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Agenda Item 7.b.
Meeting of July 25, 2023

CITY OF SANIBEL
PLANNING COMMISSION RESOLUTION 23-24

A RESOLUTION OF THE PLANNING COMMISSION OF THE CITY OF SANIBEL; RELATING TO A DEVELOPMENT PERMIT (APPLICATION NO. SPLT-2022-000074 AND DP-2021-001803) FILED PURSUANT TO SECTIONS 82-421(1), 82-422, AND 114-106 OF THE LAND DEVELOPMENT CODE FOR PRELIMINARY PLAT, TO ALLOW A UNIFIED RESIDENTAIL HOUSING (CLUSTER HOUSING) DEVELOPMENT INCLUDING SIX PARCELS FOR SINGLE-FAMILY RESIDENTIAL USE AND ASSOCIATED IMPROVEMENTS, KNOWN AS "COASTAL CREEK" SUBDIVISION, ON PROPERTY OWNED BY BUCKINGHAM 225 DEVELOPMENT, INC. (DANIEL W. DODRILL), AND LOCATED AT 5301-5326 SANIBEL CAPTIVA ROAD, PARCEL NO. 13-46-21-T2-00002.2000 AND 13-46-21-T2-00002-4000, MORE FULLY DESCRIBED HEREIN; AND PROVIDING AN EFFECTIVE DATE.

WHEREAS, Sections 82-421(1) and 82-422 of the Land Development Code detail the application and notice requirements for development permit consideration by the Planning Commission; and Section 114-106 of the Land Development Code provides for requirements and procedures for preliminary plats; and

WHEREAS, Buckingham 225 Development, Inc. (Daniel W. Dodrill), owner of the property located at 5301-5326 Sanibel Captiva Road, has authorized Brian Smith, Ensite, Inc., to submit Development Application No. DP-2021-001803 and Major Subdivision Plat Application No. SPLT-2022-000074 (the "subject applications") to allow for a Unified Residential Housing (Cluster Housing) development including six parcels for single-family residential use and associated improvements, known as "Coastal Creek" subdivision; and

WHEREAS, the subject applications were initially approved by Sanibel Planning Commission Resolution 23-001; however, a timely appeal of Planning Commission Resolution 23-001 was filed pursuant to Land Development Code Section 82-98 – Appeals; and City Council, after consideration of the appeal, remanded the subject applications back to Planning Commission for a new hearing by Sanibel City Council Resolution 23-024; and

WHEREAS, the applicant has reiterated its requests for approval of the subject applications; and

WHEREAS, a duly noticed public hearing of the subject applications was held before the Planning Commission on July 25, 2023; and

WHEREAS, after providing the applicant, staff, and the public an opportunity to present testimony and evidence, and having reviewed the record, and all applicable sections of the Land Development Code including Section 86-40(b), the Planning Commission finds that the criteria for granting the applications have been met and that the application should therefore be approved.

NOW, THEREFORE, BE IT RESOLVED, THE PLANNING COMMISSION OF THE CITY OF SANIBEL finds that Development Permit Application No. DP-2021-001803 and Major Subdivision Plat Application No. SPLT-2022-000074 to allow for a Unified Residential Housing (Cluster Housing) development including six parcels for single-family residential use and associated improvements, known as "Coastal Creek" subdivision on property owned by Buckingham 225 Development, Inc. (Daniel W. Dodrill), and located at 5301-5326 Sanibel Captiva Road, and more fully identified as Tax Parcel No. 13-46-21-T2-00002.2000 and 13-46-21-T2-00002.4000, is approved.

Any approval of this Development Permit Application is pursuant to the application and attachments included with these items, and subject to the following condition(s) contained in the July 25, 2023, staff report:

