September 2017 DRAFT Per- and Poly-Fluorinated Alkyl Substances Chemical Action Plan (PFAS CAP)

The Washington State departments of Ecology and Health prepared a draft of several PFAS CAP chapters for external review. This document is one chapter to a planned multi-chapter PFAS CAP. This material may be modified in response to comments and the content re-organized for the final Action Plan.

The September 2017 Draft PFAS CAP includes: Health, Environment, Chemistry, Regulations, Uses/Sources, Intro/Scope. This draft may include cross-references to other sections/chapters in the Draft PFAS CAP or notes where additional information will be provided in a later draft.

An updated draft of the PFAS CAP will be provided in November/December 2017 for additional review and comment. The PFAS CAP Advisory Committee will discuss comments on these draft chapters at the November 1, 2017 meeting.

Ecology and Health are asking interested parties to provide feedback. Comments on these draft documents are due to Ecology by **October 20, 2017.**

Submit comments, suggestions, and questions to Kara Steward at kara.steward@ecy.wa.gov.

The Draft PFAS CAP documents are posted at <u>https://www.ezview.wa.gov/?alias=1962&pageid=37105</u> (at the bottom of the webpage).

Summary of PFAS in Washington's Environment

PFAS are released to the environment through manufacturing emissions, releases or disposal, and the use and disposal of products containing PFAS. Major pathways of <u>specific</u> PFAS <u>emissions</u> to the environment have been identified as <u>point sources such as manufacturing</u>, stormwater <u>discharges</u>, wastewater treatment plant (WWTP) effluent, discrete releases from <u>direct</u> product use (i.e., AFFF use), land application of biosolids, and atmospheric deposition. The relative importance of these pathways in Washington State is not well-characterized; however, environmental monitoring and drinking water testing suggest that stormwater, WWTP effluent, and AFFF use are primary ways <u>specific</u> PFAS are delivered to our waterbodies.

PFAS have been detected in all sample types analyzed in Washington State: surface waters, groundwater, WWTP effluent, freshwater and marine sediments, freshwater fish tissue, and osprey eggs. Other media types have not been sampled in Washington, such as biosolids and landfill leachate. Environmental monitoring in the state has shown that PFAS, particularly shortchain PFAAs, are present in WWTP effluent, urban surface water, and surface waters receiving WWTP effluent with minimal dilution. PFOS, and to a lesser extent other long-chain PFAAs, were widespread in freshwater fish tissue of waterbodies in the state, with PFOS levels in urban lakes that may trigger consumption advisories to protect human health. PFOS and long-chain PFAAs are also detected in osprey eggs, at concentrations lower than would affect offspring survival, but potentially high enough (reference please) to reduce hatchability in samples from sites affected by urban sources and WWTP inputs.

Environmental concentrations of PFAAs in Washington State surface waters, WWTP effluent, and freshwater fish tissue sampled in 2016 are consistent with PFAS levels in other parts of the U.S. 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.

Environmental monitoring in 2016 suggested that PFAA levels in surface waters and WWTP effluent have decreased since the last round of sampling in 2008. A general shift in PFAA compound make-up was evident in WWTP effluent samples, with short chain PFAAs replacing PFOA as the most dominant compounds in effluent. PFAS concentrations in freshwater fish tissue and osprey eggs (primarily made up of PFOS) have remained unchanged between 2008 and 2016. PFOS continues to be a ubiquitous contaminant in Washington State aquatic biota.

- Washington State currently has no known PFAS manufacturing facilities, and therefore most contamination in the state's environment likely comes from product use (past and/or present) and disposal and atmospheric deposition. Currently, the relative importance of different environmental pathways (e.g., WWTP effluent, AFFF use, etc.) is not characterized for Washington State.
- Relatively few studies have been conducted on PFAS in Washington. PFAS analyses in Washington State have primarily been limited to the perfluoroalkyl acids (PFAAs), such as perfluoroalkane sulfonic acids (PFSA) and perfluoroalkyl carboxylic acids (PFCA).

Commented [A1]: Can you please provide a copy of Ecology, 2017 to allow for a more comprehensive review of this document? Consider extending the comment period for this paper since not all publications referenced and used in this report are available to the public.

The Chapter on Marine Fish should be removed as there is no content to review.

Please comment on "data quality" in the "Summary" chapter.

Has Ecology reviewed the UCMR3 data for Washington and summarized the findings in any of these reports?

Commented [A2]: Below please see a suggested, revised Summary of this Chapter listing the key findings from the key reports.

Commented [A3]: Need to justify this comment given the historical/legacy use of long-chain chemistries. Additionally, what is the reason for calling out short-chain PFAAs when they are not the dominate PFAS in some waters/media tested?

Commented [A4]: As a comment here and on the entire chapter, it would be useful to also say which PFAS were sampled for in the discussed study(s), but not found. For example, PFOA and PFHxA were not found in biota in the 2016 sampling.

Additional compounds, including precursors that are known to potentially break down into PFAAs, were analyzed in surface water and WWTP effluent samples collected in 2016. Only few of these were detected, most of them one-time at very low concentrations.

Key findings from the occurrence studies are summarized below by environmental matrix. For some matrices, data from multiple sampling events are available and provide insights on the changes in PFAS profiles over time, both in magnitude and relative percent composition. Changes in analytical detection limits may contribute to increased frequency of detection of some PFAS, particularly for measurements of [list which matrices this applies to...].

Surface Water

• Freshwater Monitoring (2008, 2016)

- <u>The 2008 Study focused on 14 sites. 11 sites had total PFAAs concentrations</u> below 20ng/L, one site had total PFAAs concentrations of 185ng/L (spring) and 170ng/L (fall) with PFOA being the largest contributor, two sites had total PFAAs concentrations below 80ng/l and above 20ng/L. All spring samples contained measurable concentrations of at least one of the target 11 PFAAs. Fall samples detected PFAAs in all but two samples.
- The 2016 Study focused on 15 sites. Less than half of the surface water samples contained any of the 25 targeted PFAS compounds. Only heavily impacted waterbodies had detections those with WWTP inputs and in urban areas. Of the 13 precursor PFAS monitored, only 3 were detected and only one-time.
- Detection frequencies and total concentrations in the 2016 Study were generally lower than those of surface water samples collected in 2008 at the same sites.
- Saltwater Monitoring (2014)
 - Puget Sound Study (2014): Monitoring for 14 PFAAs at seven Puget Sound sites and six sites in British Columbia. At least one PFAA compound was detected in all samples analyzed. PFH_pA, PFOA and PFOS were the most frequently detected compounds in the sample. Overall concentrations in marine water were lower than for urban freshwater sites.

Drinking Water and Groundwater

- UCMR3
- State monitoring studies

WWTP Effluent

• WWTP Studies (2008, 2016)

Commented [A5]: Please see the comment above. Without being able to read the report, it is difficult to provide a detailed summary suggestion.

- The 2008 Study analyzed 11 PFAAs in effluent from 4 WWTPs in the Spring and in the Fall. All but one PFAA were detected with PFOA being the dominant compound detected. The Spring median concentration for the sum of PFAAs was 218.3 ng/l (mean 97 ng/l). The Fall median concentration for the sum of PFAAs was 139.6 ng/l (mean 148.4 ng/l).
- <u>The 2016 Study analyzed 35 PFAS in effluent from 5 WWTPs in the Spring and in the Fall.</u>
- PFAA concentrations in effluent samples collected in 2016 were consistently lower than PFAA concentrations measured in the 2008 Study 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 PFBA.
- <u>Puget Sound (Phase 3) Study: 12 PFAAs and PFOSA were monitored in effluent</u> from 10 WWTPs during the winter and summer of 2009. Six to ten of the compounds were detected in every sample. PFHxA, PFNA, PFOS, and PFOA were present in the greatest concentrations and were detected in all samples.

Sediment

- Freshwater Locations (2012)
 - Sediment cores from three freshwater lakes were collected in 2012 for analysis of 12 PFAAs and PFOSA. PFOS and long-chain PFAAs were the dominant compounds measured. PFAA concentrations in the surface sediments ranged from 0.392 ng/g dw to 7.0 ng/g dw. 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.
- Marine Locations (2013 2015)

Marine sediment from Puget Sound urban bays was collected in 2013, 2014 and 2015 for analysis of specific PFAAs and PFOSA. Not all sediment samples showed presence of the PFASs tested. In several samples PFOS, PFOSA and PFOA were detected.

