 80. Washingto	n laws. res	gulations, and	executive ord	ders applicable to PFAS.
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Regulation	Responsible agency	Reference
Pollution Prevention for Healthy People	Ecology and Health	Chapter 70.365 RCW
and Puget Sound Act Law		
Firefighting Agents And Equipment—Toxic	Ecology	Chapter 70.75A RCW
Chemical Use Law		
Packages Containing Metals And Toxic	Ecology	Chapter 70.95G RCW
Chemicals Law		
Dangerous Waste Regulations Rule	Ecology	Chapter 173-303 WAC
Persistent Bioaccumulative Toxins Rule	Ecology and Health	Chapter 173-333 WAC
Children's Safe Products Act Reporting	Ecology and Health	Chapter 173-334 WAC
Rule		
Group A Public Water Supplies Rule	Health	Chapter 246-290 WAC
Governor's Executive Order	All state agencies	EO 04-01
Governor's Executive Order	All state agencies	EO 18-01

9.1.1 Washington state laws

Chapter 70.240 RCW

The Children's Safe Products Act (CSPA), Chapter 70.240¹³³ RCW, authorized Ecology and Health to develop a list of chemicals of high concern to children and a process for manufacturers to report on the presence of those chemicals in children's products. Manufacturers are required to annually report the presence of listed chemicals present in children's products sold in

¹³³ https://app.leg.wa.gov/RCW/default.aspx?cite=70.240

Washington state. Annual reports include the manufacturer name, product category and component, chemical function, and concentration. The Children's Safe Products Reporting Rule (WAC <u>173-334-130</u>¹³⁴) included PFOS in the list of chemicals of high concern to children upon initial rule adoption in 2011. PFOA was added to the reporting list in 2017.

Chapter 70.365 RCW

Chapter 70.365¹³⁵ RCW, implemented through the <u>Safer Products for Washington program</u>, ¹³⁶ creates a process for Ecology, in consultation with Health, to regulate classes of chemicals in consumer products. The law designates PFAS as a priority chemical class in the first five-year cycle of the program. The law requires Ecology to designate priority chemicals, identify products that contain these chemicals, determine regulatory actions, and, if needed, adopt rules to implement regulatory actions. Chemical restrictions require that safer alternatives are feasible and available.

The law outlines steps that involve stakeholder consultation, legislative reporting, and rulemaking. Ecology published a <u>draft report</u>¹³⁷ in January 2020 and a <u>final report</u>¹³⁸ in July 2020 recommending priority consumer products for further research, including carpeting and rugs, aftermarket stain and water resistance treatments, and leather and textile furnishings containing PFAS.

Chapter 70.75A RCW

Firefighting Agents and Equipment Toxic Chemical Use law (Chapter 70.75A¹³⁹ RCW) applies restrictions to Class B firefighting foam (i.e., PFAS-containing firefighting foam) and PFAS-containing firefighting personal protective equipment. PFAS in this law is defined as a class of fluorinated organic chemicals containing at least one fully fluorinated carbon atom. The law bans the use of PFAS-containing firefighting foam for training purposes by any user as of July 1, 2018. Starting in July 2020, purchase of PFAS-containing firefighting foam is not allowed by most users, including municipal fire departments. Exceptions to the purchase restrictions include federally required purchases (for example, military or federally certified airports), petroleum refineries, and chemical plants. The law requires notification to purchasers of firefighting personal protective equipment if PFAS is used in that equipment and serves a protective function. Ecology is required to enforce these requirements.

In 2020, the law was amended in three ways (Engrossed Substitute H.B. 2265, 2020). First, the allowance for federal facilities (including Department of Defense (DOD) facilities and airports) to purchase PFAS-containing foam will end two years after federal regulations are amended to

¹³⁴ https://apps.leg.wa.gov/wac/default.aspx?cite=173-334-130

¹³⁵ https://app.leg.wa.gov/RCW/default.aspx?cite=70.365

¹³⁶ https://ecology.wa.gov/Safer-Products-WA

¹³⁷ https://fortress.wa.gov/ecy/publications/documents/2004004.pdf

¹³⁸ https://fortress.wa.gov/ecy/publications/documents/2004019.pdf

¹³⁹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.75A&full=true

allow the use of non-PFAS foams. After that date, federal facilities will be required to use non-PFAS foams that comply with the new federal regulation.

Second, 18 months after the federal regulations change, airports will be required to inform Ecology about their ability to switch to non-PFAS foams and Ecology may provide additional time for them to comply with the non-PFAS foam requirements.

Finally, the restriction on purchases of PFAS-containing foams do not apply to oil terminals, oil refineries, and chemical plants until 2024, and extensions may be approved by Ecology under certain circumstances.

Chapter 70.95G RCW

Packages Containing Metals and Toxic Chemicals law (Chapter 70.95G¹⁴⁰ RCW) prohibits PFAS in paper or paperboard food packaging where safer alternatives for specific applications have been determined to exist. PFAS in this law is defined as a class of fluorinated organic chemicals containing at least one fully fluorinated carbon atom. Ecology is required to identify whether safer alternatives to PFAS in food packaging are available, through the completion of an alternatives assessment. A ban on specific PFAS in food packaging takes effect in January 2022 if Ecology identifies that safer alternatives are available for those food packaging applications. If Ecology does not identify safer alternatives, Ecology must review its analysis annually and resubmit findings to the Legislature. The prohibition for specific food packaging applications takes effect two years after the submittal of Ecology's report.

9.1.2 Washington state rules

Chapter 173-303 WAC

Under state hazardous waste law (Chapter 70.105¹⁴¹ RCW), Washington regulates the designation, handling, and disposal of hazardous waste under the state's Dangerous Waste Regulations (WAC 173-303¹⁴²). These regulations include a category of state-only dangerous waste based on either toxicity or persistence. Halogenated organic compounds are state-only persistent wastes. All PFAS are halogenated, therefore any waste containing PFAS at concentrations above 100 parts per million (ppm) designates as a state-only dangerous waste and must be handled and disposed as required by the Dangerous Waste Regulations. Appendix 3: Sources and Uses, Section 3.4.4 Dangerous waste disposal reports, provides a summary of dangerous waste disposal records submitted to Ecology reporting use of firefighting foams.

¹⁴⁰ https://app.leg.wa.gov/RCW/default.aspx?cite=70.95G&full=true

¹⁴¹ https://app.leg.wa.gov/RCW/default.aspx?cite=70.105

¹⁴² https://apps.leg.wa.gov/wac/default.aspx?cite=173-303

Chapter 173-333 WAC

Under the authority of 2004 c276, Executive Order (EO) 04-01, and state hazardous waste law (Chapter 70.105¹⁴³ RCW), Ecology adopted a rule outlining the processes for efforts to reduce and phase out the uses, releases, and exposures to PBT chemicals. This rule includes a list of 28 PBT chemicals, chemical groups, or metals of concern to be considered for CAP development. PFOS and its salts are listed as a chemical group on the list of PBT chemicals in this rule. Table 81 summarizes these below.

CAS Number	Substance	Formula	R-Group
1763-23-1	Perfluorooctane sulfonic acid (PFOS)	C ₈ F ₁₇ SO ₃ H	SO₃H
29081-56-9	Perfluorooctane sulfonic acid, ammonium salt	C ₈ H ₄ F ₁₇ NO ₃ S	SO₃NH₄
70225-14-8	Perfluorooctane sulfonic acid, diethanolamine salt	C ₁₂ H ₁₂ F ₁₇ NO ₅ S	C ₄ H ₁₂ NO ₅ S
29457-72-5	Perfluorooctane sulfonic acid, lithium salt	C ₈ F ₁₇ SO ₃ Li	SO₃Li
2795-39-3	Perfluorooctane sulfonic acid, potassium salt	C ₈ F ₁₇ SO ₃ K	SO₃K

Ecology and Health work together on CAP development, collaborating with an external advisory committee to identify, characterize, and evaluate uses and releases of PBTs. CAPs recommend actions to protect human health or the environment, including actions to reduce and phase out uses and releases of the PBT, such as through the use of safer alternatives. Ecology and Health have completed five CAPs: mercury, lead, polyaromatic hydrocarbons, flame retardants, and polychlorinated biphenyls.

Chapter 173-334 WAC

The Children's Safe Products Act (CSPA) (Chapter 70.240¹⁴⁵ RCW) authorized Ecology and Health to develop a list of chemicals of high concern to children and a process for manufacturers to report on the presence of those chemicals in children's products. The CSPA Reporting Rule chemical list includes PFOS and its salts, and PFOA and related substances in the list of 85 chemicals of high concern to children (WAC 173-334-130¹⁴⁶). PFOS and its salts was included in the first list of reporting chemicals adopted in rule in 2011. PFOA and related substances was added to the reporting list in 2017.

Manufacturers are required to annually report the presence of PFOS or PFOA in children's products sold in Washington State. Annual reports include the manufacturer name, product category and component, chemical function, and concentration. Appendix 3: Sources and Uses,

¹⁴³ https://app.leg.wa.gov/RCW/default.aspx?cite=70.105

¹⁴⁴ https://apps.leg.wa.gov/wac/default.aspx?cite=173-333-310

¹⁴⁵ https://app.leg.wa.gov/RCW/default.aspx?cite=70.240

¹⁴⁶ https://apps.leg.wa.gov/wac/default.aspx?cite=173-334-130

<u>Section 3.3.1 PFAS in children's products</u>, provides a summary of the <u>PFOS and PFOA reported</u> in children's products.

Chapter 246-290 WAC

In December 2017, the Washington State Board of Health started rulemaking for Chapter 246-290¹⁴⁷ WAC Group A Public Water Supplies, to consider setting a standard for certain PFAS. The draft rule released for public comment in November 2019 included state action levels for five PFAS: PFOA, PFOS, perfluorononanoic acid (PFNA), perfluorohexane sulfonate (PFHxS), and perfluorobutane sulfonate (PFBS). It also included requirements for testing and reporting results to the Department of Health and for public notification, follow-up monitoring, and other actions when PFAS are detected. The rulemaking is intended to improve public health protection by setting a regulatory standard for PFAS chemicals in Washington for Group A public water systems. See Appendix 7: Health for additional information.

9.1.3 Executive orders

EO 04-01

In 2004, Governor Locke issued EO 04-01, requiring Ecology to establish, through rulemaking, specific criteria for use in identifying persistent, toxic chemicals that pose human health or environmental impacts in Washington state, and a clear process for developing chemical action plans to address those impacts (EO 04-01, 2004). Ecology developed Chapter 173-333 WAC in response to this EO.