1. A copy of the approved preliminary plat is attached to this resolution as Exhibit "A". Preliminary plat approval shall be effective and valid for a period of three years. The planning commission may, however, extend this effective period for an additional two years, provided that, at the time such extension is granted, the preliminary plat continues in compliance with all requirements of this Land Development Code and the Sanibel Plan. If all requirements for approval of a final subdivision plat, and the recording of such plat, are not completed within the time period for which preliminary approval is valid and effective, such preliminary approval and all rights conferred thereby shall be terminated and expire.
2. Preliminary plat approval may be extended by city council for an additional period of time beyond the three-year effective time period and the two-year extension that may be approved by the planning commission due to the necessity of achieving compliance with federal, state or local regulations pertaining to endangered or protected species and respective habitat or due to other unforeseen environmental conditions.
3. The applicant shall submit for final approval, a final subdivision plat in accordance with the requirements of *Land Development Code Section 114-89 – Final plat review and approval*.
4. A copy of the approved Coastal Creek Site Development Plans is attached to this resolution as Exhibit "B".
5. All future land use and development shall comply with Exhibit "A" and "B", and all associated requirements of the Land Development Code including, but not limited to:
    a. Single-family residential and associated accessory uses
    b. Lot sizes
    c. Setbacks
    d. Limitations on vegetation removal and developed area
    e. Limitation on coverage with impermeable surfaces
    f. Minimum distance between buildings
    g. Environmental performance standards
6. The applicant shall execute, and record in the public records of Lee County, a declaration that the residential density allocation for the property has been fully utilized. If the development is to be phased and only a portion of the residential density allocated to the property is to be utilized, then the declaration shall reflect that portion so utilized. The declaration shall be prepared, executed and recorded in accordance with the formalities required under state law for the conveyance of real property, for which the developer shall be responsible. Completion permits shall not be issued for any dwelling units in the development and the subdivision shall not be final until such time as the developer shows compliance with this section.
7. The applicant shall establish a legal entity (a property owner's association) with responsibility for complying with the requirements of this development permit and maintaining the private road, drainage system, utilities, landscape buffers, and wetland preservation. Restrictive covenants for the subdivision are to include the following provisions:
    a. A provision that requires compliance with the conditions of development permit DP-2021-1803;
    b. A provision that prohibits any dwelling unit in the subdivision from use as resort housing;
    c. A provision requiring the Association to maintain the private street, stormwater management system, utilities, landscape buffers, and wetland preservation as conditions of approval for the preliminary plat and construction plans. The Association shall have the authority to make assessments and enforce liens to

cover the costs and assessments;

d. A provision giving the City of Sanibel the right to enforce the Associations obligations to maintain the private street, the stormwater management system, utilities, landscape buffers, and wetland preservation required as conditions of approval for the preliminary plat and construction plans;

e. A provision authorizing the City of Sanibel to make repairs and perform maintenance on required facilities, if not done by the Association, and giving the City the right to make assessments and enforce liens for costs of repairs and maintenance;

f. A provision providing a notice to lot purchasers that the street within the subdivision is not a public street, is to be maintained by the owners at their expense and is intended to remain a private street in perpetuity;

g. A provision that specifies as to each lot how many dwelling units are permitted thereon, including the amount of developed area and coverage assigned to each single-family lot;

h. A provision requiring the applicant to provide each property purchase/owner with information about the common open space and preservation areas, including Tracts B, C, D-1, and D-2; and

i. A provision in the amendment section of the documents prohibiting amendments to the preceding provisions without the written jointer or consent of the City of Sanibel.

8. Preliminary plat approval shall not relieve the applicant from the requirement of obtaining permits from and complying with lawful requirements imposed by the Florida Department of Environmental Protection, South Florida Water Management District, and any other applicable local, State and Federal law.
9. The applicant shall install all monuments as required by Chapter 177, Florida Statutes, and construct all streets, street signs, drainage facilities, sewage treatment facilities, and other improvements as are necessary to bring the proposed subdivision in full compliance with the following requirements of the Land Development Code:
    a. All streets and other improvements in proposed subdivisions shall be constructed in accordance with all specifications as provided in this chapter and as may be adopted by the city council by resolution as "Subdivision Improvement Construction Requirements;"
    b. All necessary street signs shall be installed by the applicant in accordance with the prevailing scheme of identifying public and private streets in the city; and
    c. All necessary drainage facilities shall be constructed as required and approved by the city manager so as to meet the minimum requirements of this Land Development Code and as to accomplish the intent and purpose of this Land Development Code.
10. The applicant shall obtain formal approval of the proposed subdivision and street names from the City in accordance with Ordinance No. 86-27 and City Council Resolution No. 86-67 prior to final plat approval.
11. The applicant may not begin construction of required improvement until construction plans are approved by the City Manager and a construction bond with good and sufficient surety, an irrevocable letter of credit, or equivalent security is provided to the City in an amount equal to the cost of all required improvements, and in a form approved by the City Attorney, conditioned upon satisfactory completion of required improvements in accordance with he approved construction plans during the effective period of the preliminary plat approval.
12. No facility installed underground may be covered over until the facility has been inspected by the City Manager. Other periodic inspections may be required. Construction of required improvements shall not be complete unless and until all the improvements have been finally inspected and approved by the City Manager.
13. Prior to the commencement of development activity, the applicant shall obtain a

vegetation permit to approve revisions of the vegetation plan.