Biota Surveys

- Freshwater Fish (2008, 2016)
 - Ecology collected 15 freshwater fish samples from 7 waterbodies throughout the state in 2008 for analysis of 10 PFAAs. A total of 11 different species were collected and analyzed as composite fillet and composite liver samples. Of the PFAAs analyzed, only PFOS, PFDA, PFUnDA, and PFDoDA were detected and quantified. PFOS was detected in 67% of the liver samples and 40% of fillet samples. Concentrations of PFOS in liver samples ranged from <10-527 ng/g ww with a median value of nnnn and a mean value of mmm. Fillet samples had PFOS

Commented [A6]: Please include median and mean values.

concentrations of <10-75.5 ng/g ww with a median value of vvv and a mean value of ccc. PFDoDA, PFUnDA, and PFDA were each detected once at concentrations of 21.0-46.1 ng/g ww for liver tissue and 5.5-7.5 ng/g ww for fillets.

- <u>Construction</u>
 <u>Construction</u>
 <u>Ecology collected freshwater fish of various species from 11 waterbodies in</u>
 <u>Washington State in 2016 as part of the follow-up study to 2008 sampling. A total of 22 composite samples of freshwater fish fillet tissue and 22 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 PFAA concentrations ranged from <1-87.3 ng/g ww (median = 3.92 ng/g ww) and liver PFAS concentrations ranged 5.12 to 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.
 </u>
- o Please include a brief summary comparing results from the two studies.
- PFOS concentrations in six of the fillet samples were above (greater) than the Washington Department of Health's (DOH's) provisional general population screening level for PFOS in edible fish tissue (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 DOH'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 DOH, but determined to have insufficient sample sizes for a fish advisory assessment.
- PBT Screening Study (2011)

In 2011, Ecology collected common carp and largescale 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. 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.

- Osprey Egg Studies (2008, 2016)
 - Ecology collected eleven osprey eggs in 2008 from the Lower Columbia River and tested the inner contents (whole egg without shell) for 13 PFAAs-. Egg homogenates contained PFAA concentrations ranging from 38 – 910 ng/g fresh weight (fw)-. Similar to fish tissue, PFOS was the dominant compound, followed by PFUnDA and PFDA. Some PFAA were detected less frequently and at low concentrations. Some PFAA were not detected.
 - In 2016, Ecology collected osprey eggs from the Lower Columbia River, Lake Washington, and West Medical Lake-. A total of 11 osprey eggs were analyzed for 12 PFAAs and PFOSA. All eggs contained one or more PFAAs, with PFAA concentrations ranging 11.7- 820 ng/g fw (median = 99.8 ng/g fw).

Commented [A7]: See comment above.

Commented [A8]: Please include value for the mean.
Commented [A9]: See comment above.

Commented [A10]: Please include the mean and the median value.

Commented [A11]: Please include the range, median and mean value.

Commented [A12]: Please be specific which PFAAs were detected and which ones were not. Please include the range, median and mean for the dominant compound.

Commented [A13]: Please include data for the mean.

- None of the osprey eggs analyzed for this study had PFOS concentrations exceeding a Practical No Effects Concentration of 1,000 ng/g for offspring survival in a top avian predator (Newsted et al., 2005).
- <u>PFOS concentrations in five of the samples were above a Lowest Observable</u> Adverse Effect (LOAE) 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.
- 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.

Environmental Transport Pathways and Environmental Fate

PFAS can be released to the environment as emissions during manufacturing, and during the use and disposal of products containing PFAS (Figure 1, from OECD, 2013). After products containing PFAS are used or disposed of, the certain non-polymeric PFAS are transported into and through the environment through pathways such as stormwater, discrete releases from product use, wastewater treatment plant (WWTP) effluent, biosolids application, landfill leachate, and atmospheric deposition.

Stormwater and runoff can transport PFAS to aquatic systems when the accumulation of dry deposition and indoor/outdoor releases of PFAS from consumer and industrial products are washed into rivers and streams during rain events (Muller et al., 2011). This is a particularly important pathway in urban areas, as industrial and commercial activities have been identified as primary determinants of PFOS levels in lakes, with surface runoff as the primary pollution carrier (Xiao et al., 2013).

The use of AFFFs have been identified as a direct mode of PFAS release to the environment (Paul et al., 2009; Ahrens and Bundschuh, 2014). Levels of environmental PFAS contamination found in groundwater, surface water, sediments, and fish near sites that use AFFFs, such as airports and military bases, are much higher than those found in non-point source areas (Gewurtz et al., 2014; Ahrens et al., 2015; Barzen-Hanson et al., 2017). AFFF use may continue to be a source to the environment for many decades at sites with a history of repeated applications, as in fire training areas (Baduel et al., 2015). <u>Need to reference FFFC documents including best practices as well as training with AFFF has decreased very significantly everywhere. Please note, the Best Practice Guidance for Use of Class B Firefighting Foams published by the Fire Fighting Foam Coalition</u>

(https://docs.wixstatic.com/ugd/331cad_188bf72c523c46adac082278ac019a7b.pdf) recommends the following: a) Use training foams that do not contain fluorosurfactants for training purposes. **Commented [A14]:** Please be specific throughout the document as your statements indicate properties and presence of specific classes of PFAS that are not reflective of the entire PFAS category – please see a few (but not all) examples highlighted. Throughout the document, mainly PFAAs are addressed that belong to the sub-group of PFSAs and PFCAs.

Commented [A15]: Is it possible to quantify the contribution to the total for each such categories for Washington State, for the USA or globally?

Commented [A16]: The report identifies landfill leachate as a potential source of PFAS and cites the recent article by Lang et al. (2017). There is extensive literature on this topic, including results from studies of landfills in the Pacific Northwest (Schultz et al. 2006), New York (Sinclair and Kanann 2006); Canada (Benskin et al. 2012; Li et al. 2012), Germany (Busch et al. 2010), Denmark (Bossi et al. 2008), Sweden (Woldegiorgis et al. 2006), Finland and Norway (Kallenborn et al. 2004), and Japan (Murakimi et al. 2008; Zushi et al. 2007). Consider using some of these study findings to help guide future investigations in Washington.

Formatted: Highlight

Formatted: Highlight

<u>B) Use surrogate liquid test methods that do not contain fluorosurfactants for testing fixed system and vehicle foam proportioning systems.</u>

WWTP effluent has been identified as a major transport pathway of PFAS to the aquatic environment (Sinclair and Kannan, 2006). PFAS enter WWTPs through the collection systems in domestic waste and non-domestic waste discharges, and, for plants with combined collection systems, in stormwater. WWTPs are not designed to remove or break down these chemicals, and PFAS enter the environment through the discharged effluent (Sinclair and Kannan, 2006; Alder and van der Voet, 2015). Some PFAS, particularly the long-chain PFAAs, will partition to sludge in WWTPs and may be released to the environment through land applications of biosolids (Sepulvado et al., 2011; Venkatesan and Halden, 2013).

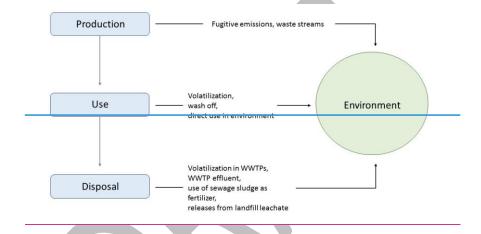


Figure 1. Diagram of PFAS Release to the Environment Adapted from OECD (2013).

Landfill leachate often contains concentrated (? Quantify this statement please) levels of PFAS, which can be released to groundwater in unlined landfills or transported to WWTPs for treatment and become part of effluent discharge (Lang et al., 2017). Landfill leachate may be locally important because of direct discharge to groundwater or surface water (Busch et al., 2010). Due to the slow release of landfill leachate, PFAS will continue to be discharged for many years after product disposal (Lang et al., 2017).