The EO also ordered the Department of General Administration's Office of State Procurement (now called Department of Enterprise Services) to make available for purchase and use by all state agencies equipment, supplies, and other products that do not contain persistent, toxic chemicals unless there is no feasible alternative. State agencies were also ordered to reduce the use and purchase of products that contain PBT compounds (EO 04-01, 2004). As a result of the implementation of this EO, several state purchasing efforts have focused on reducing the presence of PBTs in state products. State purchasing preferences efforts related to PFAS have focused on PFAS-free carpet and food packaging (Simcich, 2020).

EO 18-01

In 2018, Governor Inslee issued EO 18-01, including the requirement that state agencies produce simple, clear, and targeted guidance that ensures agency compliance with environmentally preferable purchasing including opportunities for toxics reduction (EO 18-01, 2018).

¹⁴⁷ https://apps.leg.wa.gov/wac/default.aspx?cite=246-290

¹⁴⁸ https://apps.leg.wa.gov/wac/default.aspx?cite=173-333

9.2 Federal

Federal agencies that review PFAS include the Environmental Protection Agency (EPA), Food and Drug Administration (FDA), and Agency for Toxic Substances and Disease Registry (ATSDR). DOD is also required to track and reduce PFAS emissions resulting from its activities. The following EPA regulatory actions summarized in the Interstate Technology Regulatory Council (ITRC) Fact Sheet (ITRC, 2020a) are not repeated in this document (refer to Appendix 7: Health or ITRC for more discussion):

- Lifetime health advisory of 70 nanograms per liter (parts per trillion) under the Safe Drinking Water Act (SDWA).
- Unregulated contaminant monitoring rule data under the SDWA.
- PFAS reported at 14 sites under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA).

9.2.1 Environmental Protection Agency

PFAS are not currently regulated under the Resource Conservation and Recovery Act (RCRA), nor the Clean Air Act (CAA) or through numeric standards under the Clean Water Act (CWA). Nevertheless, EPA has initiated various PFAS-related activities as articulated in its 2019 PFAS Action Plan (EPA, 2019a, 2020a). The plan includes four main actions:

- Initiating steps to evaluate the need for a maximum contaminant level (MCL) for PFOA and PFOS.
- Beginning the steps to propose designating PFOA and PFOS as "hazardous substances" through available federal statutory mechanisms.
- Developing groundwater cleanup recommendations for PFOA and PFOS at contaminated sites.
- Developing toxicity values or oral reference doses for Hexafluoropropylene oxide dimer acid (HFPO-DA, also known as GenX) chemicals and PFBS.

The plan also identifies actions related to the development of new analytical methods, promulgating Significant New Use Rules (SNURs), and using enforcement actions to help manage PFAS risk.

Voluntary actions

PFOS voluntary phase-out

The 3M Company, the only U.S. manufacturer of PFOS, voluntarily phased out manufacture and use of PFOS in the U.S. in 2000 (EPA, 2000). 3M substantially completed the phase out of PFOA and PFOS in its products in the U.S. by 2002 (3M, 2020; Rutherford, 2019). These PFAS were further regulated under the Toxic Substances Control Act (TSCA) as described below.

PFOA stewardship program

EPA and eight major fluoropolymer and fluorotelomer manufacturers established a Voluntary PFOA Stewardship Program in 2006. Participants include Arkema, Asahi Glass, Ciba (now BASF), Clariant (now Archroma), Daikin, DuPont, 3M/Dyneon, and Solvay Solexis. Manufacturers agreed to reduce PFOA, precursor chemicals, and related higher homologue chemicals by 95% no later than 2010. The agreement committed companies to work toward eliminating PFOA emissions and products by 2015. All participating companies state that they met the PFOA Stewardship Program goals (EPA, 2018).

Interim recommendations for addressing contaminated groundwater

On December 19, 2019, EPA issued interim recommendations for addressing groundwater contaminated with PFOA and/or PFOS at sites being evaluated and addressed under federal cleanup programs, including programs for cleanup under CERCLA and RCRA (EPA, 2019b). The guidance recommends:

- Using a screening level of 40 parts per trillion (ppt) to determine if PFOA and/or PFOS is present at a site and may warrant further attention.
- Using EPA's PFOA and PFOS Lifetime Drinking Water Health Advisory level of 70 ppt as the preliminary remediation goal (PRG) for contaminated groundwater that is a current or potential source of drinking water, where no state or tribal MCL or other applicable or relevant and appropriate requirements (ARARs) are available or sufficiently protective.

In January 2020, the House of Representatives passed the PFAS Action Act (H.R. 535, S. 638), omnibus PFAS legislation. The Senate has not yet taken up companion legislation. The PFAS Action Act would require EPA to designate PFAS chemicals as hazardous substances under CERCLA within one year of enactment of the legislation. CERCLA imposes liability on responsible parties for response costs incurred in the cleanup of sites contaminated with hazardous substances. Designating the family of PFAS chemicals as "hazardous substances" would trigger cleanups of contaminated groundwater under CERCLA.

National Defense Authorization Act (NDAA) for Fiscal Year 2020

The NDAA includes PFAS-related mandates for both EPA and DOD. <u>Section 9.2.4</u> below addresses DOD requirements.

Effective January 1, 2020, Section 7321 of the NDAA includes 172 PFAS in the TRI, which are now subject to TRI reporting due July 2021 for calendar year 2020 data (EPA 2020b; EPA, 2020c). The NDAA establishes TRI manufacturing, processing, and otherwise use reporting thresholds of 100 pounds for each of the listed PFAS. EPA will revise the Emergency Planning and Community Right to Know Act (EPCRA) Section 313 list of reportable chemicals in the Code of Federal Regulations (CFR) to include the 172 PFAS added by the NDAA.

Additionally, the NDAA provides a framework for PFAS to be added automatically to the TRI list on January 1 of the year following certain EPA actions (NDAA Section 7321(c)). For example, the NDAA automatically adds a PFAS to the TRI list in response to the EPA finalizing a toxicity value for it. The NDAA also instructs EPA to consider certain other PFAS to the TRI list for possible addition (EPA, 2020c). Finally, EPA is directed to issue a "data rule" pursuant to section 8(a) of TSCA by January 1, 2023, requiring PFAS manufacturers to submit existing information concerning the environmental and health effects and estimates of the number of people exposed to each PFAS.

Toxic Substances Control Act

Significant New Use Rules

Under the provisions of TSCA, EPA has issued Significant New Use Rules (SNUR), to require notification regarding use, or restricting the use, of certain PFAS as follows (EPA, 2020d):

- March 11, 2002: SNUR requiring notification to EPA before any future manufacture (including import) of 13 PFAS chemicals specifically included in the voluntary phase out of PFOS by 3M that took place between 2000 and 2002—allowing the continuation of a few specifically limited uses.
- December 9, 2002: SNUR requiring notification to EPA before any future manufacture (including import) of 75 PFAS chemicals specifically included in the voluntary phase out of PFOS by 3M that took place between 2000 and 2002 allowing the continuation of a few specifically limited uses.
- October 9, 2007: SNUR addressing 183 PFAS chemicals believed to no longer be manufactured (including imported) or used in the United States.
- October 22, 2013: SNUR requiring companies to report all new uses (including import) of certain PFOA related chemicals as part of carpets.
- January 2015: EPA proposed a SNUR requiring persons who import a subset of
 articles containing long-chain perfluoroalkyl carboxylate and perfluoroalkyl sulfonate
 (LCPFAC) substances (as well as their salts and precursors) to notify EPA (Significant
 New Use Rule, 2015). The supplement would narrow the application of the LCPFAC
 SNUR to the subset of articles where the LCPFAC are part of a surface coating.
- March 3, 2020: EPA published its proposed rule in the Federal Register regarding supplementation of its 2015 PFAS SNUR Significant New Use Rule (EPA Supplemental Proposal, 2020). The supplement would require importers to notify EPA at least 90 days before beginning the import of such articles. Upon notification, EPA would begin evaluation of the conditions of use. Manufacturing (including import) or processing for the use would be prohibited until EPA has conducted a review of the notice, made an appropriate determination on the notice, and taken such actions as are required in association with that determination. This SNUR became final in June 2020 (EPA, 2020e).

New Chemical Review

Since 2000, EPA has also reviewed substitutes for PFOA and PFOS and other long-chain PFAS under TSCA Section 5, New Chemical Program Review. EPA reviews the new substances to identify whether the range of toxicity, fate, and bioaccumulation issues that have caused past concerns with PFAS may be present, as well as any issues that may be raised by new chemistries, in order to ensure that the new chemical may not present an unreasonable risk to health or the environment (EPA, 2020f). For those substances allowed to be manufactured or to enter the market, EPA issues TSCA Section 5(e) consent orders. These consent orders establish, for example, conditions on testing, release monitoring, and use specification.

9.2.2 Food and Drug Administration (FDA)

The FDA oversees the safety of food, drugs, and cosmetics under the Federal Food, Drug, and Cosmetic Act (U.S. Code Title 21, Chapter 9). Ingredients added to food and indirect food additives regulated under the Food, Drug and Cosmetics Act include substances that migrate from food packaging materials (FDA, 2017). Since the 1960s, PFAS have been used as grease-proofing agents for food packaging such as fast-food wrappers, to-go boxes, and pizza boxes.

In 2011, the FDA and several manufacturers reached a voluntary agreement to stop interstate distribution of products containing long-chain PFAS (FDA, 2017). In 2016, because the industry had discontinued the use, two PFAS were removed from the list of approved substances for oil and water repellants for paper and paperboard for use in contact with food (FDA, 2016).

9.2.3 Agency for Toxic Substances and Disease Registry (ATSDR)

The ATSDR, under the U.S. Department of Health and Human Services, is an advisory health agency, working with other federal agencies, state and local jurisdictions, tribes, and healthcare providers. Its focus includes preventing harmful exposures to hazardous substances using the best science, taking responsive public health actions, and providing trusted health information to prevent harmful exposures and diseases related to toxic substances. In June 2018, ATSDR released a draft Toxicological Profile for 15 PFAS (ATSDR 2018). See Appendix 7: Health for more information.

9.2.4 Department of Defense

The NDAA included PFAS-related provisions related to military use of products containing PFAS chemicals. The legislation included (State Energy & Environmental Impact Center, 2020):

- Phasing out the use of AFFF after October 1, 2024, except where it is used on ships, in emergency responses, and in limited testing and training circumstances.
- Establishing conditions for incineration of AFFF, and how wastes from such destruction are to be managed.
- Requiring blood testing for PFAS chemicals as part of routine physicals for military firefighters.

- Authorizing DOD to temporarily supply uncontaminated water or treated water to agricultural users whose irrigation water is contaminated with PFAS chemicals from military installations, as well as acquire property within the vicinity of an Air Force base that has shown signs of contamination due to activities at the base.
- Promoting cooperation on and monitoring of PFAS contamination in water supplies with local and state governments.
- Requiring EPA to take action on PFAS chemicals under TSCA, and promulgating a rule to require any manufacturer that has produced PFAS chemicals since 2011 to maintain records and report on the production of PFAS chemicals under TSCA.
- Banning the use of PFAS chemicals in packaging of military field food rations after October 1, 2021.