14. A native vegetation buffer shall be installed along the north property line, pursuant to LDC Chapter 122, Article II, Division 3 – Residential Development Along Arterial and Collector Roads.
15. The proposed native plant buffer along the western boundary shall be permitted to mature to the natural height and growth pattern of the plants and maintained in perpetuity to prevent encroachment into the adjacent city-owned preserve.
16. Pursuant to Sec. 122-169(3) and Sec. 122-191, all City-listed and Florida Invasive Species Council (formerly FLEPPC) listed plant species shall be removed from the site and maintained free in perpetuity.
17. The applicant shall submit a copy of the SFWMD preserve management plan annual monitoring reports to the City.
18. The trimming of mangroves or other vegetation within Tract C and Tract D-2 is strictly prohibited, except to access utilities. The Coastal Creek subdivision does not abut the Heron's Landing Lake; property owners do not have riparian rights and are not entitled to a view of the waterbody.
19. All landscaping within the Coastal Creek subdivision shall be 100% native except the six platted lots which are required to be at least 75% native; the use of sod is prohibited.
20. The use of fertilizer containing nitrogen and phosphorus is prohibited within the Coastal Creek subdivision.
21. All new vegetation within stormwater retention areas, including lands between designated home sites, shall comply with Sec. 118-286 – Planting. A revised vegetation plan shall be submitted detailing the native plants installed to vegetate the common area tracts to ensure compliance with this condition.
22. All soil excavated from the site shall be disposed of properly at an off-island location.
23. The applicant shall comply with all provisions of Chapter 86 – Site Preparation, including but not limited to:
    a. Development activities shall be designed to minimize the amount of fill used in preparation of the site. Soil and other materials shall not be temporarily nor permanently stored in locations which would result in the unnecessary destruction of vegetation. Excess soil, or other material, including dredged spoil, to be disposed of off-site, shall be deposited at specified locations in a manner causing minimal environmental damage.
    b. Erosion and sediment control measures shall be coordinated with the sequence of grading, development, and construction operations; and shall be maintained for the duration of construction until final landscaping has been installed.
    c. Removal of vegetation shall be limited to the minimum necessary to carry out development activity, except as required by other provisions of the Land Development Code. The removal of vegetation, by any means other than the use of hand-held tools, shall not occur prior to issuance of a development permit.
    d. Permanent vegetation shall be installed on the construction site as soon as utilities are in place and final grades are completed. This permanent vegetation must be cared for and maintained in a healthy condition.
    e. All on-site facilities shall be properly maintained by the owner so that they do not become nuisances. Nuisance conditions shall include but not be limited to:
        1. Improper storage resulting in uncontrolled runoff and overflow;
        2. Stagnant water with concomitant algae growth, insect breeding and odors;
        3. Discarded debris;
        4. Unnecessary noise; and
        5. Safety hazards created by the facility's operations.
    f. The design, location and construction and the maintenance of all development shall be in a manner that minimizes environmental damage. The developer shall completely restore any environmentally sensitive area or wetland area damaged

during construction. Complete restoration means that the damaged area shall, within two years, be operating as effectively as the natural system did before being destroyed or altered. In designing the site for its ultimate end use, the site shall be graded in such a manner, and development and use shall take place in such a manner, so that there are no point discharges into coastal waters resulting from stormwater runoff and/or from wastewater effluent.

24. Planning Commission recommends that "Tract C", as shown and described by Exhibit "A", approved as conservation use for the protection of wetlands, is designated for inclusion within the Environmentally Sensitive Lands Conservation District and map upon City Council consideration of an amendment to the Sanibel Plan.

EXPIRATION OF PLANNING COMMISSION ACTION; In accordance with Land Development Code Section 82-424(f), when a development order is approved with conditions imposed thereon, such conditions shall be satisfied within the time limit specified in the development order issued by the Planning Commission. When such conditions specify requirements to be completed before a development permit is issued, and no particular time limit is specified for satisfaction of the conditions, such conditions must be satisfied within six months after issuance of the development order. Failure to satisfy a condition imposed upon the approval of a development permit, within the time limit specified therefor, or such extended time period as the Planning Commission may approve upon timely application of the permittee, shall cause the development order approving the development permit to be null and void and of no further force or effect.

EFFECTIVE DATE OF PLANNING COMMISSION ACTION: In accordance with Land Development Code Section 82-97, all actions of the Planning Commission, including those which constitute final decisions, shall be effective upon the date of filing of the adopted Resolution with the City Manager, or at a later date if provided in the Resolution. However, permits authorized by final decisions shall not be issued until one of the following has occurred: 1) The time for filing an appeal to City Council has elapsed; 2) The applicant and all other persons having appeal rights have filed a written waiver of appeal rights; 3) If an appeal has been timely filed, the City Council has finally disposed of the matter.

RIGHT TO APPEAL PLANNING COMMISSION ACTION: In accordance with Land Development Code Section 82-98. Appeals. The applicant is hereby advised that the following persons have the right to appeal a final decision of the Planning Commission adverse to their interests: 1) The applicant; 2) The owner of the property proposed for development; 3) The developer of the property proposed for development; and 4) Any other person residing upon, or owning property within the City, or owning or operating a business within the City, who participated by written comment before or at the Planning Commission hearing or who participated in person or through an authorized agent at the Planning Commission hearing. The appeal shall be filed within 15 days after the date that the Planning Commission decision was filed. The appeal shall be filed with the City manager, and the filing fee shall be paid as a prerequisite to filing.