PFAAs and their precursors can be transported atmospherically and deposited through wet and dry deposition (Stock et al., 2004; Prevedouros et al., 2006). PFAAs are generally not volatile and are removed from the atmosphere relatively quickly (Hurley et al., 2004). Volatile precursor compounds, such as fluorotelomer alcohols, have longer atmospheric lifetimes and are capable of long-range transport (Ellis et al., 2004; Martin et al., 2006). These precursors then degrade to PFAAs either in the atmosphere or after deposition, resulting in PFAA contamination in remote areas (Stock et al., 2004; Young et al., 2007; Ellis et al., 2004). PFAAs also reach remote areas, such as the Arctic, through oceanic transport (Prevedouros et al., 2006).

Formatted: Highlight

Formatted: Highlight

The ultimate sink for PFAAs is the global reservoir of oceans and sediments (Prevedouros et al., 2006). Long-chain PFAAs will continue to be released into the environment for some time through degradation of PFAA precursors, legacy products still in use, and remobilization between environmental compartments (Ahrens and Bundschuh, 2014).

Environmental Pathways in Washington State

Washington State currently has no known PFAS manufacturing facilities, and therefore most contamination in the state's environment likely comes from product use (past and/or present) and disposal and atmospheric deposition. Currently, the relative importance of different environmental pathways (e.g. WWTP effluent, AFFF use, etc.) is not characterized for Washington State. Environmental monitoring in Washington suggests that PFAS concentrations are highest in waterbodies located in urban settings and where WWTP effluent makes up a significant portion of the flow or hydrologic dilution is minimal. Monitoring in the state has focused on releases of PFAS to surface water and the aquatic food chain. Background levels in soil or geographical distribution of PFAS in soils, groundwater, or air has not been investigated.

PFAS in Washington's Environment

The following sections discuss available PFAS data on environmental media collected in Washington State. Relatively few studies have been conducted on PFAS in Washington. PFAS analyses in Washington State have primarily been limited to the perfluoroalkyl acids (PFAAs) included in EPA Method 537 (see Section X.X [Chemistry chapter] for a more detailed discussion on PFAS analytical methods). Additional PFAS compounds, including precursors that are known to or potentially break down into PFAAs, were analyzed in surface water and WWTP effluent samples collected in 2016.

Air

I

Ecology did not identify any studies or analyses of PFAS compounds in Washington's air.

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). This soil sample had an elevated concentration of PFOS, at 12,000 ug/g, followed by PFUnDA (1,100 ug/g), PFNA (120 ug/g), and PFDS (110 ug/g) (accessed from Ecology's Environmental Information Management Database on 8/30/2017 at:

http://www.ecy.wa.gov/eim/). Other perfluoroalkyl acids were detected at levels less than 100 ug/g. Following this sampling event, the impacted soils were excavated and removed from the site for proper disposal.

Commented [A17]: The report states that Washington currently has no known PFAS manufacturing facilities. However, there should be cross reference to the Uses/Sources chapter (especially Tables 3 and 4). For example, there's many potential industrial sources of PFAS when one considers the range of materials, industrial processes, and products that may involve PFAS. Examples include: textiles; wood flooring; newspaper printing; yarn manufacturing, bleaching, dyeing; cloth production, laundry and dry cleaning; paper and corrugated board production; lamp production, varnishing, pickling, phosphating; and chromium plating (Clara et al. 2008). The State should acknowledge that an assessment of these types of sources has not been conducted and remains a data gap; but also, there are steps that could be taken to address this data gap. This discussion should be included in the section on Data Gaps on p. 13.

Formatted: Highlight

Commented [A18]: Please include information on overall data quality and please provide how many samples were collected and analyzed for each site/specimen and how many repeats were run in the quantitative analysis. Is it correct to assume that each data point provided is an average of 3 different samples with 3 repeated analysis? Can you provide the standard deviation for each of these single data points?

Groundwater

Ecology did not identify any ambient groundwater monitoring of PFAS in Washington State. However, PFAS have been detected in groundwater wells used for drinking water in several areas. Section X.X [Health chapter] describes this sampling.describes this sampling. Furthermore, UCMR3 provides occurrence data for three PFSAs and three PFCAs in drinking water. Samples were collected between 2013 and 2015. (https://www.epa.gov/dwucmr/occurrence-data-unregulated-contaminant-monitoring-rule)

Surface Water

Ecology's 2008 Statewide Study: In 2008, Ecology carried out a study measuring <u>11 different</u> PFAS in a variety of environmental media throughout the state to determine their occurrence in the state's freshwater systems (Ecology, 2010). This study collected freshwater from 14 waterbodies 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 (T-) PFAAs¹ from 1.11 - 185 ng/L. Fall samples contained detected PFAAs in all but two samples, ranging in T-PFAAs from <0.9 - 170 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. The majority of total concentrations (78%) recorded during both seasons were less than 10.5 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 waterbodies were collected in the spring and fall for analysis of 12 PFAAs and 13 known or potential precursors to PFAAs². Less than half of the surface water samples contained PFAS compounds. T-PFAA³ concentrations ranged from <2-153 ng/L in the spring and <2-170 ng/L in the fall. Only heavily impacted waterbodies had detections – those with WWTP inputs and 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 waterbodies lacking point sources collected throughout Michigan, Rhode Island, and New York (MDEQ, 2015; Zang et al., 2016). All surface water samples were 1-2 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; MDEQ, 2015; Newton et al., 2017).

Commented [A19]: Expand the subtitle from

"Groundwater" to be "Groundwater and Drinking Water", since both are discussed. There is a cross reference to the Health chapter, which includes a detailed discussion of drinking water occurrence data (including UCMR3) on p.5. It would be clearer to present the summary of the drinking water occurrence data in this Environment chapter, and reserve the Health chapter for discussion of studies that associate the drinking water concentrations with serum measurements or health surveys.

Commented [A20]: Please include the reference here for public review or only refer to a chapter in another document.

Commented [A21]: The report states that precursors were detected in a single sample. Include the total sample size (i.e., 1 of how many?).

Commented [A22]: Please consider including Figure 3 and Table 4 from that report here.

Commented [A23]: Please consider including a summary Figure (similar to Figure 3 in the 2008 study) and a summary table (similar to Table 4 in the 2008 study)

Commented [A24]: How many sample locations had no detection? Please include.

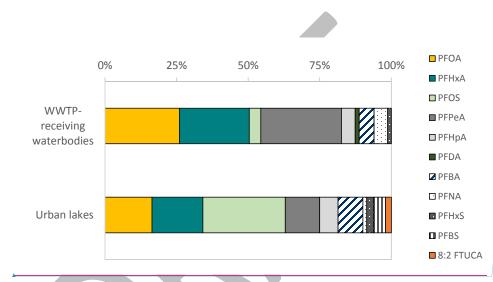
Commented [A25]: Samples from these locations displayed the highest values measured in the 2008 study. Is that true for the 2016 sampling cycle as well?

¹ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFDA, PFDA, PFBS, PFHxS, PFOS, and PFDS.

 $^{^2}$ Precursors analyzed included polyfluorinated sulfonamides, fluorotelomer carboxylates (saturated and unsaturated), and fluorotelomer sulfonates.

³ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS.

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 surface water samples. The only precursor PFAS compounds detected were 8:2 FTUCA, 4:2 FTS, and 6:2 FTS, which were all detected only once at 1.02, 11.3, and 6.87 ng/L(ppt's)., respectively. In the waterbodies impacted by WWTP effluent (West Medical Lake and South Fork Palouse River), PFPeA, PFOA, and PFHxA were the most dominant compounds, each contributing an average of 24% to 28% of the total PFAS concentration. The urban lakes were dominated by PFOS first, and then by the compounds seen in the WWTP-impacted sites.



Commented [A26]: Can you please include the concentrations in ng/l? Can you please a similar graph for the samples from West Medical Lake and South Fork Palouse River as they might have had the highest PFAA concentrations.

Formatted: Font color: Green, Kern at 14 pt, Contextual Alternates

Figure _. Average PFAS Compound Profiles in Two Types of Surface Waters Collected from Washington State Waterbodies in 2016. WWTP-receiving waterbodies = South Fork Palouse River and West Medical Lake; Urban lakes = Angle, Meridian, and Washington Lakes.

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-PFAA⁴ concentrations ranged from 1.5 - 41 ng/L (therefore the sum of all detected species ranged from 1.5 - 41 ppts). The highest concentrations were found in two urbanized sites draining to Puget Sound (First Creek in Tacoma and Portage Bay in Seattle). T-PFAAs 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

Commented [A27]: Would be helpful to have absolute levels in ppts for each of these species in Figure.