Various branches of DOD have also implemented many initiatives to address PFAS contamination issues. For example, the Department of the Navy is implementing a comprehensive strategy to manage and address PFAS in drinking water on and off Navy installations, cleanup of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) contaminated sites, and destruction of legacy AFFF (DON, 2017). The strategy includes:

- Controlling, removing, and disposing AFFF (DON, 2016). The Department of the Navy intends to remove, dispose, and replace legacy AFFF that contains PFOS or PFOA once environmentally suitable substitutes are certified to meet MIL-SPEC requirements.
- Identifying potential areas of concern from use of AFFF for fire and emergency response and test and training activities.
- Testing for PFOS and PFOA in Navy public water systems to determine if PFAS are known or suspected to have been released within one mile of the water source.

In 2019, DOD established a task force to address PFAS issues related to its installations nationwide (DOD, 2019). The task force has focused on three goals (DOD, 2020):

- Mitigating and eliminating the use of the current AFFF.
- Understanding the impacts of PFAS on human health.
- Fulfilling its cleanup responsibility related to PFAS.

Research is being funded by DOD in many areas—for example, through its environmental research programs, the Strategic Environmental Research and Development Program (SERDP), and the Environmental Security Technology Certification Program (ESTCP) (SERDP, 2020).

9.3 Other U.S. PFAS regulations and advisories

There are many PFAS regulations, advisories, and criteria, and they are changing rapidly. The ITRC fact sheet on regulations and guidance provides a summary. Supplemental tables to this fact sheet are updated to track changes in state and federal criteria and guidance:

ITRC PFAS regulations, guidance, and advisories fact sheets (ITRC, 2020a).

- ITRC PFAS Basis for PFOA and PFOS Values in Water established by ten U.S. states and Canada (ITRC, 2020b).
- Water and soil tables of regulations, guidance and advisories for PFAS by 23 U.S. states and 12 nations (ITRC, 2020c).

PFAS-related activities in other U.S. States have also been summarized by the Environmental Council of the States (ECOS) and the National Conference of State Legislatures (NCSL) (ECOS, 2020; NCSL, 2020). The EPA website also compiles <u>state PFAS resource information</u>. ¹⁴⁹ For example, in addition to developing standards or guidance for certain PFAS in in drinking water, groundwater, surface water, and soil, states have adopted requirements to report the presence of certain PFAS in consumer products, restrict certain PFAS in firefighting foam applications, and implement fish consumption advisories.

9.4 International

ITRC tables described in Section 9.3 above identify international standards or guidance for PFAS in drinking water, groundwater, surface water, and soil. The OECD also maintains a list of PFAS risk reduction strategies, including regulations, implemented by <u>countries around the world</u>.¹⁵⁰

9.5 Data gaps and recommendations

9.5.1 Data Gaps

There are no data gaps specific to Washington state regulatory activity on PFAS. Additional information collected as a result of CAP recommendations may clarify whether additional regulatory action is required to minimize PFAS risks.

9.5.2 Recommendations

The following is a summary of recommendations in this CAP which would require legislative or agency regulatory action.

- Recommendation 1.2: Health would complete rulemaking for PFAS in drinking water
- Recommendation 2.1: Ecology would develop cleanup levels for PFOA, PFOS, or other PFAS under MTCA.
- Recommendation 4.1: Ecology would require that domestic or industrial wastewater treatment plants monitor for PFAS.
- Recommendation 4.2: Ecology would update Chapter <u>173-350</u>¹⁵¹ WAC to require PFAS testing of leachate and landfill monitoring.

¹⁴⁹ https://www.epa.gov/pfas/us-state-resources-about-pfas

¹⁵⁰ https://www.oecd.org/chemicalsafety/portal-perfluorinated-chemicals/countryinformation/

¹⁵¹ https://apps.leg.wa.gov/wac/default.aspx?cite=173-350

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Chapter 70.95G RCW: Packages Containing Metals and Toxic Chemicals

Chapter 173-303 WAC: Dangerous Waste Regulations

Chapter 173-333 WAC: Persistent Bio accumulative Toxins

Chapter 173-334 WAC: Children's Safe Products Reporting Rule

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List of acronyms

General acronyms

Table 82. Acronyms found in the regulations appendix.

Acronym	Definition
ARARs	Applicable or relevant and appropriate requirements
ATSDR	United States Agency for Toxic Substances and Disease Registry
CAA	Clean Air Act
CAP	Chemical Action Plan
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
CFR	Code of Federal Regulations
CSPA	Children's Safe Products Act
CWA	Clean Water Act
DOD	United States Department of Defense
Ecology	Washington State Department of Ecology
ECOS	Environmental Council of the States
EO	Executive Order
EPA	United States Environmental Protection Agency
EPCRA	Emergency Planning and Community Right to Know Act
FCN	Food contact notification
FDA	United States Food and Drug Administration
Health	Washington State Department of Health
ITRC	Interstate Technology Regulatory Council
MCL	Maximum contaminant level
NCSL	National Conference of State Legislatures
NDAA	National Defense Authorization Act
OECD	Organisation for Economic Co-operation and Development
PBT	Persistent Bioaccumulative Toxic
ppm	Parts per million
ppt	Parts per trillion
PRG	Preliminary remediation goal
RCRA	Resource Conservation and Recovery Act
RCW	Revised Code of Washington
SDWA	Safe Drinking Water Act
SNUR	Signficant new use rule
TRI	Toxics Release Inventory
TSCA	Toxic Substances Control Act
U.S.	United States of America
WAC	Washington Administrative Code

Chemical Names

Table 83. Chemical name acronyms found in the regulations appendix, excluding the acronyms listed in the table above.

Acronym	Chemical Name
HFPO-DA (GenX)	Hexafluoropropylene oxide dimer acid
LCPFAC	Long-chain perfluoroalkyl carboxylate and perfluoroalkyl sulfonate
PFAS	Per and polyfluorinated alkyl substances
PFBS	Perfluorobutane sulfonate
PFHxS	Perfluorohexane sulfonate
PFNA	Perfluorononanoic acid
PFOS	Perfluorooctane sulfonic acid
PFOA	Perfluorooctanoic acid

Appendix 10: Economic Analysis

10.0 Overview

10.0.1 Findings

Information about the costs of actions to reduce per- and polyfluoroalkyl substance (PFAS) exposure is limited, due to limited understanding about the extent and characteristics of PFAS contamination statewide, and the variability and often site-specific nature of actions considered. Some PFAS contamination is known and well characterized, while overall statewide contamination and associated cleanup actions are likely to be highly variable and require ongoing sampling and testing to fully characterize.

Moreover, the current status of mitigating ongoing impacts of contamination offers limited insight into overall costs of complete cleanup. Similarly, extent and understanding of the human health impacts of PFAS contamination are variable, and testing depends on factors ranging from individual test costs and bulk discounts, to affected population and duration of exposure before mitigation measures are taken. Public involvement and education can take many forms, and depends on the attributes and needs of the affected population. Finally, understanding of known existing sources and potential actions to reduce exposure suffers from limited information that clearly identifies products containing PFAS (or what kinds of PFAS they contain).

In light of these limitations, we have sought to provide what information is available. For some actions, this allowed for full quantification of some potential costs. For others, only illustrative examples were possible. Overall, potential costs of PFAS remediation and exposure mitigation are likely very significant, and beg the question of whether cleanup or ongoing mitigation is most feasible. Characterizing the extent of the problem, such as testing landfill leachate, is less costly, but does not include the costs of any necessary cleanup or other action identified as a result.

The greatest uncertainty in costs surrounds product purchasing, due to limited or absent ability to identify whether current products contain PFAS, and what PFAS-free alternatives are available. Absent testing, we were not able to identify which paints in state contracts contain PFAS. To the extent that shifting to consumer products labeled as more environmentally friendly, such as cleaning products and floor coverings, coincides with a shift away from PFAS-containing products, some products may double in price. Changing purchasing priorities for floor covering purchases in state and municipal contracts could increase costs by millions of dollars.

10.0.2 Introduction

The Persistent Bioaccumulative Toxins rule (Washington Administrative Code (WAC) <u>173-333</u>(3)¹⁵²) requires that, as part of any Chemical Action Plan (CAP), the Washington State Departments of Ecology (Ecology) and Health (Health) should "identify costs of implementing the recommendations. This may include a qualitative and/or quantitative analysis of the probable benefits and costs of the CAP." This appendix is intended to meet these requirements. Here, we identify and estimate, to the extent possible, the costs of implementing the recommendations of the CAP. Where possible, we identify the resulting costs and benefits of implementing the recommendations.

Cost estimates in this appendix include external costs borne by parties other than Ecology or Health, which would not be funded through agency budget requests. Recommendations presented in the CAP identify estimates of agency implementation costs that could potentially be funded through additional budget allocations.

Our analyses compare potential actions to the current situation where no action is taken. This is the baseline, and it reflects legal requirements that exist regardless of whether proposed actions are taken (e.g., state dangerous waste regulations). The baseline can also include actions that are already planned or occurring (e.g., ongoing removal, disposal, and replacement of aqueous film forming foam (AFFF) at military facilities).

Because of developing knowledge about PFAS—from scientific research, to testing and identification, to understanding the extent of use and contamination—our ability to fully quantify implementation costs and resulting costs and benefits is limited. Where full quantification (total costs or benefits) was not possible, we have included what partial quantification was possible, such as unit costs, costs per event, or costs per firm. Where no quantification was possible, we have included qualitative discussion of impacts.

Recommended actions analyzed

- Action 1.1: Identify funding for PFAS drinking water mitigation.
- Action 1.2: Technical support for site characterization, source investigation, and mitigation at contaminated sites.
 - Ecology will continue to collaborate with and provide technical assistance to involved parties at PFAS contamination sites in the state. These efforts will help to better understand the sources, composition, and distribution of PFAS contamination in soil and water. This work will also inform evaluation of appropriate cleanup actions and their costs.
- Action 1.3: Support biomonitoring to support impacted residents and help answer important health questions.
- Action 2.2: Partner with local organizations in communities with contaminated water or contaminated sites.

¹⁵² https://apps.leg.wa.gov/wac/default.aspx?cite=173-333

- Action 2.3: Work to prevent PFAS releases from AFFF use and manufacturing processes.
 - Ensure that industrial use of AFFF provides for containment procedures along with collection of AFFF and contaminated soil or sediment for proper designation and disposal. Costs to industrial users to collect and dispose of released PFAScontaining AFFF include plan development, employee training, methods for containment, and disposal of waste.
- Action 3.1: Reduce PFAS exposure from carpet and rugs, water and stain resistance treatments, and leather and textile furnishings.
 - Implement a purchasing preference policy for PFAS-free carpet. Work with vendors on the flooring contract to offer PFAS-free carpet on all state master contracts and all agency contracts.
- Action 3.3: Implement other reduction actions for PFAS in products.
 - Establish a purchasing preference policy for PFAS-free products. Work with vendors to offer PFAS-free textiles, furniture, and paints. Apply this policy to all state master contracts and all agency contracts.
- Action 4.2: Evaluate PFAS in landfill leachate.
 - For this action, Ecology would consider adding requirements for PFAS testing, monitoring, and reporting for landfill leachate emissions.