DISCLAIMER & PERMIT CONDITION (APPLICABLE ONLY IF FEDERAL OR STATE PERMITS ARE REQUIRED): Issuance of a development permit by the City does not create any right to obtain a permit from a State or Federal agency and does not create any liability on the part of the City for issuance of the permit if the applicant fails to obtain requisite approvals or fulfill the obligations imposed by a State or Federal agency or undertakes actions that result in a violation of State or Federal law. If applicable, all other State or Federal permits must be obtained before commencement of the project.

PASSED IN OPEN AND REGULAR SESSION OF THE PLANNING COMMISSION OF THE CITY OF SANIBEL, FLORIDA, THIS 25TH DAY OF JULY 2023.

Attest:

~~Scotty Lynn Kelly, City Clerk~~
Anna M. Hicks, Acting

Roger Grogman, Chair

Approved as to form and legality:

John D. Agnew, City Attorney

Date filed with City Clerk: 7/25/23

Vote of Commission Members:

Grogman	Aye
Pfeifer	Aye
DeBruce	Excused
Colter	Aye
Nichols	Aye
Welch	Excused
Symroski	Nay

CITY OF SANIBEL
RESOLUTION 23-049

A RESOLUTION OF THE CITY COUNCIL OF THE CITY OF SANIBEL, FLORIDA, UPHOLDING IN ALL RESPECTS THE PLANNING COMMISSION'S DECISION DETAILED IN SANIBEL PLANNING COMMISSION RESOLUTION 23-024 EXCEPT TO MODIFY CONDITION 22, REGARDING DEVELOPMENT PERMIT APPLICATION NO. DP-2021-001803 AND MAJOR SUBDIVISION APPLICATION NO. SPLT 2022-000074 DATED JULY 25, 2023, FOR THE PROJECT KNOWN AS COASTAL CREEK SUBDIVISION AND LOCATED AT 5301-5325 SANIBEL CAPTIVA ROAD; AND PROVIDING AN EFFECTIVE DATE.

WHEREAS, on July 25, 2023, the Sanibel Planning Commission conducted a quasi-judicial hearing for consideration of Development Permit Application No. DP-2021-001803 and Major Subdivision Application No. SPLT 2022-00007 (the "Applications"); and

WHEREAS, the Applications, filed by Brian Smith with Ensite, Inc. on behalf of owner Buckingham 225 Development, Inc. (the "Applicant"), seek approval to allow for a unified residential housing (cluster housing) development including six parcels for single-family residential use and associated improvements, known as "Coastal Creek" subdivision, located at 5301-5325 Sanibel Captiva Road, Sanibel; and

WHEREAS, Land Development Code Sections 82-421(8) and 82-422 detail the application and notice requirements for development permit consideration by the Planning Commission; and Land Development Code Section 114-106 provides the requirements and procedures for preliminary plats; and

WHEREAS, at the conclusion of its July 25, 2023 hearing, the Planning Commission, by a 4-1 vote, approved the Applications together with 24 conditions; and

WHEREAS, a Notice of Appeal of Planning Commission Resolution 23-024 was timely filed on August 8, 2023, by Ralph Brookes, Esq., on behalf of the Heron's Landing Homeowners Association of Sanibel, Inc. and six Sanibel residents (the "Appellants"), opposing the approval of the applications with the 24 conditions, as approved; and

WHEREAS, pursuant to Land Development Code Section 82-98(d), the City Council's consideration on appeal is limited to whether the Planning Commission properly interpreted and applied the provisions of the Land Development Code, based upon the Applications and evidence presented to the Planning Commission; and

WHEREAS, on appeal, the Appellants argue, among other things, that the Coastal Creek site should have certain additional testing and site remediation performed before the site is developed, as approved; and

WHEREAS, the City Council conducted a duly noticed hearing on appeal on September 11, 2023, for which the City Council reviewed and considered the record before the Planning Commission, Planning Commission Resolution 23-024, minutes of the Planning Commission, and the written and oral arguments presented by or on behalf of the Appellants, the Applicant, and Planning Staff; and

NOW, THEREFORE, BE IT RESOLVED BY THE CITY COUNCIL OF THE CITY OF SANIBEL, FLORIDA:

SECTION 1. The above "whereas" recitals are hereby found to be correct and are incorporated herein as part of this Resolution.