⁴ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFBS, PFHxS, PFOS, and PFDS.

Barkley Sounds. PFHpA, PFOA, and PFOS were the most frequently detected compounds in the samples.

WWTP Effluent

Ecology's 2008 Statewide Study: 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-PFAAs⁵ ranging 61-418 ng/L in the spring and 73-188 ng/L in the fall. PFOA, the dominant compound detected, contributed an average of 36% and 32% to the T-PFAA concentration in the spring and fall, respectively. In spring samples, PFHxA was the next most-dominant compound (average of 28% contribution to T-PFAA concentration.<u>or ?? ppt's</u>), followed by PFPeA (average of 10%). PFHxA and PFPeA had similar percent contributions in the fall samples (16-17% of the total).

Ecology's 2016 Statewide Study: Ecology collected effluent from 5 WWTPs in during the spring and fall of 2016 for analysis of 35 PFAS compounds (12 PFAAs and 23 known or potential precursor compounds⁶) (Ecology, 2017). PFAS were detected in all WWTP effluent samples analyzed. Spring T-PFAA⁷ concentrations ranged from 42.1 to 107 ng/L, with a median of 68.9 ng/L. Fall concentrations were similar, ranging in T-PFAA from 41.8 to 125 ng/L, with a median of 71.4 ng/L. The PFAA concentrations from all WWTP 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 waste containing AFFF (Appleman et al., 2014; Houtz et al., 2016).

Perfluoroalkyl acids were the primary PFAS compound type found in the effluent samples. Only four of the precursor compounds were detected: PFOSA, 6:6 PFPi_(identify this please),-6:2 diPAP, 8:2 diPAP, which ranged in concentration from 2.8 to 19.3 ng/L. 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).

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 PFBA.

- ⁶ Precursors analyzed included polyfluorinated sulfonamides, fluorotelomer carboxylates (saturated and
- unsaturated), fluorotelomer sulfonates, perfluoroalkyl phosphonates, and polyfluoroalkyl phosphates.
- ⁷ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS.

Commented [A28]: Can you please provide the values?

Commented [A29]: Please consider including Figure 6 and Table 6 from this report.

Commented [A30]: Please consider including a summary figure and summary table listing the concentrations in ng/l so that a good comparison with the 2008 study is possible.

Commented [A31]: Can you please provide more context? Where those concentrations significant when compared to the total?

⁵ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, PFOS, and PFDS.

Control of Toxic Chemicals in Puget Sound (Phase 3) Study: Ecology and Herrera (2010) analyzed 12 PFAAs and PFOSA in effluent from 10 WWTPs during the winter and summer of 2009. All 10 WWTPs discharge treated effluent to Puget Sound tributaries, as the goal of the study was to provide estimates of contaminant loadings to the Puget Sound. Six to ten of the compounds were detected in every sample. T-PFAA⁸ concentrations ranged from 35.3 - 194 ng/L in the winter and from 46.3 - 146 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.

Sediments

Sediment Cores: In 2012, Ecology collected sediment cores from three freshwater lakes for analysis of 12 PFAAs and PFOSA (Ecology, 2013). PFOS and long-chain PFAAs were the dominant compounds measured. T-PFAA⁹ concentrations in the surface sediments ranged from 0.392 ng/g 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.

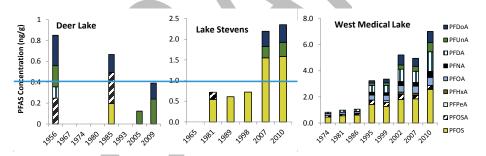


Figure _. PFAS Concentrations (ng/g dw) in Sediment Core Samples Collected in Washington State in 2012.

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 PFAAs and PFOSA. T-PFAA values were not reported. In Elliott Bay, PFOS was detected in 7 out of 30 stations, with detected concentrations ranging

Commented [A32]: How many?

⁸ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS.

⁹ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS.

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 to 0.57 ng/g dw (*accessed from Ecology's Environmental Information Management Database on 3/21/2017 at:* http://www.ecy.wa.gov/eim/).). Six out of 30 Commencement Bay stations (20%) contained one or more detected 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 to 1.6 ng/g dw (*accessed from Ecology's Environmental Information Management Database on 3/21/2017 at:* http://www.ecy.wa.gov/eim/). The highest concentration of PFOS (1.6 ng/g dw) was found in a sediment sample collected from Sinclair Inlet.

Freshwater Fish

Ecology's 2008 Statewide Study: Ecology collected 15 freshwater fish samples from 7 waterbodies throughout the state in 2008 for analysis of 10 PFAAs (Ecology, 2010). A total of 11 different species were collected and analyzed as composite fillet and 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 <10-527 ng/g ww. Fillet samples had PFOS concentrations of <10-75.5 ng/g ww. PFDoDA, PFUnDA, and PFDA were each detected once at concentrations of 21.0-46.1 ng/g ww for liver tissue and 5.5-7.5 ng/g ww for fillets.

Ecology's 2016 Statewide Study: Ecology collected freshwater fish of various species from 11 waterbodies in Washington State in 2016 (Ecology, 2017) as part of the follow-up study to 2008 sampling (Ecology, 2010). A total of 22 composite samples of freshwater fish fillet tissue and 22 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-PFAA¹⁰ concentrations ranged from <1-87.3 ng/g ww (median = 3.92 ng/g ww) and liver T-PFAS concentrations ranged 5.12 to 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 concentrations found near point sources by recent U.S. and Canadian studies, and within the range seen in other waterbodies lacking point sources (MDEQ, 2015; Lanza et al., 2016; and Gewurtz et al., 2014).

PFOS concentrations in six of the fillet samples were above (greater) than the Washington Department of Health's (DOH's) provisional general population screening level for PFOS in edible fish tissue (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 DOH'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 **Commented [A33]:** Please include Figure 8 from this study as it provides an excellent overview of the findings..

Commented [A34]: Please include median and mean values.

Commented [A35]: See comment above.

Commented [A36]: Please include value for the mean. Commented [A37]: See comment above.

¹⁰ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS.

the provisional general population screening level. This data was evaluated by DOH, but determined to have insufficient sample sizes for a fish advisory assessment.

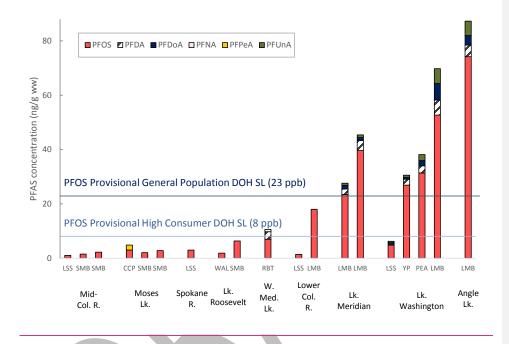


Figure _. PFAA Concentrations (ng/g ww) of Freshwater Fish Fillet Samples Collected in Washington State in 2016.

Eleven freshwater fish tissue samples analyzed for PFAS in 2016 had paired species/waterbody data from 2008. 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 LOQ, showing no overall apparent pattern. No temporal pattern was evident with liver samples, either, despite higher detection frequencies.

2011 PBT Screening Study: In 2011, Ecology collected common carp and largescale 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 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

Commented [A38]: Figure 8 in Ecology, 2010 shows much higher values for samples taken from Lk. Washington and Lower Columbia. Please explain in more detail why the two data sets cannot be compared.

detected infrequently or not at all. T-PFAA¹¹ concentrations across both species and sample types ranged from 2.1 - 91.9 ng/g ww, with the highest concentration in the Lake Washington largescale sucker whole body sample.

Marine fish

[Placeholder for juvenile salmonid and mussel data/summary from WDFW. If not received by December, this section will be removed.]

Osprey

Ecology's 2008 Study: 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-PFAA¹² concentrations ranging from 38 - 910 ng/g fresh weight (fw) (Ecology, 2010). Similar to fish tissue, PFOS was the dominant compound, followed by PFUnDA and PFDA. Other Some acids were detected less frequently and at low concentrations. Some acids were not detected (< LOD). The majority of eggs contained less than 100 ng/g fresh weight (fw); however, three eggs contained levels greater than 250 ng/g fw_r. Please consider inserting Figure 13 from Ecology, 2010 and insert a similar figure from Ecology, 2017 (if it is available).