Additional options analyzed

In developing this CAP, Ecology considered additional actions that were ultimately not recommended. They include:

- Requiring municipal wastewater treatment plants (WWTPs) to test influent and effluent.
- Recommending that the Legislature require alternatives assessments for specific products.

10.1 Costs of recommended actions

Action 1.1 Identify funding for PFAS drinking water mitigation

When concentrations of PFAS above the health advisory are detected in a drinking water supply, mitigation is required to bring drinking water quality back into compliance. Mitigation activities incur unanticipated costs to water utilities in a variety of ways, including but not limited to:

- Site-specific investigations to determine the location, extent, and source of PFAS contamination.
- Information campaigns to notify affected water users.
- Identification and development of appropriate technologies to reduce or remove contaminants in order to meet drinking water quality standards.
- Implementation and maintenance of such measures in the short and long term.

In the short term, clean water may need to be supplied by purchase of drinking water from a neighboring system or distribution of bottled water to customers. Longer term mitigation measures focus on finding and accessing more permanent clean water sources or installing additional equipment, such as an expensive filtration system to remove PFAS, to treat water to meet applicable standards (see Appendix 4: Fate and Transport, Section 4.4). Without funding to defray these costs, public water systems and their ratepayers must absorb the costs of response.

Total site specific or statewide costs for PFAS mitigation in drinking water will not be known until further water testing defines the scope of the problem in Washington state. At this time, a few illustrative examples of drinking water mitigation are available. Some examples do not separate the investigation costs.

- The City of Issaquah spent \$600,000 to install a treatment system on one PFAS-contaminated city well. Filter maintenance and monitoring also require ongoing expenditures of \$56,000 per year (York, 2020).
- The Naval Air Station Whidbey Island spent \$1.37 million, to date, for investigation and mitigation efforts, including providing bottled water and filters to impacted residences. Adding an activated carbon treatment system to the Town of Coupeville's water system is estimated to cost \$1.5 million (Washington State Department of Health, 2019).
- In response to PFAAs detection in April 2017, the public water system of Airway Heights shut down their contaminated wells and used an emergency intertie with the City of Spokane water system to flush their system with clean water. Flushing included draining reservoirs and water towers. During the flushing, Fairchild AFB provided bottled water to water customers. Airway Heights has since added another connection to the City of Spokane to supply drinking water while they pursue treatment options for the contaminated wells. This water purchase could cost over \$687,000 in the first year, for over 439 million gallons of drinking water, for which the Air Force has agreed to pay the city (Sokol, 2017).
- Treatment of drinking water in Lakewood, using activated carbon filtration, is estimated to cost \$5.2 million in initial capital costs, with ongoing operating and maintenance costs of \$96,000 per year (Health, 2019).
- At Joint Base Lewis McChord, McChord Field System, treatment of water from three wells, using activated carbon filtration, is estimated to cost \$10.3 million in initial capital costs, with ongoing operating and maintenance costs of \$830,000 per year (Health, 2019).

These costs are in line with similar drinking water remediation activities in other states. For example, Moose Creek, Alaska, has already incurred \$3.5 million for a granular activated carbon system to treat groundwater contaminated by AFFF releases at Eilson Airforce Base (Gardner,

2019). The Pentagon will pay \$30 million to extend the municipal water system from a neighboring community to Moose Creek (Ellis, 2020; DeFazio & Tynan, 2019).

Action 1.2: Technical support for site characterization, source investigation, and mitigation at contaminated sites

Parties that released PFAS into the environment are responsible for cleaning it up and may also need to reimburse the water purveyor or Ecology for cleanup or exposure-mitigation activities. As part of the cleanup process, Ecology establishes cleanup levels, which are concentrations of hazardous substances in the environment that are considered sufficiently "protective of human health and the environment under specified exposure conditions."

Action 1.2 includes the recommendation that Ecology collaborate with and provide technical support to involved parties at PFAS contamination sites in the state. These efforts will help to better understand the sources, composition, and distribution of PFAS contamination in soil and water. Evaluation of appropriate cleanup actions and their costs will be informed by this work.

The costs of developing and evaluating methods for investigating and cleaning up PFAS contamination are currently difficult to estimate due to significant uncertainties in our understanding regarding:

- How most PFAS affect people, animals, and plants, and in what concentrations.
- How to best measure the types and amounts of PFAS in the environment.
- How PFAS move through the environment and change over time.
- How to best clean up environmental PFAS contamination, including consideration of protectiveness, feasibility, and cost.

Ongoing research continues to expand our knowledge base on these issues and may significantly alter the way that Ecology's Toxics Cleanup Program (TCP) approaches cleanup in the future. TCP is currently working with the City of Issaquah and the Eastside Fire District to identify possible sources of the PFAS contamination affecting the city's drinking water. As of the end of 2018, Ecology has contributed \$330,000 to this investigation (Ecology, 2019a).

Environmental PFAS contamination in Washington and examples of cost of interim cleanup actions in Washington and elsewhere

Known sites

Known areas with PFAS contamination in drinking water in Washington include:

- Issaquah (Eastside Fire and Rescue).
- Joint Base Lewis-McChord.
- Naval Air Station Whidbey Island.
- Naval Base Kitsap-Bangor.
- Fairchild Air Force Base, including Airway Heights.
- Cities of Lakewood, DuPont, Tacoma, and Parkland.

In some of these areas, concentrations of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) in groundwater used for drinking exceed the Environmental Protection Agency (EPA) health advisory level (see <u>Appendix 7: Health, Section 7.4, Known areas of PFAS contamination</u>). The primary source of contamination at all of these areas is believed to be releases of legacy PFAS-based firefighting foam (AFFF).

Investigation and exposure reduction actions in and around the military bases are being conducted by the Department of Defense (DOD). The City of Issaquah has installed a filtration system to reduce PFAS concentration in its finished drinking water. As noted above, TCP is currently working with the City of Issaquah and the Eastside Fire District to identify possible sources of the PFAS groundwater contamination affecting the city's drinking water. Detections of PFOA and PFOS at levels above the EPA health advisory level resulted in removing wells from operation.

Potential sites

In addition to fire suppression, PFAS have many industrial uses (see <u>Appendix 3: Sources and Uses</u>). Future investigations may identify releases at or near these types of facilities:

- Tanneries.
- Shoe manufacturing facilities.
- Textile treatment facilities.
- Plastics manufacturing facilities.
- Metal plating facilities.

Also, AFFF may have been released at many locations to extinguish fires, particularly petroleum fires. If PFAS releases are suspected at these types of facilities, testing should be conducted to evaluate the presence of environmental contamination.

Remediation costs are difficult to estimate and are expected to depend on variables such as:

- Extent and attributes of contamination.
- Affected populations.
- Geographic location and site attributes.
- Amount of contamination in soil versus groundwater.

Example costs

Moreover, the total cost of remediation for PFAS-contaminated groundwater is not yet known. Ecology is not aware of any completed cleanups of PFAS contamination. Instead, we are including illustrative costs of interim actions and options to scale them to full cleanup cost. Costs already incurred to address contaminated drinking water in Washington (noted above) are illustrative of sites undergoing mitigation and investigation, but no site has yet completed remediation. The cost to remediate a site will depend on site-specific factors such as the number of sources of contamination contributing to a site, the specific contaminants present,

and how the contamination migrates from the sites. In the case of complex manufacturing contaminated sites, costs have escalated rapidly, for example:

- It was reported that Hoosick Falls, New York budgeted \$10 million for temporary municipal and private residential water filtration systems and investigation into an alternate drinking water source (Safer States, 2019). The capital, operation, and maintenance costs of alternatives to address the drinking water contamination on a long term basis ranged from \$6 to \$48 million dollars (ERM & CHA, 2019). However, full remediation of the complexity of contamination from multiple sources will exceed these preliminary costs (New York Department of Environmental Conservation 2020).
- Statewide, Minnesota has estimated PFAS cleanup costs resulting from PFAS manufacturing to exceed \$1 billion (Bjorjus, 2020; Minnesota 3M PFC Settlement, 2020a, 2020b).

Potential total cost of a cleanup in Washington

The Model Toxics Control Act (MTCA) Biennial Report provides data on remediation costs by stage for addressing contaminated sites (Ecology, 2016, 2018b, 2019b). These are costs associated with characterizing the site, mitigating the impact if drinking water is affected, and cleaning up contaminated soil and groundwater. The table below summarizes how costs are typically distributed between site investigation and cleanup phases.

Table 84.	Percentage of	cost k	oy remedial	activity	phase.

Remedial activity category	2013 – 15 biennium	2015 – 17 biennium	2017 – 19 biennium
Cleanup	67%	72%	70%
Investigations	26%	16%	28%
Other	7%	12%	2%

Scaling the cost examples (related primarily to investigation and pre-cleanup activities using the MTCA remediation ratios with highest weighted cleanup) could result in overall remediation costs (excluding interim mitigation costs) of between \$5.3 million and \$62.8 million. Additionally, spending on interim solutions such as filtering or alternative sources of drinking water could result in ten-year costs of \$6.5 million to \$10 million.

Cleanup or mitigation?

Additional complexity in estimating potential costs comes from the developing nature of PFAS drinking water cleanups. Additional unknowns resulting in uncertainty include:

- The degree of remedial and preventative soil cleanup needed to reduce ongoing and future groundwater contamination.
- The best remediation method(s) and their cost-effectiveness in balance with mitigation efforts. This includes comparisons between actions such as:
 - Only treating drinking water prior to consumption.

- o Pumping, treating, and returning water to the aquifer.
- o Treatment at a single point versus multiple wells.
- Treatment limiting the scope or spread of existing contamination.
- Multiple types of PFAS might be contaminating groundwater but do not currently have viable test methods.
- Size of populations consuming contaminated groundwater.
- Liability to other property owners, water purveyors, or consumers.
- Developing knowledge in PFAS toxicity to humans and the environment.
- Ability of liable parties to cover cleanup costs.
 - Parties such as small firefighting districts, that provide crucial services, may not be able to fund remedial actions on the same time scale or size as larger entities.
 Extending timeframes could increase interim mitigation costs and potential scope of contamination.