SECTION 2. The City Council makes the following findings:

(a) The Appellants have standing to appeal and timely filed their appeal to City Council; and

(b) All parties had notice and opportunity to be heard in the appeal; and

(c) No objections were raised to form or procedure of the appeal; and

(d) In approving the Applications, the Planning Commission properly interpreted and applied the provisions of the Land Development Code, based upon the Applications and evidence presented to the Planning Commission; and

(e) Condition 22 of Planning Commission Resolution 23-024 should be modified to clarify the definition of "excavated soil."

SECTION 3. Planning Commission Resolution 23-024 is hereby **UPHELD** in all respects, except Condition 22 is **MODIFIED** to read as follows:

> 22. All excavated soil from the site shall be disposed of properly at an off-island location. For this project, "excavated soil" shall include all soil dug up or scraped from predevelopment grade, other than *de minimus* removal of soil for the planting of vegetation or other, similar minor soil disturbances.

SECTION 4. This resolution shall take effect immediately upon adoption.

PASSED IN OPEN AND REGULAR SESSION OF THE CITY COUNCIL OF THE CITY OF SANIBEL, FLORIDA THIS 3RD DAY OF OCTOBER 2023.

Attest:

Scotty Lynn Kelly, City Clerk

Richard Johnson, Mayor

Approved as to form and legality:

John D. Agnew, City Attorney

Date filed with City Clerk: October 3, 2023

Vote of Council Members:

Johnson	Aye
Miller	Aye
Crater	Nay
Henshaw	Aye
Smith	Aye

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PFOS in U.S. Ambient Surface Waters: A Review of Occurrence in Aquatic Environments and Comparison to Global Concentrations

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Abstract

PFOS is one of the dominant PFAS detected in aquatic ecosystems. PFOS has been used in a wide range of industrial and consumer products for decades. The unique properties of PFOS, including its stability and resistance to degradation, have made it highly persistent in the aquatic environment. Due to its persistence, potential toxicity, and occurrence in aquatic ecosystems, interest in PFOS has increased in recent decades. Despite this interest, current information on the environmental distribution of PFOS in ambient surface waters of the United States (U.S) is fairly limited. This critical review summarizes currently available literature on PFOS occurrence in surface waters across the U.S. and highlights existing data gaps. Available data are largely from a handful of study areas with known PFAS manufacturing or industrial uses, with much of the data collected from freshwater systems in eastern states and the upper midwest. Measured PFOS concentrations in surface waters vary widely, over eight orders of magnitude, with the highest concentrations occurring downstream from manufacturing and industrial use plants, areas near aqueous film-forming foams (AFFF) use sites, and sites where PFOS precursors were used in textile treatment. Non-point source related occurrences are highest near urbanized areas with high population densities. Current data illustrates the occurrence of PFOS in surface waters across multiple U.S. states. Additional data are needed to better understand PFOS occurrence in U.S. aquatic ecosystems, particularly in estuarine and marine systems and where monitoring data are not available (e.g., southwestern, central, and western U.S.). Additional PFOS occurrence data would provide valuable information on potential spatial and temporal variability in surface waters, and possible risks posed to aquatic ecosystems.

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Disclaimer—The views expressed in this manuscript are those of the authors and do not necessarily represent the views or policies of the U.S. Environmental Protection Agency.

Supplemental Data—The Supplemental Data are available on the Wiley Online Library at DOI: 10.1002/etc.xxxx.

Keywords

Perfluoroalkyl Substance (PFAS); Perfluorooctane Sulfonate (PFOS); Fate and Transport; Occurrence; Surface Waters

BACKGROUND

Perfluorooctane sulfonate (PFOS), and its salts, belong to the Per- and Polyfluoroalkyl Substances (PFAS) group of chemicals. PFAS are synthetic, organic compounds that consist of a carbon backbone and a unique functional group, such as sulfonate or carboxylic acid (C_nF_{2n+1}-R; Ahrens 2011; Buck et al. 2011; Lindstrom et al. 2011; Wang et al. 2017). Specifically, PFOS consists of an eight-carbon backbone and a sulfonate functional group (formula is C_8F_{17} SO_3^-; CAS No. 45298–90-6 for anionic form). PFOS, and its salts, have been incorporated into a wide range of consumer and industrial products for decades (since the 1950s), including surface treatments for soil and stain resistance of textiles, paper, metals, pesticides, and are used in applications such as in aqueous film-forming foam (AFFF; Ahrens 2011; Buck et al. 2011; Lindstrom et al. 2011; Ahrens and Bundschuh 2014) and by 2002 the use of PFOS was phased with the exception of a few small applications (i.e., AFFF and hard chrome plating mist suppression) ((Lindstrom et al. 2011; U.S. EPA 2000).