Ecology's 2016 Study: 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 one or more PFAAs, with T-PFAA¹³ concentrations ranging 11.7- 820 ng/g fw (median = 99.8 ng/g fw). 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.

Commented [A39]: Please include the mean and the median value.

Commented [A40]: Please include the range, median and mean value.

Commented [A41]: This sentence is unclear.

Commented [A42]: Please be specific which PFAAs were detected and which ones were not. Please include the range, median and mean for the dominant compound.

Commented [A43]: Please include data for the mean.

¹¹ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, and PFOS.

¹² Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, PFOS, and PFDS.

¹³ Sum of detected perfluoroalkyl acid concentrations: PFBA, PFPeA, PHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, PFOS, and PFOSA.

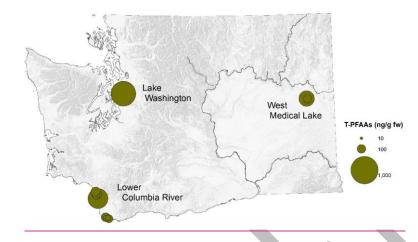


Figure _. Total Perfluoroalkyl Acid Concentrations (ng/g fw) Measured in Osprey Eggs Collected in 2016.

PFOS made up 69% to 94% of the PFAA burden in the osprey eggs. 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, these concentrations were quite low, at 0.45 – 1.83 ng/g fw, and made up less than 2% of the total.

None of the osprey eggs analyzed for this study had PFOS concentrations exceeding a Practical 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 (LOAE) 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 LOAE 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).

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.

Wildlife Studies outside of Washington

PFAS have been detected throughout the world in wildlife that haven't been sampled in Washington State. Giesy and Kannan (2001) were the first to find detectable levels of PFOS in a wide range of biota, including species such as bald eagles, polar bears, and seals, from urbanized **Commented [A44]:** Please include the range, median and mean values.

Commented [A45]: Reference is made to Practical No Effects Concentration (PNEC) given by Newsted et al. (2005). To assist readers, include a brief explanation of how this metric is calculated and how it is interpreted.

Commented [A46]: Please be specific which PFAS have been detected.

centers in North America to remote regions of the Arctic and North Pacific Oceans. Literature reviews done in the mid-2000s confirmed **PFAS** contamination at all levels at the food chain, and particularly elevated levels in fish-eating animals living near industrialized areas (Houde et al., 2006; Lau 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).

Environmental Data Gaps in Washington

Washington State is lacking in some key areas of characterizing PFAS contamination in the environment, such as ambient groundwater monitoring, source assessments of PFAS in urban waterbodies, 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 State. <u>PFAS-PFAA</u> 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 urban lakes as sites of elevated <u>PFAS_PFAA</u> contamination in Washington State, yet we do not a have a full understanding of what the sources to these waterbodies are. Other research on <u>PFAS_PFAA</u> in urban waterbodies has suggested sources related to traffic or automobile/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).

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 biota, though levels have generally been lower than PFOS (Chu et al., 2016; Shi et al., 2015; Baygi et al., 2016). Other novel PFAS, such as cyclic perfluoroalkyl acids and fluorosurfactants, have been found to accumulate in fish from waterbodies directly impacted by AFFF use (Wang et al., 2016; Munoz et al., 2017). Recent research has identified hundreds of new PFAS, many of which have been identified in the aquatic environment (Xiao, 2017). Aside from a limited list of precursor compounds measured in surface waters in 2016, none of these emerging PFAS compounds have been analyzed in Washington State samples.

Commented [A47]: Please be specific which PFAS have been detected.

Commented [A48]: As noted on pp. 3-4, landfills are another potential source of PFAS. References such as Lang et al. (2017) which provides results from a recent national survey, and numerous others cited in Comment #1, can be used to prioritize future sampling efforts. For example Washington should consider mapping the landfills by category of material received (e.g., municipal, industrial, private), leachate capture and treatment technology, and climate conditions, all of which may be predictors of the type and amount of annual releases.

Appendix Table

Table A-_. PFAS Concentrations in Washington State Environmental Media.

Sample Matrix	Sample Type	Collection Year	Units	n	# PFAA compounds analyzed	T-PFAAs	PFBA	PFPeA	PFHxA	PFHpA	PFBS	Ref.
Surface water	Spring	2008	ng/L	14	11	1.1-185 (7.5)	<0.1- 3.6	<0.1- 26.5	<1.0- 10.5	<1.0- 28	<0.1- 0.6	Ecology, 2010
Surface water	Fall	2008	ng/L	14	11	<0.9- 170 (3.6)	<0.1- 5.5	<0.5- 32	<0.1- 37	<0.9- 22	<0.1- 2.0	Ecology, 2010
Surface water (fresh and marine)	Spring/summer/ fall/winter mean	2009-2010	ng/L	13	14	1.5-40	NR		NR	NR	NR	Dinglasan-Panlilio et al., 2014
Surface water	Spring	2016	ng/L	15	12	<2-153 (<2)	<1.0- 13	<1.0- 29	<1.0-33	<1.0- 13	<2.0- 2.1	Ecology, 2017
Surface water	Fall	2016	ng/L	15	12	<2-170 (<2)	<1.0- 12	<1.0- 39	<1.0- 32.5	<1.0- 13	<2.0- 13	Ecology, 2017
WWTP effluent	Spring	2008	ng/L	4	11	61-418 (218)	0.7-3.3	3.8-31	14.5-141	4.1-35	<0.1- 1.5	Ecology, 2010
WWTP effluent	Fall	2008	ng/L	4	11	73-188 (140)	1.9-5.4	13-47	11-30	<3.5- 13	<0.5- 6.6	Ecology, 2010
WWTP effluent	Summer	2009	ng/L	10	12	46-146 (93)	<1.0- 4.9	<1.0- 18	9.6-44	3.4-9.7	<2.0- 18	Ecology and Herrera, 2010
WWTP effluent	Winter	2009	ng/L	10	12	35-194 (73.5)	<1.0- 3.6	<1.5- 16	11-52	2.1-10	<2.0	Ecology and Herrera, 2010
WWTP effluent	Spring	2016	ng/L	5	12	42-107 (69)	2.2-7.1	5.5-28	12-36	2.2-5.5	<2- 3.4	Ecology, 2017
WWTP effluent	Fall	2016	ng/L	5	12	42-125 (71)	1.6-7.1	6.1-57	10.5-49	2.6-3.7	<2.0-14	Ecology, 2017
Sediment	Freshwater (0-2 cm)	2013	ng/g dw	3	12	0.4-7.0 (2.35)	<0.2	<0.2	<0.2	<0.2	<0.4	Ecology, 2013
Sediment	Marine (0-3 cm)	2013-2015	ng/g dw	101	12		<0.1	<0.1	<0.1	<0.1	<0.2	EIM database*
Freshwater fish	Fillet - mult. sp.	2008	ng/g ww	15	10	<10- 76 (<10)			<5.0	<5.0	<5.0	Ecology, 2010
Freshwater fish	Liver - mult. sp.	2008	ng/g ww	15	10	<25- 527 (48)			<10.0	<10.0	<10.0	Ecology, 2010
Freshwater fish	Fillet - CC	2011	ng/g ww	4	12	2.1-21.5 (12)	<loq< td=""><td><loq< td=""><td><loq< td=""><td><0.3</td><td><loq< td=""><td>Ecology, 2012</td></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td><loq< td=""><td><0.3</td><td><loq< td=""><td>Ecology, 2012</td></loq<></td></loq<></td></loq<>	<loq< td=""><td><0.3</td><td><loq< td=""><td>Ecology, 2012</td></loq<></td></loq<>	<0.3	<loq< td=""><td>Ecology, 2012</td></loq<>	Ecology, 2012
Freshwater fish	Whole body - LSS	2011	ng/g ww	4	12	3.3-92 (23)	<loq< td=""><td><loq.< td=""><td><loq< td=""><td><0.2-0.6</td><td><loq< td=""><td>Ecology, 2012</td></loq<></td></loq<></td></loq.<></td></loq<>	<loq.< td=""><td><loq< td=""><td><0.2-0.6</td><td><loq< td=""><td>Ecology, 2012</td></loq<></td></loq<></td></loq.<>	<loq< td=""><td><0.2-0.6</td><td><loq< td=""><td>Ecology, 2012</td></loq<></td></loq<>	<0.2- 0.6	<loq< td=""><td>Ecology, 2012</td></loq<>	Ecology, 2012
Freshwater fish	Fillet - mult. sp.	2016	ng/g ww	22	12	<1-87 (3.9)	<0.5	<0.5- 1.8	<0.5	<0.5	<1.0	Ecology, 2017
Freshwater fish	Liver - mult. sp.	2016	ng/g ww	22	12	5.1-394 (20)	<1.0	<0.5-<2.9	<0.5- 2.5	<0.5- 1.1	<0.9- 6.2	Ecology, 2017
Osprey	Egg	2008	ng/g fw	11	13	37.5-910 (91)	<0.5	<0.5	<0.5- 0.8	<0.5- 0.8	<0.5	Ecology, 2010
Osprey	Egg	2016	ng/g fw	11	12	12-820 (100)	<0.5	<0.4- 1.8	<0.5	<0.5	<1.0	Ecology, 2017