Action 1.3 Support funding for biomonitoring to support impacted residents and help answer important health questions

This action could involve finding competitive grant funding sources to offer subsidized biomonitoring for residents in areas impacted by PFAS-contaminated drinking water. Biomonitoring would let people know their exposure level relative to national averages and relative to other populations with elevated PFAS exposure. This information could help residents connect to health information that becomes available in the future.

Testing costs vary by number of analytes and whether they include drawing blood (Wagner & Bagenstose, 2017):

- Tests for PFOA or PFOS that do not include blood draws cost about \$300 per test.
- Tests for 13 PFAS analytes cost between \$450 and \$500.
- Tests including drawing blood or a blood sampling and mailing kit cost between \$528 and \$797.

Testing performed by a centralized company or agency may receive different rates (e.g., bulk rates) but incur additional administrative costs, resulting in different and variable average perperson testing costs (Bagenstose, 2018):

- Serum testing of over 69,000 people in the mid-Ohio Valley for PFAS, for \$70 million, averaged approximately \$1,000 per person tested.
- The state of New York tested 3,000 people in Hoosick, New York for \$3 million, averaging \$1,000 per person tested.
- The Centers for Disease Control and Prevention (CDC) and state of New Hampshire tested 1,600 people in Pease, New Hampshire for nearly \$340,000, averaging \$211 per person tested.
- Under a CDC grant to the state of Pennsylvania, the state tested 250 people for \$175,000, averaging \$700 per person tested.

Action 2.2: Partner with local organizations in community outreach and support community involvement

This action involves providing funding to local organizations to engage communities affected by PFAS contamination. This could involve example activities including (as demonstrated in other states' public involvement):

- Educational materials.
- Rapid response information.

Ecology's Public Participation Grant (PPG) program funds activities similar to what is included in this recommendation. The PPG program, however, applies to a broad set of activity types, and is already limited in funds it provides for such activities, meaning the program is not likely to have funding available for PFAS activities. Additional funding under this action would be directed specifically to PFAS-related impacts to communities, rather than competing with (and potentially displacing) existing PPG grantees.

A component of the PPG program that funds information for communities impacted by contaminated sites is the Contaminated Site Project category of grants. An example of this type of grant project is the multi-component Futurewise program. This program is for communities affected by contamination in Algona, stemming from past Boeing manufacturing activities (Ecology, 2013). The two-year, \$120,000 grant covered a large scope of activities, but included \$25,000 for educational materials specifically:

- Printed and electronic education and outreach materials.
- Display booth.
- Health fair.
- Translated materials.

This action is likely to cover the types of activities listed above, as well as potential engagement of disadvantaged populations in problem solving and collective action. Provision of these educational materials would be helpful for communities that are at elevated risk of PFAS exposure. The degree and types of activity covered would depend on the funds available, as well as the number and types of projects requesting funding.

Examples of larger types of community-based action

While not envisioned as part of this recommended action, we note there are additional types of action taken through community-based grant programs in other states in response to PFAS drinking water contamination and exposure.

- Population PFAS testing.
- Public meetings.
- Health guidance and information provision.
- Information fairs.
- Involvement and representation in public processes for PFAS regulation.

PFAS projects developed under funding from this action are envisioned to be significantly smaller in scope, primarily intending to avoid displacing projects already using the PPG program. The \$120,000 grant agreement for the Futurewise project covered (between October 1, 2013 and June 30, 2015) the activities below. Depending on the types of community engagement that would occur, and based on circumstances and grant applications, this action could include similar additional components.

- Administration (\$13,625)
 - Tracking of spending and objectives.
 - Evaluation and reporting.
 - Final project report.
- Public events and outreach (\$81,230)
 - o Immediate resident audience
 - One-to-one outreach.
 - o House parties with invited speakers such as health consultants.
 - Healthy home visits.
 - Outreach specialist.
 - o Translation to relevant languages.
 - Greater Algona audience
 - o Attending annual community-wide public events.
 - Creating exhibits, games, and interactive activities for education.
 - Holding a health fair.
 - Fact sheets: contamination, cleanup, participation in the Ecology public process.
 - o Community meeting with speakers and cleanup updates.
 - Business audience
 - One-to-one outreach identifying questions and concerns.
 - Business-specific outreach materials.
 - Two outreach events with speakers and updates.
 - Student audience
 - Outreach to teachers and other adult youth leaders about cleanup and groundwater science.
 - Engaging youth volunteers in youth education and involvement.
- Education tools (\$25,145)
 - Printed and electronic education and outreach materials:
 - PowerPoint presentations.
 - Factsheets.
 - Display materials for event tables.
 - Meeting agendas.
 - Evaluation tools.
 - Display booth.
 - Health fair.
 - Translated materials.

Action 2.3: Work to prevent PFAS releases from firefighting foam use and manufacturing processes

One of the recommended actions is to ensure that industrial use of AFFF provides for containment procedures along with collection of AFFF and contaminated soil or sediment for proper designation and disposal. Costs to industrial users to collect and dispose of released PFAS-containing AFFF include plan development, employee training, methods for containment, and disposal of waste.

For this action, Ecology would need to inform users of AFFF of the requirements and provide guidance on how to comply with them. Then users of AFFF would need to collect, treat, and properly dispose of PFAS-containing waste from AFFF use.

To prevent discharge of AFFF to the environment (or minimize it), industry would need to:

- Develop a plan for compliance.
- Purchase and carry compliance equipment.
- Collect runoff containing PFAS.
- Treat and dispose of runoff.

Runoff collection plan

We assumed development of a runoff collection plan would require 80 hours of technical, administrative, and managerial staff time at AFFF-using facilities. Assuming third-party median environmental engineer hourly wages of \$46.89, loaded with additional overhead costs to reflect higher consultant prices (if external consultants are used—using exclusively internal staff, if possible, could reduce costs), and updated for inflation, the loaded hourly wage for this work would be \$89.77 (Ecology, 2018a; U.S. Bureau of Labor Statistics, 2017, 2019). The cost per facility for this task would be \$7,182. Potential additional costs would be incurred if additional internal staff time is required, such as for ongoing interaction with consultants during plan development.

Training

We further assumed four personnel would need to be trained in the runoff collection plan facilities using AFFF. Using the median firefighting wage of \$35.28 per hour, updated for inflation and overhead to \$68.51 per hour, and an estimate of 320 hours (80 hours each for four trainees) this cost becomes approximately \$22,000 per facility (Ecology, 2018a; U.S. Bureau of Labor Statistics, 2017, 2019).

Training costs are, of course, more nuanced. Training materials may need to be developed, as well as labels and signage reminding firefighters of best practices. These materials may be generated by one party, and then shared with, or sold to, other facilities. Highly location-specific training needs, as well as staff turnover, may result in additional materials, instruction, and hours of employee time.

Containment, collection, and disposal

Effectively containing, collecting, and disposing of AFFF-contaminated runoff would likely entail measures such as:

- Portable booms, berms, and drain blocks.
- Pumps, hoses, and tanks.
- Potential pre-disposal treatment.
- Disposal of untreated or treated runoff.
- Disposal of treatment byproducts such as filters, sorbents, or solidifiers.

While these measures could be taken individually, facilities are likely to hire a specialist in wastewater and runoff management to properly manage PFAS-containing runoff (ESTCP, 2003). While volumes of existing product are identifiable to some extent, actual volumes of runoff captured, treated, and disposed of will vary by site and firefighting activities (e.g., how much water is used, site characteristics). We could not, therefore, estimate total costs. We have, however, identified unit costs associated with elements of capture, treatment, and disposal.

Portable booms, berms, and drain blocks

We evaluated response equipment costs based on Grainger (2019). Depending on the style and length of boom, the per-foot cost ranges between \$10 and \$45, with a median price of \$26 per foot. At typical purchasing lengths of up to 100 feet, this cost would be \$260 per boom, at the median.

Similarly, depending on the style and length of berm, the per-foot cost ranges between \$22 and \$81, with a median price of \$34 per foot. At typical purchasing lengths of up to 12 feet, this cost would be up to \$408 per berm, at the median.

Drain blocks and seals vary in size and quality, but range between \$110 and \$581, with a median price of \$238.

These prices do not include additional labor required for set-up during AFFF use, which will vary by site and firefighting characteristics.

Pumps, hoses, and tanks

Depending on the size needed and location of use, purchasing a portable trailer pump and tank (rather than hiring a contractor) could cost tens of thousands of dollars. A smaller-volume (50 gallon) portable pump and tank could cost up to \$5,000 (Edson, 2019; JME Ellsworth, 2019).

Treatment and disposal

The cost of on-site treatment of AFFF-contaminated runoff water varies significantly by technology and type of product disposed. A DOD study comparing multiple treatment technologies and disposal products at large facilities indicates the following—with costs updated to 2018 values (U.S. Bureau of Labor Statistics, 2019):

- Existing off-site treatment and disposal costs ranged between \$0.14/pound (lb) and \$0.44/lb, with an average cost of \$0.25/lb, if waste is approximately the density of water.
- Rental generators to run treatment would cost \$137/day with anticipated work days lasting eight hours to treat 24,000 48,000 thousand gallons of wastewater.
- A potential treatment technology could incur capital costs of \$236,000 to \$306,000, but result in 94% reductions in disposal costs by reducing water waste to sludge.

Existing stored product

The exact makeup of most AFFF products is confidential business information (CBI), making a detailed economic analysis of replacement products impossible. However, technical documents from the United Nations (UN) Stockholm Convention (2012) provide a general assessment on replacing AFFF that contain PFOS, which may translate to PFAS replacement activities. Using this assessment as a guide, costs would likely be incurred during PFAS AFFF replacement from:

- Destruction or storage of the retired chemicals.
- Cleanup of impacts areas.
- Replacement of or upgrades to existing equipment.
- Potential changes in operations.

In lieu of ongoing use of AFFF, facilities have options of either disposal or other removal of the product, and replacement with an alternative PFAS-free product. Washington state does not prohibit removal of AFFF product from the state, but does encourage proper disposal at a permitted treatment, storage, and disposal facility.

Large scale replacement costs have included \$6.2 million for replacement of AFFF at 180 U.S. Air Force facilities. For context, we note that the U.S. Air Force budget is in the hundreds of billions of dollars. More remote facilities incur higher transportation costs to airlift in new AFFF (U.S. Air Force, 2017). A recent DOD contract opportunity offered \$5 million for removal, destruction, and disposal of AFFF from three geographic regions of facilities, but without replacement (DOD, 2018). The existence of these ongoing replacement and disposal activities, however, indicates that some of the costs of this action are part of the baseline (happening regardless of recommendations).