The manufacture of PFOS started in 1949 with the 3M Company (Paul et al 2009). Prior to 2000, the 3M Company was the major producer of perfluorooctanesulfonyl fluoride (PFOSF also known as POSF), the raw material that undergoes base-catalyzed hydrolysis in the Electrochemical Fluorination (ECF) process to make PFOS, with smaller producers in Europe and Asia (Paul et al. 2009; Lehmler 2005; Lindstrom et al. 2011). In 2000, the 3M Company manufactured approximately 78% of the estimated global PFOSF production (approximately 3,665 tons of 4,650 tons; OECD 2002). The estimated total cumulative production of PFOSF from the 3M Company and other western companies through 2002 is between 44,000 and 96,000 tons. Information on previous and current production of PFOSF from Asia and other production sources is limited (Prevedourous et al. 2006; Smithwick et al. 2006; Paul et al. 2009).

In May 2000, following negotiations between EPA and 3M, the 3M Company agreed to a voluntary phase out and to find substitutes for PFOS chemistry used to produce all but a few small applications (i.e., AFFF and hard chrome plating mist suppression) across their range of products by 2002 (Lindstrom et al. 2011; U.S. EPA 2000). Starting around the same time, a series of Significant New Use Rules (SNUR) were put into place by the EPA to restrict the production and use of chemicals that contain PFOS and its precursors in the U.S. (Lindstrom et al. 2011). Additionally, Canada phased out PFOS, its salts, and precursors (ECCC 2018). In 2009, PFOS and related compounds were listed under Annex B of the Stockholm Convention on Persistent Organic Pollutants; restricting global manufacturing and use of PFOS (OECD 2002; Ahrens 2011). Homologues, neutral precursor compounds, and new classes of PFAS continue to be produced; and therefore, are potential sources of PFOS (Ahrens 2011). The production of PFOS was estimated to be approximately 1,000

tons from 2002 and onward (Paul et al. 2009). However, while industrialized countries, like the U.S., phased-out the use of PFOS and its precursors, producers in other countries, such as China and Brazil, have scaled up their production to fill remaining demand (Wang et al. 2013). Despite the wide use in an array of industrial and consumer products globally, information on the sources, volumes, and emission of PFOS and its precursors are and have been limited (Paul et al. 2009; Zhang et al. 2016; Ankley et al. 2020).

PFOS is resistant to hydrolysis, photolysis, microbial degradation, and metabolism, making it persistent in aquatic environments (OECD 2002; Ahrens et al. 2011; Buck et al. 2011; Lindstrom et al. 2011). PFOS has been detected in tissues of aquatic organisms and both field and laboratory data show a propensity of PFOS to bioaccumulate and move through aquatic food webs (Houde et al. 2006 and 2011; Giesy et al. 2010). Current toxicity literature reports that PFOS exposures can have adverse effects on a diversity of aquatic organisms (Beach et al. 2006; Giesy et al. 2010). Despite the persistence, toxicity, and potential bioaccumulation of PFOS in the aquatic environment, current information on the environmental distribution of PFOS in surface waters of the U.S. is relatively limited (Ankley et al. 2020).

SOURCES OF PFOS TO AQUATIC ENVIRONMENTS

Aquatic environments and soil are thought to serve as a reservoir of PFOS, with 42,000 tons emitted to aquatic environments compared to 235 tons released globally into air between 1980 and 2002 (Paul et al. 2009; Rankin et al. 2016). Unlike other contaminants commonly found in aquatic ecosystems, such as metals, PFAS are synthetic compounds with no natural source. Thus, the occurrence of any PFAS compound in the environment is an indication of anthropogenic sources (Ahrens 2011). The occurrence of PFOS in aquatic environments can be attributed to both point and non-point sources, entering aquatic environments from industrial and consumer products during manufacturing, along supply chains, and during product use and/or disposal (Paul et al. 2009; Ahrens 2011; Kannan 2011; Ahrens and Bundschuh 2014). However, quantitative assessments of PFOS production, point and non-point source discharges, and environmental measurements are limited compared to other persistent bioaccumulative pollutants (Ahrens and Bundschuh 2014; Zhang et al. 2016)

Potential point sources of PFOS to the aquatic environment include both industrial facilities and municipal wastewater treatment plants (WWTPs). Additional point sources may include surface water runoff from industrial use sites such has metal plating facilities, areas that have received AFFF applications, landfills, and contaminated soils. Of these, industrial facilities, specifically those for fluorochemical manufacturing and other use facilities, are a primary source of PFOS to aquatic systems (Ahrens et al. 2011; Houtz et al. 2016; Sedlak et al. 2017). Estimated total global releases to water arising from discharge of PFOS during manufacturing from 1970 to 2002 ranged between 230 and 1,450 tons (Paul et al. 2009). Several studies have found increased concentrations of PFOS in municipal WWTP effluent compared to influent (Schultz et al. 2006; Sinclair and Kannan 2006). Schultz et al. (2006) observed an increase of PFOS concentrations following treatment of wastewater using standard technologies in a municipal WWTP. Similarly, Sinclair and Kannan (2006) observed a statistically significant increase (by 227 times ± 119%) of PFOS measured in

effluent compared to influent from a municipal WWTP that received industrial inputs in New York. These studies indicate conventional municipal WWTP processes (i.e., primary and secondary treatment) may not be effective at removing PFOS and that the degradation of other PFAS may be contributing to the increased concentrations of PFOS in effluent (Schultz et al. 2006; Sinclair and Kannan 2006; Ahrens 2011).