Commented [A49]: Please include values for the mean and the standard deviation. Please include information on how many samples were collected at each site and analyzed and how many repeats were run for each sample.

* accessed from Ecology's Environmental Information Management Database on 3/21/2017 at: http://www.ecy.wa.gov/eim/

Sample Matrix	Sample Type	Collection Year	Units	n	PFOA	PFNA	PFDA	PFUnA	PFDoA	PFHxS	PFOS	PFDS	PFOSA	Ref.
Surface water	Spring	2008	ng/L	14	<1.0- 96	<0.1- 17	<0.1- 4.9			<1.0- 3.3	<0.1- 6.5	<1.0		Ecology, 2010
Surface water	Fall	2008	ng/L	14	<0.5- 48	<0.5- 7.0	<0.1- 3.8			<0.5- 4.5	<0.5- 7.6	<0.1- 1.3		Ecology, 2010
	Spring/summer/ fall/winter mean	2009-2010	ng/L	13	NR	NR	NR	NR	NR	NR	NR	NR		Dinglasan-Panlilio et al., 2014
Surface water	Spring	2016	ng/L	15	<1.0- 42.5	<1.0- 5.2	<1.0- 1.9	<1.0-<1.2	<1.0-<1.2	<2.0- 5.3	<2.0- 9.2		<1.0-<2.6	Ecology, 2017
Surface water	Fall	2016	ng/L	15	<1.0-55	<1.0- 5.8	<1.0- 3.2	<1.0-<1.1	<1.0-<1.1	<2.0- 3.0	<2.0- 12.5		<1.0-<1.2	Ecology, 2017
WWTP effluent	Spring	2008	ng/L	4	16.5-128	3.6-18	3.6-13			1.3-16	3.9-31	<0.1		Ecology, 2010
WWTP effluent	Fall	2008	ng/L	4	22-63	5.7-14	3.7-13			2.2-12	9.4-18	<0.1-<0.5		Ecology, 2010
WWTP effluent	Summer	2009	ng/L	10	11-52.5	3.3-29	1.5-10	<1.0	<1.0	<2.0- 8.3	<2.0-55		<2.5	Ecology and Herrera, 2010
WWTP effluent	Winter	2009	ng/L	10	11-70	1.4-134	1.4-7.9	<1.0	<1.0	<1.9- 6.9	<2.0- 19.5		<1.0- 2.0	Ecology and Herrera, 2010
WWTP effluent	Spring	2016	ng/L	5	7.2-20	<1.0- 1.9	<1.0- 4.9	<1.0	<1.0	<2.0-11	2.6-16		<2.5- 2.8	Ecology, 2017
WWTP effluent	Fall	2016	ng/L	5	6.6-18	<1.0- 4.0	<1.0- 5.0	<1.0	<1.0	<2.0- 7.1	<2.0- 6.5		<1.0	Ecology, 2017
Sediment	Freshwater (0-2 cm)	2013	ng/g dw	3	<0.1- 0.6	<0.1-0.4	<0.1- 1.5	0.2-0.7	0.2-0.9	<0.4	<0.2- 2.6		<0.1- 0.3	Ecology, 2013
Sediment	Marine (0-3 cm)	2013-2015	ng/g dw	101	<0.1- 0.21	<0.1	<0.1- 0.14	<0.1- 0.2	<0.1- 0.2	<0.2	<0.2- 1.6		<0.1- 0.3	EIM database*
Freshwater fish	Fillet - mult. sp.	2008	ng/g ww	15	<5.0	<5.0	<5.0- 7.5	<5.0- 7.2	<5	<5.0	<10.0- 76			Ecology, 2010
Freshwater fish	Liver - mult. sp.	2008	ng/g ww	15	<10.0	<10.0	<21.0- 25	<10.0- 46	<10.0- 21	<10.0	<10.0-527			Ecology, 2010
Freshwater fish	Fillet - CC	2011	ng/g ww	4	<0.25	<0.3	<0.25- 1.2	<0.25- 1.3	<0.3- 1.8	<loq< td=""><td>2.1 - 20</td><td></td><td><0.4</td><td>Ecology, 2012</td></loq<>	2.1 - 20		<0.4	Ecology, 2012
Freshwater fish	Whole body - LSS	2011	ng/g ww	4	<0.2- 0.8	<0.2- 1.6	0.3-10	<0.24- 20	<0.2- 9.5	<loq< td=""><td>2.9 - 46</td><td></td><td><0.3-3.4</td><td>Ecology, 2012</td></loq<>	2.9 - 46		<0.3- 3.4	Ecology, 2012
Freshwater fish	Fillet - mult. sp.	2016	ng/g ww	22	<0.5	<0.5- 0.9	<0.5- 5.5	<0.5-5.5	<0.5- 6.0	<1.0	<1.0- 74		<0.5	Ecology, 2017
Freshwater fish	Liver - mult. sp.	2016	ng/g ww	22	<0.7	<0.5- 7.3	<0.5- 20	<0.5-26	<0.5- 17	<1	1.4-336		<0.5- 4.9	Ecology, 2017
Osprey	Egg	2008	ng/g fw	11	<0.2-<1.0	<0.5- 6.4	2.0-10	3.5-13	<5.0-11	<0.5- 1.8	24-884	<1.0- 5.8		Ecology, 2010
Osprey	Egg	2016	ng/g fw	11	<0.5	<0.5- 5.7	1.0-47	1.1-45	0.6-47	<1.0	9.1-675		<0.5	Ecology, 2017

* accessed from Ecology's Environmental Information Management Database on 3/21/2017 at: http://www.ecy.wa.gov/eim/

References

Alder, A.C. and J. van der Voet, 2015. Occurrence and point source characterization of perfluoroalkyl acids in sewage sludge. Chemosphere, vol. 129: 62-73.

Ahrens, L., K. Norstrom, T. Vikton, A.P. Cousins, and S. Josefsson, 2015. Stockholm Arlanda Airport as a source of per- and poly-fluoroalkyl substances to water, sediment and fish. Chemosphere, Vol. 129: 33-38.

Anderson, R.H., G.C. Long, R.C. Porter, and J.K. Anderson, 2016. Occurrence of select perfluoroalkyl substances at U.S. Air Force aqueous film-forming foam release sites other than fire-training areas: Field-validation of critical fate and transport properties. Chemosphere, Vol. 150: 678-685.

Appleman, T.D., C.P. Higgins, O. Quinones, B.J. Vanderford, C. Kolstad, J.C. Zeigler-Holady, and E.R.V. Dickenson, 2014. Treatment of poly- and per-fluoroalkyl substances in full-scale water treatment systems. Water Research, Vol. 51: 246-255.

Barzen-Hanson, K.A., S.C. Roberts, S. Choyke, K. Oetjen, A. McAlees, N. Riddell, R. McCrindle, P. Lee Ferguson, C.P. Higgins, and J.A. Field, 2017. Discovery of 40 Classes of Per- and Polyfluoroalkyl Substances in Historical Aqueous Film-Forming Foams (AFFFs) and AFFF-Impacted Groundwater. Environmental Science and Technology, DOI: 10.1021/acs.est.6b05843.