As identified in Appendix 9: Regulations, Section 9.1.1 Washington state laws, AFFF qualifies as a state-designated dangerous waste in Washington, and its disposal must therefore be managed in compliance with WAC 173-303¹⁵³ Dangerous Waste Regulations. Small quantity generators may transport their own hazardous waste, whereas medium and large quantity generators must hire a hazardous waste contractor. Small quantity generators (generating less than 220 lbs of dangerous waste in any month) may take waste to a facility licensed to accept hazardous waste. These facilities vary by county and charge their own set of fees, ranging up to

¹⁵³ https://apps.leg.wa.gov/wac/default.aspx?cite=173-303

\$100 per ton equivalent. Equipment used in deploying AFFF may need to be retrofitted or replaced. This largely depends on the change in viscosity of the replacement product.

Medium and large quantity generators must dispose of their AFFF at permitted Transfer, Storage, and Disposal facilities, via a licensed hazardous waste contractor. This can cost in the tens of thousands of dollars, depending on the quantities of product being disposed of, and hauling distance.

There may also be costs associated with changing existing operations due to differing requirements of new AFFF products. The Stockholm Convention alternatives documents suggested that some AFFF users reported no change in operational costs when retiring PFOS chemicals, while others incurred higher costs (United Nations Environmental Programme (UNEP), 2012).

Action 3.1: Reduce PFAS exposure from carpet and carpet care products

One of the recommended actions is to implement a state agency purchasing preference policy for PFAS-free carpet.

Recent and previous research has shown carpet to be a repository for pollutants and that indoor air quality declines when carpeted areas are disturbed (Becher et al, 2018). According to the Carpet and Rug Institute (CRI), carpet accounts for 48% of the U.S. flooring market (CRI, 2020). PFAS, largely used for stain repellent in carpet, were worth close to \$1 billion worldwide in 2006 for this use (Renner, 2006). Two North American studies frequently detected PFAS particles in significant concentrations in vacuum cleaner bags. The studies found a significant correlation between the presence of PFAS and the age of the house and floor covering type (Fromme et al, 2008).

For this action, Ecology would develop a purchasing preference policy (for purchases by the State of Washington) for carpet. A purchasing preference policy would not ban state purchases of PFAS-containing carpet. Instead, the policy could give points to state contract bidders, when they declare (and provide supporting data) that their carpet does not contain PFAS. These points would be part of the bid scoring process. This would create incentive for bidders to provide PFAS-free carpet to state agencies and municipalities that use the state contracting process.

We estimated the difference in costs for state carpet purchases that contain PFAS versus PFAS-free. We were able to estimate annual quantities of carpet purchased under the state contract, based on 2018 invoiced costs per quarter from Washington State Department of Enterprise Services (DES) (Table 85). Multiple types of facilities purchase carpet under the state contract:

- State agencies
- Cities
- Counties

- Fire districts
- Higher education establishments
- Libraries
- Nonprofits
- Ports
- School districts

Total invoiced amounts were identified by quarter, and are summarized in Table 85. Only one year of data was available for this analysis, so it may not be representative of all other years.

Table 85. Carpet invoices by group and quarter, 2018 (DES, 2018a).

Group	Q1	Q2	Q3	Q4	Annual
State	\$822,883	\$2,545,349	\$332,531	\$403,054	\$4,103,817
Cities	\$267,691	\$451,481	\$334,562	\$373,953	\$1,427,687
Counties	\$501,283	\$338,855	\$194,010	\$415,423	\$1,449,571
Fire Districts	\$57,094	\$101,975	\$186,309	\$60,371	\$405,749
Higher Education	\$113,484	\$702,653	\$733,069	\$345,657	\$1,894,863
Libraries	\$33,325	\$173	\$34,605	\$159,470	\$227,573
Nonprofits	\$1,944	\$1,431	\$0	\$1,771	\$5,146
Ports	\$51,053	\$10,178	\$5,534	\$6,483	\$73,248
School Districts	\$193,700	\$543,974	\$3,751,886	\$942,441	\$5,432,001
ALL GROUPS	\$2,042,457	\$4,696,069	\$5,572,506	\$2,708,623	\$15,019,655

While these invoiced amounts tell us the total cost of carpet purchases, they do not tell us the types of carpet purchased, or the square yards (quantity) purchased. We therefore made various combinations of assumptions to develop a range of square yardage potentially reflected in these invoices.

Using the multiple carpet options available under the state contract—which includes PFAS-containing and PFAS-free options or options that use alternative technologies to "permanently or inherently" make their carpet stain-resistant—we identified a price difference between carpets with and without topically applied treatment (see Table 86). Approximately 35.7% of offered carpet products have topically applied stain resistance, whereas 64.3% have permanent or inherent stain resistance (DES, 2018b).

Table 86. Price per square yard of carpet.

Summary statistic	Topically applied stain resistance	Permanent or inherent stain resistance
Minimum	\$6.74	\$11.43
Median	\$17.06	\$21.96
Maximum	\$37.04	\$45.56
Average	\$17.09	\$23.51

Uncertainty arises from whether these various types of carpet—many of which contain proprietary chemicals or fibers—do, in fact, contain PFAS. Topical treatments may or may not contain PFAS. Similarly, fiber stain-resistance technologies that are described as non-degrading and "permanent," even when subjected to heat and cleaning in addition to normal wear, may potentially contain PFAS. Without comprehensive reporting of testing or knowledge of upstream production processes and treatment chemical or technology contents, these distinctions are not possible to make with reasonable certainty.

However, because the average price of inherently stain-resistant carpet was significantly higher than the price of carpet with topically applied stain-resistance treatment, and for simplified estimation, we assumed in this estimation that carpet with topically applied treatment contained PFAS, while those with inherent stain-resistance did not. This resulted in carpet containing PFAS costing less than the PFAS-free alternatives. We have also included a worst-case cost scenario, in which all carpet currently being purchased contains PFAS, and would be incentivized under this alternative to be replaced with PFAS-free carpet, by scoring PFAS-free carpet contractors higher than those not offering documented PFAS-free carpet.

If current carpet purchases are in line with the proportions of products offered, and based on the average topically applied price of \$17.09/square yard (sy), the total invoiced costs reflect 725,000 sy of carpet. In this case, 35.7% of purchases would be incentivized to change their purchasing under this action. The total cost increase would then be 35.7% of 725,000 sy, purchased at the difference between the average permanently stain-resistant price of \$23.51 and the topically applied price of \$17.09. This total cost increase statewide would be \$1.7 million per year.

In a worst-case cost scenario, however, all current carpet purchases contain PFAS in some form or another, potentially because of least-cost purchasing preferences. In this case, based on the average topically applied price of \$17.09/sy, the total invoiced costs reflect 879,000 sy of carpet. In this case, all buyers would be incentivized to change their purchasing under this action. The total cost increase would then be 879,000 sy purchased at the difference between the average permanently stain-resistant price of \$23.51 and the topically applied price of \$17.09. This total cost increase statewide would be \$5.6 million per year.

The table below summarizes how the cost increases under the two scenarios above would be distributed across various types of buyers.

Table 87. Total cost increase by group and scope of change, annual.

Group	Cost increase if 35.7% switch	Cost increase if 100% switch
State	\$454,012	\$1,542,781
Cities	\$157,947	\$536,722
Counties	\$160,368	\$544,949
Fire districts	\$44,889	\$152,537
Higher education	\$209,632	\$712,351

Group	Cost increase if 35.7% switch	Cost increase if 100% switch
Libraries	\$25,177	\$85,553
Nonprofits	\$569	\$1,935
Ports	\$8,104	\$27,537
School districts	\$600,951	\$2,042,096
ALL GROUPS	\$1,661,648	\$5,646,461

We note there are also types of carpet that are not included in existing state contract rates. The prices above reflect various types of backed nylon carpet, either in tiles or broadloom. They do not include polyester carpets that do not need stain resistance added. They also exclude potential emerging new technologies in stain-resistance using alternative fibers designed to be more hard-wearing, such as nylon carpet (see, for example, Dupont's "Sorona Fiber" and Invista's "non-fluorinated Duratech") (Antron, 2016; Dupont, 2020). We could not identify the relative cost of these options.

These estimates also assume that the same quantities of carpet will be purchased. Facing higher unit prices, buyers may substitute away from carpet and choose other floor coverings that are PFAS-free instead of PFAS-free carpet. This could lower overall costs, though we note that a specific floor covering may be chosen for any number of purposes, including, but not limited to, aesthetics, safety, acoustics, or other qualities.

Action 3.2: Implement reduction actions for PFAS in priority consumer products

One of the recommended actions is that a state agency purchasing preference policy be established for other PFAS-free products. As identified in Appendix 3: Sources and Uses, Section
3.3 Consumer Products, PFAS have been detected in numerous commercial products, including cleaning products, paint, and treated upholstery. For this action, Ecology would develop a purchasing preference policy (for purchases by the State of Washington) for:

- Cleaning products—dispersed when used and are often discharged down the drain.
- Paint—used for any surface and could release PFAS to the environment.
- Other potential products with likely PFAS treatment, such as furniture or textiles.

A purchasing preference policy would not ban state purchases of PFAS-containing products. Instead, the policy would give points to state contract bidders when they declare (and provide supporting data) that their product does not contain PFAS. These points would be part of the bid scoring process. This would create incentive for bidders to provide PFAS-free products to state agencies and municipalities that use the state contracting process.

Cleaning products

Due to the broad nature of cleaning products and floor finishes that potentially contain PFAS, as well as the ability of janitorial services under state contract to purchase their preferred cleaning products, we could not identify the extent of PFAS-containing or PFAS-free cleaning product use

in Washington. We do note that, of the identifiable products available for direct purchase under the state green janitorial products contract, only one floor polish (sold in three sizes) is explicitly listed as being PFAS-free. Other cleaning products surveyed and identified in supplier product searches do not mention PFAS.

While there is moderate literature on the replacement of PFAS-containing cleaning products, prices and price differences specifically for PFAS are not mentioned. However, we can approximate the price difference by using the example of the price difference between green cleaning products in general, and non-green cleaning products. While this does not necessarily show PFAS contents, it is the nearest available quantification of this potential price difference. We note also that prices will depend on green product availability and market share, as reflected in price differentials by country. These price differences for four countries are summarized below (McCabe, 2008).

Table 88. Price difference between green and non-green products.

Country	All-purpose and floor care products	Sanitary cleaning products	Window cleaners
Sweden	-74%	-82%	-9%
Germany	+36%	+148%	-36%
Spain	+131%	+92%	-94%
Czech Republic	+158%	+2%	N/A

In markets with long-standing incentives and regulation for certain green products, where green substitutes for toxic chemicals are more prevalent, we see that green products are generally less expensive than non-green products. Where regulation or incentives are more recent or nonexistent, green products have a more niche market, and can be significantly more expensive. Data was not reported for the U.S., but we may assume that since 1) nontoxic substitutes for PFAS are currently limited or unknown, and 2) there are observationally few mentions of PFAS-free products in marketing and labeling, initial prices for PFAS-free products (holding other product attributes constant) would be higher than current prices.