Potential non-point PFOS sources to aquatic environments include: dry and wet atmospheric deposition, discharge of contaminated groundwater from manufacturing sites, runoff from impervious surfaces in urban environments, discharge of contaminated groundwater from use of AFFF, and land application of contaminated biosolids (OECD 2002; Paul et al. 2009; Ahrens et al. 2011; Kannan 2011). Identification of non-point PFOS sources and understanding their relative contribution to aquatic ecosystems can be difficult (Paul et al. 2009; Ahrens 2011). Overall, the presence of non-point PFOS sources and their relative contributions are dependent on the aquatic system, air, groundwater, and soil levels, and nearby land uses. For example, concentrations of PFAS, including PFOS, have been influenced by urban land use (Ahrens 2011; Zhang et al. 2016) and overall PFAS concentrations in the environment have been positively correlated with human population density. PFOS was detected in aquatic systems at elevated concentrations (ranging between 97 and 1,371 ng/L) in densely populated areas of the U.S. and Europe (Zhang et al. 2016 and Loos et al. 2009, respectively). Paul et al. (2009) estimated the total global PFOS emissions to air and water from 1970 to 2009 resulting from consumer use and disposal to be between 420 and 2,100 tons.

PFAS are still produced that can transform or degrade into compounds belonging to the perfluoroalkane sulfonic acids (PFSA) family of PFAS, including PFOS (Ahrens 2011). The metabolic transformation of PFAS precursors such as fluorotelomer sulfonates (FTSAs) and perfluoroalkyl sulfonamidoacetic acids (FASAAs), and the degradation of volatile PFAS such as perfluoroalkyl sulfonamidoethanols (FASEs), are known to degrade to PFOS. (Ahrens and Bundschuh 2014; Benskin et al. 2009; Boulanger et al. 2005; Buck et al. 2011; Lange 2000; Lui and Mejia Avendaño 2013; Plumlee et al. 2009; Rhoads et al. 2008; Wang et al. 2017). However, understanding of these transformation processes is limited, and additional work is needed to fully understand these processes and their role as a source of PFOS to aquatic environments (Lau et al. 2007; Buck et al. 2011; Lui and Mejia Avendaño 2013; Wang et al. 2017). The contribution of precursors to the presence of PFOS in the environment is unknown and difficult to quantify. However, these precursors can be a continuous source of PFOS. Particularly, the degradation of precursors may represent a potentially significant known source of PFOS to the aquatic environment, especially since PFOS production within the U.S. has not occurred since 2002 (Buck et al. 2011; Lui and Mejia Avendaño 2013). Nevertheless, PFOS-treated articles, such as fabrics, paper, and other treated materials, are still being imported in the U.S. and end up being released into the environment (Allred et al. 2015; Lang et al. 2016; Lui et al. 2014). The importation of PFOS treated articles is considered as production under the Toxic Substances Control Act (TSCA; U.S. EPA 2020).

PFOS can also be re-emitted to aquatic environments from PFOS sinks such as soil, groundwater, ice, and sediment. Sediment is an important sink of PFOS in the aquatic

environment (Ahrens et al. 2011). Like other persistent organic pollutants, the movement of PFOS between groundwater, surface water, and sediment is complex and depends on the chemical properties of PFOS and site-specific physiochemical characteristics (including pH, temperature, organic carbon content, and salinity) of the aquatic environment. In general, PFOS may adsorb to sediments (with a K_d greater than 1 mL/g; Giesy et al. 2010). However, this sorption to sediments is limited and PFOS has a K_{OC} of 2.57 indicating that PFOS is relatively mobile in water and the physicochemical characteristics of the sediment ultimately influence the sorption of PFOS (Ahrens et al. 2011; Higgins and Luthy 2006). While the release of PFOS from the transformation of other PFAS and the historical products still in use (e.g., consumer goods manufactured, imported and/or obtained before the PFOS discontinuation and regulations) will continue into the future, the re-emissions of PFOS from existing sinks are assumed to be slowly decreasing since the restrictions and regulations of PFOS have gone into place (Paul et al. 2009; Ahrens 2011; Ahrens and Bundschuh 2014, Washington and Jenkins 2005; Washington et al. 2015).