Baygi, S.F., B.S. Crimmins, P.K. Hopke, and T.M. Holsen, 2016. Comprehensive Emerging Chemical Discovery: Novel Polyfluorinated Compounds in Lake Michigan Trout. Environmental Science and Technology, Vol. 50: 9460-9468.

Busch, J., L. Ahrens, R. Sturm, and R. Ebinghaus, 2010. Polyfluoroalkyl compounds in landfill leachates. Vol. 158: 1467-1471.

Chu, S., R.J. Letcher, D.J. McGoldrick, and S.M. Backus, 2016. A New Fluorinated Surfactant Contaminant in Biota: Perfluorobutane Sulfonamide in Several Fish Species. Environmental Science and Technology, Vol. 50: 669-675.

Cousins, I.T., R. Vestergren, Z. Wang, M. Scheringer, and M.S. McLachlan, 2016. The precautionary principle and chemicals management: The example of perfluoroalkyl acids in groundwater. Environment International, Vol. 94: 331-340.

Dinglasan-Panlilio, M.J., S.S. Prakash, and J.E. Baker, 2014. Perfluorinated compounds in the surface waters of Puget Sound, Washington and Clayoquot and Barkley Sounds, British Columbia. Marine Pollution Bulletin, Vol. 78: 173-180.

Ecology, 2010. Perfluorinated Compounds in Washington Rivers and Lakes. Washington State Department of Ecology, Olympia, WA. Publication Number: 10-03-034. https://fortress.wa.gov/ecy/publications/summarypages/1003034.html

Ecology, 2012. PBTs Analyzed in Bottom Fish from Four Washington Rivers and Lakes: Hexabromocyclododecane, Tetrabromobisphenol A, Chlorinated Paraffins, Polybrominated Diphenylethers, Polychloinated Napthalaenes, Perfluorinated Organic Compounds, Lead, and Cadmium. Washington State Department of Ecology, Olympia, WA. Publication Number 12-03-042. <u>https://fortress.wa.gov/ecy/publications/summarypages/1203042.html</u>

Ecology, 2013. PBT Chemical Trends in Washington State Determined from Age-Dated Lake Sediment Cores, 2012 Sampling Results. Washington State Department of Ecology, Olympia, WA. Publication Number: 13-03-036.

https://fortress.wa.gov/ecy/publications/SummaryPages/1303036.html

Ecology, 2014. Pharmaceuticals, Personal Care Products, and Perfluoroalkyl Substances in Elliott Bay Sediments: 2013 Data Summary. Washington State Department of Ecology, Olympia, WA. Publication Number: 14-03-049. https://fortress.wa.gov/ecy/publications/SummaryPages/1403049.html

Ecology, 2016a. Moses Lake Port Aircraft Rescue and Firefighting School – VCP Opinion on Site Cleanup – No Further Action.

Ecology, 2016b. Quality Assurance Project Plan: Statewide Survey of Per- and Poly-fluoroalkyl Substances in Washington State Rivers and Lakes. Washington State Department of Ecology, Olympia, WA. Publication Number 16-03-110. https://fortress.wa.gov/ecy/publications/SummaryPages/1603110.html

Ecology, 2017. Survey of Per- and Poly-fluoroalkyl Substances in Rivers and Lakes, 2016. Washington State Department of Ecology, Olympia, WA. Publication Number 17-03-021. [publication link to be added by late September]

Ecology and Herrera, 2010. Control of Toxic Chemicals in Puget Sound – Summary Technical Report for Phase 3: Loadings from POTW Discharge of Treated Wastewater. Washington State Department of Ecology, Olympia, WA. Publication Number: 10-10-057. https://fortress.wa.gov/ecy/publications/SummaryPages/1010057.html

Ellis, D.A., J.W. Martin, A.O. De Silva, S.A. Mabury, M.D. Hurley, M.P. Sulbaek Andersen, and T.J. Wallington, 2004. Degradation of Fluorotelomer Alcohols: A Likely Atmospheric Source of Perfluorinated Carboxylic Acids. Environmental Science and Technology, Vol. 38: 3316-3321.

Eriksson, U., A. roos, Y. Lind, K. Hope, A. Ekblad, and A. Karrman, 2016. Comparison of PFASs contamination in the freshwater and terrestrial environments by analysis of eggs from osprey (Pandion haliaetus), tawny owl (Strix aluco), and common kestrel (Falco tinnunculus). Environmental Research, Vol. 149: 40-47.

Gewurtz, S.B., S.P. Bhavsar, P.W. Crozier, M.L. Diamond, P.A. Helm, C.H. Marvin, and E.J. Reiner, 2009. Perfluoroalkyl Contaminants in Window Film: Indoor/Outdoor, Urban/Rural, and Winter/Summer Contamination and Assessment of Carpet as a Possible Source. Environmental Science and Technology, Vol. 43: 7317-4323.

Gewurtz, S.B., S.P. Bhavsar, S. Petro, C.G. Mahon, X. Zhao, D. Morse, E.J. Reiner, S.A. Tittlemier, E. Braekevelt, and K. Drouillard, 2014. High levels of perfluoroalkyl acids in sport fish species downstream of a firefighting training facility at Hamilton International Airport, Ontario, Canada. Environment International, Vol. 67: 1-11.

Giesy, J.P. and K. Kannan, 2001. Global Distribution of Perfluorooctane Sulfonate in Wildlife. Environmental Science and Technology, Vol. 35: 1339-1342.

Higgins, C.P. and R.G. Luthy, 2006. Sorption of Perfluorinated Surfactants on Sediments. Environmental Science and Technology, Vol. 40: 7251-7256.

Houde, M., J.W. Martin, R.J. Letcher, K.R. Solomon, and D.C.G. Muir, 2006. Biological Monitoring of Polyfluoroalkyl Substances: A Review. Environmental Science and Technology, Vol. 40: 3463-3473.

Houde, M., A.O. De Silva, D.C.G. Muir, and R.J. Letcher, 2011. Monitoring of Perfluorinated Compounds in Aquatic Biota: An Updated Review. Environmental Science and Technology, Vol. 45: 7962-7973.

Houtz, E.F., R. Sutton, J-S Park, and M. Sedlak, 2016. Poly- and perfluoroalkyl substances in wastewater: Signficance of unknown precursors, manufacturing shifts, and likely AFFF impacts. Water Research, Vol. 95: 142-149.

Kim, S.K. and K. Kannan, 2007. Perfluorinated Acids in Air, Rain, Snow, Surface Runoff, and Lakes: Relative Importance of Pathways to Contamination of Urban Lakes. Environmental Science and Technology, Vol. 41: 8328-8334.

Lang, J.R., B. McKay Allred, J.A. Field, J.W. Levis, and M.A. Barlaz, 2017. National Estimate of Per- and Polyfluoroalkyl Substance (PFAS) Release to U.S. Municipal Landfill Leachate. Environmental Science and Technology, Vol. 51: 2197-2205.

Lanza, H.A., R.S. Cochran, J.F. Mudge, A.D. Olson, B.R. Blackwell, J.D. Maul, C.J. Salice, and T.A. Anderson, 2016. Temporal Monitoring of Perfluorooctane Sulfonate Accumulation in Aquatic Biota Downstream of Historical Aqueous Film Forming Foam Use Areas. Environmental Toxicology, Vol. 9999: 1-8.

Lau, C., K. Anitole, C. Hodes, D. Lai, A. Pfahles-Hutchens, and J. Seed, 2007. Perfluoroalkyl Acids: A Review of Monitoring and Toxicological Findings. Toxicological Sciences, Vol. 99: 366-394.

Martin, J.W., D.A. Ellis, and S.A. Mabury, 2006. Atmospheric Chemistry of Perfluoroalkanesulfonamides: Kinetic and Product Studies of the OH Radical and Cl Atom Initiated Oxidation of *N*-Ethyl Perfluorobutanesulfonamide. Environmental Science and Technology, Vol. 40: 864-872.

MDEQ, 2015. Reconnaissance Sampling of Perfluorinated Compounds in Michigan Surface Waters and Fish 2010-2014. Michigan Department of Environmental Quality, Water Resources Division, Publication No. MI/DEQ/WRD-15/019.