Paint

We based cost estimation for paints on historic bids for state waterborne road marking paint contracts (DES, 2018c). This use is more likely to prefer the qualities PFAS provides for products of reduced adherence and staining. The quantity of different types of paint varied, as summarized below. The total quantity and types of paint purchased are expected to vary annually by the needs of planned projects and locations.

Table 89. Paint quantities purchased, by type.

Paint type	Quantity (gallons)
Standard, white, sold by the truckload	216,750
Standard, yellow, sold by the truckload	137,750
Standard, white, sold by less than truckload	12,500
Standard, yellow, sold by less than truckload	8,500
Cold weather, white, sold by the truckload	18,000
Cold weather, yellow, sold by the truckload	18,000
Cold weather, white, sold by less than truckload	0
Cold weather, yellow, sold by less than truckload	0
High build, white, sold by truckload	0
High build, yellow, sold by the truckload	0
High build, white, sold by less than truckload	0
High build, yellow, sold by less than truckload	0
TOTAL	411,500

Across all paint types and quantities, prices were generally consistent across brands. The table below provides summary statistics for waterborne road marking paint prices in the current state contract.

Table 90. Paint price per gallon.

Summary statistic	Price per gallon
Minimum	\$8.50
Median	\$10.99
Maximum	\$19.79
Average	\$11.93

Based on existing prices reflected in state contracts, and allowing for variance in the types of paint purchased, we estimate that purchasing the quantities of paint shown in Table 89 would currently cost between \$3.5 million and \$8.1 million per year.

At this time, it is not possible to identify which road paints do or do not contain PFAS. Acceptable road paint choices are based on a large set of usability, functionality, and wear criteria, some of which may be tied to PFAS or other surfactant contents, but use of PFAS is not identified in choice criteria. Comprehensive paint contents is proprietary, so it is similarly difficult to directly identify PFAS use in existing contracted paints by brand and type. It is therefore also difficult to ascertain, from the literature, the pricing of adequate alternatives, or whether such alternatives are sufficiently available. The limited studies explicitly addressing road paint show that water-based alternatives are available that meet the same functionality and durability criteria (without identifying price differences), but also indicate that further study of alternatives is needed (Kougoulis et al., 2012).

Action 4.2: Evaluate PFAS in landfill leachate

For this action, Ecology would consider adding long term requirements for PFAS testing, monitoring, and reporting for landfill leachate. This could require a rule revision.

Landfill leachate

Landfills in Washington are regulated by local health districts under rules authored by Ecology. Chapters 173-350¹⁵⁴ and 173-500¹⁵⁵ WAC allow health districts to include stipulations in permits that require landfills to sample for additional constituents (e.g. PFAS). If, for some reason, a health district does not want to make that stipulation, then a rule change would be necessary to ensure sampling for PFAS. The process to adopt landfill leachate regulations into rule would likely span over several rulemakings as the science and policy surrounding PFAS continues to evolve. Ecology would likely pursue a single, complex rulemaking to create the initial policy followed by a series of less complex rule updates to incorporate new science.

The request for landfill testing for PFAS in leachate would be generated by Ecology and local health districts. The cost to a landfill operator to test for PFAS would depend both on the frequency of monitoring, and on whether there are requirements to test monitoring wells. Current laboratory cost to analyze for PFAS ranges from \$600 to \$1,200 per sample. Depending on landfill leachate collection configuration, more than one sample may need to be collected. The landfill operator would need to add PFAS to their existing landfill monitoring plan.

There are 53 landfills identified as operating in the state which are likely to store waste containing PFAS (limited purpose and municipal solid waste). Assuming a single sample is taken, testing leachate from 53 landfills would cost \$63,600. Limited purpose landfills, however, are not required to collect leachate. Some limited purpose landfills may collect it voluntarily.

Depending on the scope of additional regulation surrounding PFAS in leachate, additional costs to the industry could also include:

- The cost to update their existing landfill monitoring plan.
- The cost of monitoring well sampling if PFAS has migrated to groundwater from the landfill.

These costs are landfill specific and Ecology cannot estimate them at this time.

10.2 Costs of other options analyzed

In developing this CAP, Ecology considered additional actions that were ultimately not recommended. They include:

¹⁵⁴ https://apps.leg.wa.gov/wac/default.aspx?cite=173-350

¹⁵⁵ https://apps.leg.wa.gov/wac/default.aspx?cite=173-500

- Requiring municipal WWTPs to test influent and effluent. The costs of this option are unknown without the development of individual monitoring plans.
- Recommending that the Legislature require alternatives assessments for specific products (discussion of costs below).

10.2.1 Alternatives assessments

For this action, Ecology could have recommended that the Legislature:

- Allocate funding for Ecology to conduct alternatives assessments of the use of PFAS in products, OR
- Require manufacturers of PFAS-containing products (specific products or industries) to work with independent third-party contractors to conduct alternatives assessments of the use of PFAS in their products.

We identified 13 industries operating in Washington that are likely to use PFAS in their production processes (see <u>Appendix 3: Sources and Uses, Section 3.1.2 Secondary</u> Manufacturing).

Table 91. Industries likely to use PFAS.

Industry	North American Industry Classification System (NAICS) code
Plastics product manufacturing	326199
Automobile manufacturing (plating activity)	3361
Carpet rug mills	314110
Corrugated solid fiber box manufacturing	322211
Electroplating, plating, polishing, and anodizing	332813
Leather hide tanning finishing	316110
Other fabricated wire product manufacturing	331222
Paper mills (except newsprint)	322121
Paper bag coated treated paper manufacturing	322220
Paperboard mills	322130
Pulp mills	322110
Semiconductors and related devices manufacturing	334413
Textile fabric finishing mills	31320

Cost if Ecology assesses alternatives

Ecology assumes that an alternatives assessment costs \$400,000 and takes up to two years. Costs and time would vary by:

- Stakeholder interest and involvement.
- Project scope.
- Robustness of analysis.

The most significant expenditure for a robust alternatives assessment comes from completing the hazardous chemical assessment, which Ecology typically contracts out to a third-party toxicology consultant.

For this analysis, Ecology anticipates completing an alternatives assessment for each of the most common applications of PFAS chemicals in secondary products. PFAS polymer treatments are widely used to provide stain, grease, or water resistance to materials such as carpets and apparel. PFAS are also added to formulated products such as paints and sealers both to improve surface characteristics and to promote even wetting and spreading (fluorosurfactants). Alternative assessments may be appropriate for any or all of these typical PFAS product categories:

- Paint.
- Textiles.
- Cosmetics.
- Cleaning products.
- Floor and car waxes.
- Waterproofing sprays (for leather, carpet, or textiles).
- Automotive fluids.

The process to complete an alternatives assessment for one product is estimated to be two years long and cost \$400,000 for each contract. Actual costs would depend on product category breadth and complexity. Oversight of the contractor and review of the assessment would require one staff person (a full time employee, specifically using one-quarter of their time) per year for two years for each assessment. At a cost of \$400,000 per assessment, the cost to complete assessments of all seven product categories would total \$2.8 million. This estimate is in nominal terms, reflecting the total cost if all costs are incurred immediately (i.e., all seven alternatives assessments are done at once). If one alternatives assessment is done at a time, the cost estimate would be lower, at \$1.6 million based on average rate of return on U.S. Treasury Department Bonds (U.S. Treasury Department, 2020).

Cost if industry assesses alternatives

If industry, as a group, contracts alternatives assessments to consultants, all assessments could be completed in the first two-year cycle. Assuming all assessments begin in the first year, and assessments were completed for the seven applications as assumed for Ecology alternatives analyses, this action would cost \$2.8 million, but assessments would be completed significantly sooner than with the above option.

Price impacts to products

If the alternative chemicals identified and subsequently required are significantly more costly than PFAS, then the prices of products could increase. Since we cannot know the attributes of substitute chemicals before an alternatives assessment is completed, we cannot determine with certainty whether assessments will identify viable alternatives that are significantly more

costly than currently used PFAS. This is the nature of recommended actions that involve research and investigation. For potential price differences for carpet, cleaning products, and paints, see discussion for Actions 3.1 and 3.2, above.

10.3 Benefits of recommended actions

10.3.1 Current state of economic and scientific knowledge about PFAS

Research is emerging on the human health and environmental impacts of PFAS exposure. Because of this, it is not possible to succinctly quantify health- or environmental-related economic benefits of reduced PFAS exposure. However, the literature on relationships between PFAS exposure and impacts to human health and the environment is robust enough to provide a high-level discussion of those impacts and potential costs resulting from those impacts.

There are several key reasons for the poor resolution in the literature on PFAS exposure and health and environmental costs. A significant amount of information pertaining to the exact compositions of PFAS is confidential business information, and is unavailable to independent researchers. Molecular composition can vary widely among different producers, even within particular uses of PFAS, and this information is not available to the public. Studies suggest that PFAS manufacturing data be made public as a method to reduce public health expenditures on toxicology research and to better understand the global effects of PFAS (Scheringer et al., 2014). Unlike substances focused on in Ecology's previous CAPs, little is known about the prevalence, locations, exposure, and quantitative effects of PFAS—which is largely related to the lack of reporting and disclosure requirements.

10.3.2 Human health and wellbeing benefits

Poor human health and related healthcare expenditures are generally associated with lower macroeconomic growth. This relationship results from reductions in:

- Consumer spending on non-medical goods.
- Worker productivity.
- Capacity for public investment in areas outside of healthcare (World Health Organization, 2009).

An increase in human health and productivity would result in macroeconomic benefits.

Despite the emerging nature of PFAS health impact research, several trends in human health conditions associated with exposure to certain PFAS are identified within the literature. These include:

- Increased risk of thyroid disease and endocrine system disruptions.
- Increased risk of certain cancers.
- Higher cholesterol levels.

Reduced antibody response to vaccinations.

<u>Appendix 7: Health, Section 7.1.2 Primary health endpoints of concern</u>, discusses these health impacts in detail. Each of these health issues are associated with direct and indirect costs. Some are terminal illnesses, while others, like high cholesterol and immune deficiencies, increase risk for other illnesses, and are associated more with their secondary costs.

The health conditions associated with PFAS exposure not only affect the lives of sick individuals and their families, but they also influence economy-wide productivity losses. Among all sicknesses and diseases, absenteeism and presenteeism impacts to business productivity can be twice as high as medical and pharmacy costs (Loeppke et al, 2009). Presenteeism occurs when workers are present at their job, but function at a reduced capacity because of a health issue; depression is often cited as an example of a condition that affects presenteeism. The Commonwealth Fund estimated a nationwide impact of \$260 billion in 2003, 2.4% of gross domestic product at the time, for reduced worker productivity, sick days, and the loss of adults from the workforce due to chronic disease and disability (Davis, 2005).

Children who are sick often and miss school may see long-term economic impacts. Recent research established a negative relationship between a child's school absenteeism and their overall performance on tests (García & Weiss, 2018). Studies dating back over 40 years have found positive associations between a person's educational attainment and their earnings, and one recent study linked high school GPA (academic performance) with annual salary, particularly during young adulthood (French et al., 2015).

10.4 Costs from likely PFAS-related health conditions

Because PFAS exposure thresholds associated with these health issues have not been established, it is not possible to quantify healthcare costs associated with PFAS chemicals at this time. Ecology assumes that rates of PFAS exposure are positively correlated with rates of the previously identified health outcomes. A reduction in exposure to PFAS chemicals would logically reduce the risk of these associated health issues and related costs. However, due to limited knowledge regarding how and to what extent PFAS contribute to these health impacts, we could not identify the degree or significance of such reductions.

Therefore, this analysis does not intend to, nor does it, provide a detailed quantitative analysis of the healthcare costs related to PFAS exposure for any one individual or any group of individuals. Instead, we review potential population-wide economic impacts by assessing costs associated with conditions that are likely related to PFAS exposure. As the science linking PFAS exposure with particular health conditions gains resolution, more detailed analyses of individual and population-wide health costs, and associated broader economic impacts, will be possible. Until then, it is not possible to determine how much of a condition's economic effects are related to PFAS exposure. The following discussion does not assume particular correlations

between health-related costs and PFAS exposure, but is meant to be a high-level identification of population-wide costs of potentially associated health outcomes.

All dollar values are reported in 2017 dollars in the analyses below.

10.4.1 Thyroid disease and endocrine disruption

Some studies have found significant associations between PFAS exposure and endocrine disruption (Ballesteros et al., 2017). Research generally shows a positive relationship between thyroid hormone levels and exposure to PFAS, but more research is needed to confirm the relationship and establish exposure thresholds. Both thyroid disease and its associated illnesses are responsible for significant costs to those impacted, and to society as a whole.

According to the Endocrine Society and the Agency for Healthcare Research and Quality, thyroid disease treatment costs for females over age 18 in the U.S. approached \$4.3 billion, with a per person mean expenditure of \$409 for ambulatory services and \$116 for prescriptions (Endocrine Society, 2015; Soni, 2008). Thyroid disease and endocrine disruptions are significantly more common in females than males. From 1996 to 2006, the occurrence of thyroidectomies in the U.S. has increased for both inpatient and outpatient services, with the most significant increases among Medicare and Medicaid patients (Sun et al., 2013). At this time, we cannot estimate the proportion of thyroid disease specifically caused by PFAS exposure, or its interactive or complementary affects in combination with other chemicals or behaviors.

10.4.2 Cancer risk

Studies have linked exposure to PFAS and cancer with varying degrees of significance for over 20 years (Australian Department of Health, 2018). Several distinct populations may have elevated risk of testicular and/or kidney cancer, including:

- Workers directly exposed in chemical plants to PFOA.
- Communities where PFOA exposure is significantly elevated (typically due to water supply contamination or being close to an industrial facility releasing PFOA).
- Those with high rates of background exposure (International Agency for Research on Cancer, 2017).

At this time, it is not possible to estimate the percentage of testicular and kidney cancer cases associated with PFAS exposure.

Cancers are generally shown to have the highest associated medical and pharmaceutical costs among common illnesses (Loeppke et al., 2009; Mitchell & Bates, 2011). Annual charges for an individual with kidney or testicular cancer are estimated at \$39,841 and \$33,747, respectively (U.S. Department of Health and Human Services, 2012). National expenditures for kidney cancer care in 2017 were \$4.7 billion, while those related to testicular cancer approached \$22

million (Aberger et al., 2014; U.S. Department of Health and Human Services, 2012; values reported in 2017 dollars).

The average annual productivity loss per employee due to cancer is commonly estimated at more than \$1,600. Accounting for medical costs and lost productivity, annual cancer impacts to an average-sized company (10,000 employees) can approach \$2.5 million (Mitchell & Bates, 2011). In 2005, productivity losses in the U.S. from testicular cancer were about \$500 million, while kidney cancer was responsible for \$3.4 billion in losses based on productivity losses from both kidney and renal pelvis cancers (National Cancer Institute, 2018).

10.4.3 Higher cholesterol levels

Several studies show links between PFAS exposure and increased cholesterol levels, although the extent to which PFAS exposure is responsible for increased cholesterol is not known (CDC, 2019). Current science suggests that diet is the most significant influencer of high cholesterol (Mayo Clinic, 2017). Although research does not suggest that PFAS exposure plays a significant role in health outcomes related to increased or high cholesterol nationally, there may be economic benefits from even slight reductions in population-wide cholesterol levels, given the widespread occurrence of the condition in the U.S. It is not possible to estimate what percentage of this impact is related to PFAS exposure at this time.

Between 2011 and 2012, just under 40% of U.S. adults had cholesterol levels high enough to be considered at risk for heart disease or stroke, dangerous conditions that are associated with significant costs (CDC, 2018). The CDC estimates that more than 43 million U.S. adults took cholesterol-lowering medications between 2005 and 2012 (Mercado et al., 2015). The cost of these drugs, known as statins, vary significantly, ranging from \$36 to more than \$600 per month (Consumer Reports, 2014). According to the American Heart and Stroke Associations, costs associated with heart disease and stroke in the U.S. exceed \$316 billion, including both medical expenditures and lost productivity. Similar costs related to heart disease alone approached \$200 billion in 2012 to 2013 (American Heart Association/American Stroke Association, 2017).

10.4.4 Secondary immunodeficiency disorders

A number of studies have found associations between PFOA and PFOS exposure and immunodeficiency conditions, including reduced antibody response to vaccinations and hypersensitivity (Chang et al., 2016; Knutsen et al., 2018; National Toxicology Program, 2016; Stein et al., 2016). The literature suggests that PFAS serum concentrations may have significant negative correlations with antibody concentrations in both children and adults, resulting in a reduced protection against pathogens treated by vaccines including tetanus, diphtheria, and rubella (Grandjean et al., 2012; Osuna et al., 2014; Stein et al., 2016).

These health conditions are very rare in the U.S. today, largely because of widespread immunization. A reduction in the effectiveness of these immunizations would increase the

occurrence of the conditions and their associated societal costs. Although it is not possible to estimate potential costs related to PFAS exposure at this time, we assume that an increase in the effectiveness of vaccinations would have economic benefits.

Asthma is a familiar hypersensitivity-related health outcome (National Toxicology Program, 2016; Knutsen et al., 2018). Affecting more than 8% of people living in the U.S., asthma's economic impacts in this country are significant. It is not possible at this time to determine the proportion of asthma cases related to PFAS exposure. Because cost estimates of this relationship are not available, we review total health costs related to asthma in the U.S.

Between 2008 and 2013, the estimated average annual medical cost per person associated with asthma was \$3,266, and the total national cost was \$50.3 billion (Nurmagambetov et al., 2018). The same study suggested that asthma was responsible for 8.7 million and 5.2 million missed days of work and school, respectively, representing a total productivity loss of \$3 billion from 2008 to 2013. In all, total economic loss associated with medical expense, productivity, and mortality was estimated to be \$81.9 billion (Nurmagambetov et al., 2018).

10.5 Environmental benefits

Similar to health benefits, there are several themes evident in the literature regarding PFAS and environmental impacts. As identified in Appendix 6: PFAS Ecotoxicology, Section 6.2
Bioaccumulation, most prevalent among the literature is its persistence within the environment and resulting bioaccumulation in animals, both of which will affect the services ecosystems provide to the public. PFAS are known to be very persistent in the environment and some bioaccumulate over time. Given the documented negative environmental impacts of PFAS emissions and related diminished ecosystem services, Ecology assumes that a reduction in PFAS emissions to the environment would have both environmental and economic benefits.

Ecosystems provide critical functions to society, like purifying water, mitigating the spread of disease, and providing raw materials. These functions are often referred to as ecosystem services. An ecosystem's ability to continuously or predictably provide services is often related to the degree of disturbance experienced by the ecosystem (Farley, 2012). Anthropogenic and natural disturbances to ecosystems often can have a more significant impact on an ecosystem's services than to the ecosystem's long-term resilience. The loss of services provided by ecosystems may threaten a society's economic well-being when the disrupted services cannot be readily substituted (Farley, 2012).

Anthropogenic disruptions to ecosystems can take a variety of forms. Most pertinent to the PFAS discussion is the emission and persistence of these chemicals in the environment and their lasting impacts. One key assumption in ecosystem services economics is that the rate of emissions to an ecosystem cannot not exceed that ecosystem's ability to process the emissions without causing disruption to the provision of ecosystem services (Daly, 1990). In the case of PFAS emissions, the persistence and bioaccumulation of the chemicals are shown to have

negative impacts on the health of water ecosystems, and impacts to organisms in the environment, as discussed in <u>Appendix 6: PFAS Ecotoxicology</u>, <u>Section 6.3 Toxicokinetics</u>.

The degradation of habitat and health impacts to key members of the trophic pyramid may negatively affect Washington's economy. For example, the health of native salmon populations is significantly related to the success of other species like Southern Resident Killer Whales and the economic well-being of tribal, recreational, and commercial fisheries. Key species often have significant cultural and spiritual value, in addition to their ecological and economic significance.

10.6 Data Gaps and Recommendations

10.6.1 Data Gaps

As addressed throughout the CAP and its appendices, data continues to be collected regarding PFAS impacts, possible future regulation, and resulting direct and indirect costs of PFAS controls, remediation activities, and environmental and human impacts.

10.6.2 Recommendations

This appendix does not propose any specific recommendations. Information collected under each of the recommendations proposed in this CAP will allow future refinement of our economic analysis.

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List of acronyms

General acronyms

Table 92. Acronyms found in this appendix.

Acronym	Definition
AFFF	Aqueous film forming foam
CAP	Chemical Action Plan
CDC	Centers for Disease Control and Prevention
DES	Washington State Department of Enterprise Services
DOD	United States Department of Defense
Ecology	Washington State Department of Ecology
EPA	United States Environmental Protection Agency
Health	Washington State Department of Health
lb	Pound
MTCA	Model Toxics Control Act
NH	State of New Hampshire
PPG	Public Participation Grant
sy	Square yard
TCP	Ecology Toxics Cleanup Program
UNEP	United Nations Environmental Programme
WAC	Washington Administrative Code
WWTP	Wastewater treatment plant

Chemical names

Table 93. Chemical name acronyms found in this appendix, excluding the general acronyms listed in the table above.

Acronym	Chemical Name
PFAS	Per- and polyfluoroalkyl substances
PFCA	Perfluoroalkyl carboxylic acid
PFHxS	Perfluorohexane sulfonate
PFOSA	Perfluorooctanesulfonamide
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFSA	Perfluoroalkyl sulfonic acid