Munoz, G., M. Desrosiers, S. Vo Duy, P. Labadie, H. Budzinski, J. Liu, and S. Sauve, 2017. Environmental Occurrence of Perfluoroalkyl Acids and Novel Fluorotelomer Surfactants in the Freshwater Fish Catostomus commersonii and Sediments Following Firefighting Foam Deployment at the Lac-Mégantic Railway Accident. Environmental Science and Technology, Vol. 51: 1231-1240.

Muller, C.E., N. Spiess, A.C. Gerecke, M. Scheringer, and K. Hungerbuhler, 2011. Quantifying Diffuse and Point Inputs of Perfluoroalkyl Acids in a Nonindustrial River Catchment. Environmental Science and Technology, Vol. 45: 9901-9909.

Molina, E.D., R. Balander, S.D. Fitzgerald, J.P. Giesy, K. Kannan, R. Mitchell, and S.J. Bursian, 2006. Effects of air cell injection of perfluorooctane sulfonate before incubation on development of the white leghorn chicken (*Gallus domesticus*) embryo. Environmental Toxicology and Chemistry, Vol. 25: 227-232.

Newsted, J.L., P.D. Jones, K. Coady, and J.P. Giesy, 2005. Avian Toxicity Reference Values for Perfluorooctane Sulfonate. Environmental Science and Technology, Vol. 39: 9357-9362.

Newton, S., R. McMahen, J.A. Stoeckel, M. Chislock, A. Lindstrom, and M. Strynar, 2017. Novel Polyfluorinated Compounds Identified Using High Resolution Mass Spectrometry Downstream of Manufacturing Facilities near Decatur, Alabama. Environmental Science and Technology, Vol. 51: 1544-1552.

OECD, 2013. OECD/UNEP Global PFC Group, Synthesis paper on per- and polyfluorinated chemicals (PFCs), Environment, Health and Safety, Environment Directorate, OECD.

Peden-Adams, M.M., J.E. Stuckey, K.M. Gaworecki, J. Berger-Ritchie, K. Bryant, P.G. Jodice, T.R. Scott, J.B. Ferrario, B. Guan, C. Vigo, J.S. Boone, W.D. McGuinn, J.C. DeWitt, and D.E. Keil, 2009. Developmental toxicity in white leghorn chickens following *in ovo* exposure to perfluorooctane sulfonate (PFOS). Reproductive Toxicology, Vol. 27: 307-318.

Prevedouros, K., I.T. Cousins, R.C. Buck, and S.H. Korzeniowski, 2006. Sources, Fate and Transport of Perfluorocarboxylates. Environmental Science and Technology, Vol. 40: 32-44.

Sepulvado, J.G., A.C. Blaine, L.S. Hundal, and C.P. Higgins, 2011. Occurrence and Fate of Perfluorochemicals in Soil Following the Land Application of Municipal Biosolids. Environmental Science and Technology, Vol. 45: 8106-8112.

Shi, Y., R. Vestergren, Z. Zhou, X. Song, L. Xu, Y. Liang, and Y. Cai, 2015. Tissue Distribution and Whole Body Burden of the Chlorinated Polyfluoroalkyl Ether Sulfonic Acid F-35B in Crucian Carp (Carassius carassius): Evidence for a Highly Bioaccumulative Contaminant of Emerging Concern. Environmental Science and Technology, Vol. 49: 14156-14165.

Sinclair, E. and K. Kannan, 2006. Mass Loading and Fate of Perfluoroalkyl Surfactants in Wastewater Treatment Plants.

Stock, N.L., F.K. Lau, D.A. Ellis, J.W. Martin, D.C.G. Muir, and S.A. Mabury, 2004. Polyfluorinated Telomer Alcohols and Sulfonamides in the North American Troposphere. Environmental Science and Technology, Vol. 38: 991-996.

Venkatesan, A.K. and R.U. Halden, 2013. National inventory of perfluoroalkyl substances in archived U.S. biosolids from the 2001 EPA National Sewage Sludge Survey. Journal of Hazardous Materials, Vol. 252-253: 413-418.

Wang, Y., R. Vestergren, Y. Shi, D. Cao, L. Xu, Y. Cai, X. Zhao, and F. Wu, 2016. Identification, Tissue Distribution, and Bioaccumulation Potential of Cyclic Perfluorinated Sulfonic Acids Isomers in an Airport Impacted Ecosystem. Environmental Science and Technology, Vol. 50: 10923-10932.

Xiao, F., J.S. Gulliver, and M.F. Simcik, 2013. Perfluorooctane sulfonate (PFOS) contamination of fish in urban lakes: A prioritization methodology for lake management. Water Research, Vol. 47: 7264-7272.

Young, C.J., V.I. Furdui, J. Franklin, R.M. Koerner, D.C.G. Muir, and S.A. Mabury, 2007. Perfluorinated Acids in Arctic Snow: New Evidence for Atmospheric Formation. Environmental Science and Technology, Vol. 41: 3455-3461.

Zushi, Y. and S. Masunaga, 2009. Identifying the nonpoint source of perfluorinated compounds using a geographic information system based approach. Environmental Toxicology and Chemistry, Vol. 28: 691-700.

Add:

Benskin, J.P., B. Li, M.G. Ikonomou, J.R. Grace, and L.Y. Li. 2012. Per- and polyfluoroalkyl substances in landfill leachate: Patterns, time trends, and sources. *Environ. Sci. Technol.* 46(21):11532-11540.

Bossi, R., J. Strand, O. Sortkjaer, and M.M. Larson. 2008. Perfluorinated compounds in Danish wastewater treatment plants and aquatic environments. *Environ. Int.* 34(4):443–450.

Busch, J., L. Ahrens, R. Sturm, and R. Ebinghaus. 2010. Polyfluoroalkyl compounds in landfill leachates. *Environ. Pollut.* 158:1467–1471.

Clara, M., S. Scharf, S. Weiss, O. Gans, and C. Scheffknecht. 2008. Emissions of perfluorinated alkylated substances (PFAS) from point sources-identification of relevant branches. *Water Sci. Technol.* 58(1):59–66.

Kallenborn, R., U. Berger, and U. Järnberg. 2004. Perfluorinated alkylated substances (PFAS) in the Nordic environment. TemaNord 2004:552. Nordic Council of Ministers, Copenhagen, Denmark. 112 pp.

Lang, J.R., B. Allred, J. Field, J. Levis, M. Barlaz. 2017. National estimate of per- and polyfluoroalkyl substance (PFAS) release to U.S. municipal landfill leachate. *Environ. Sci. Technol.* 51:2197-2205.

Li, B., M.N. Dannon-Schaffer, L.Y. Li, M.G. Ikonomou, and J.R. Grace. 2012. Occurrence of PFCs and PBDEs in landfill leachates from across Canada. *Water Air Soil Pollut*. 223:3365–3372.

Murakami, M., E. Imamura, H. Shinohara, K. Kiri, Y. Muramatsu, A. Harada, and H. Takada. 2008. Occurrence and sources of perfluorinated surfactants in rivers in Japan. *Environ. Sci. Technol.* 42(17):6566-6572.

Schultz, M. M., C.P. Higgins, C.A. Huset, R.G. Luthy, D.F. Barofsky, and J.A. Field. 2006. Fluorochemical mass flows in a municipal wastewater treatment facility. *Environ. Sci. Technol.* 40(23):7350–7357.

Sinclair, E., and K. Kannan. 2006. Mass loading and fate of perfluoroalkyl surfactants in wastewater treatment plants. *Environ. Sci. Technol.* 40(5):1408–1414.

Woldegiorgis, A., J. Anderson, M. Remberger, L. Kaj, Y. Ekheden, L. Blom, E. Brorström-Lundén, A. Borgen, C. Dye, and M. Schlabach. 2006. Results from the National Swedish Screening Programme 2005. Subreport 3: Perfluorinated alkylated substances (PFAS). IVL Report B1698. IVL Swedish Environmental Research Institute Ltd., Stockholm, Sweden. 48 pp. November 20.

Zushi, Y., T. Takeda, and S. Masunaga. 2007. Existence of nonpoint source of perfluorinated compounds and their loads in the Tsurumi River basin, Japan. *Chemosphere*. 71(8):1566-1573.