METHODS

An understanding of the occurrence of PFOS in ambient surface waters across the U.S. is needed to better identify the environmental relevance of concentrations reported in PFOS toxicity literature. In the present paper, PFOS occurrence data in U.S. ambient surface waters were obtained from publicly available literature, including peer-review journal articles, theses, and government and industry reports. Searches for such literature were conducted by developing a series of search terms related to the chemicals analyzed (e.g., perfluorooctane sulfonate or PFOS, and its salts), waterbody type sampled (e.g., ambient waterbodies such as rivers, streams, wetlands, or lakes), and location of sampled waterbody (which was specific to the U.S.). Databases searched were Science Direct, Google Scholar, and EPA's ECOTOX Database (which includes ambient water concentrations for calculation of bioconcentration and bioaccumulation factors such as those in Burkard (2021); https://cfpub.epa.gov/ecotox/). Additionally, it should be noted that many states in the U.S. have conducted monitoring of PFOS. However, most of the state monitoring data for PFOS are currently not publicly available. Thus, after extensive literature searches (including searches by individual states and on state monitoring databases/websites) and reaching out to individual states, the data presented in this manuscript appear to be inclusive of all publicly available relevant PFOS data.

The identified citations were reviewed for reported ambient surface water concentrations of PFOS, including relevant summary statistics (e.g., minimum, maximum, mean, and median concentrations) reported by the individual study authors. Citations without PFOS data and/or with only drinking or ground water data were not included. Measured PFOS concentrations in ambient surface waters from the identified citations with appropriate information were extracted into a database (Supplemental Data Table S2). For the purposes of this overview and comparison, all concentrations reported here are in nanograms per liter (ng/L). Additional data extracted from the identified citations included: the location of the waterbody sampled (as both the location by state, waterbody name, and GPS coordinates), specific site name or description (if provided in the paper), identification of possible previous exposure to PFOS (as stated by the study authors of the individual papers), the date

the sample was collected, the number of samples collected and/or analyzed, and analytical methods used to measure PFOS (including reported limits of quantification and limits of detection). Beyond the extraction of the aforementioned data, the identified citations were not further evaluated for data quality. Instead, all relevant PFOS occurrence data for ambient surface waters in the U.S. were captured in this present paper to provide an overview of PFOS occurrence in ambient surface waters across the U.S.

The extracted PFOS occurrence data were included in understanding the current distribution, frequency of detection, and summary statistics (specifically both arithmetic and geometric means, median, and overall range) of reported PFOS concentrations in ambient surface waters across the U.S. The PFOS occurrence data were also evaluated for potential spatial and/or temporal variability of PFOS in ambient surface waters. And lastly, the occurrence and concentrations of PFOS in ambient surface waters across the U.S. were generally compared to those reported globally.

RESULTS OF PFOS OCCURRENCE IN U.S. SURFACE WATERS

PFOS is one of the most commonly detected PFAS in aquatic ecosystems, along with PFOA (Ahrens 2011; Benskin et al. 2012; Zareitalabad et al. 2013; Dinglasan-Panlilio et al. 2014; Nakayama et al. 2017; Remucal 2019). Despite its wide use, and persistence in the aquatic environment, current information on the distribution of PFOS in ambient surface waters of the U.S. is relatively limited. Available data are largely collected from freshwater systems in eastern states, with most of the current, published PFOS occurrence data focused on a handful of study areas with known manufacturing or industrial uses of PFAS, such as the Mississippi River near a 3M facility, the Great Lakes, the Cape Fear Drainage Basin, and waterbodies near Decatur, Alabama and northern Georgia, along with areas of known AFFF use, such as fire-training areas on military bases (Anderson et al. 2016; Figure 1 and Table 1; Supplemental Data, Figure S1 and Table S1).

Concentrations of PFOS in surface waters vary widely, with observed concentrations ranging over eight orders of magnitude and detected generally between picogram and nanogram per liter. Some sites reported concentrations in the microgram and milligram per liter ranges. (Ahrens 2011; Zareitalabad et al. 2013). Measured surface water concentrations of PFOS in peer-reviewed journal articles and publicly available industry and government reports, range between 0.074 and 8,970,000 ng/L with an arithmetic mean concentration of 786.77 ng/L, a geometric mean concentration of 5.468 ng/L, and a median concentration of 3.6 ng/L. However, it should be noted that the mean and median concentrations reported here were calculated from the reported concentrations for individual samples. And therefore, these mean and median concentrations are not fully representative of all the measured PFOS concentrations in U.S. surface waters. In particular, should be noted that some of the papers (6 papers total without individual concentration data; see Supplemental Data Table S2) only reported summary information such as minimum and maximum concentrations and did not provide more detailed data (e.g., individual sample concentrations or sample site means) in the paper (Figure 2 and Table 1; Supplemental Data, Table S2).

Consistent with the calculated median of 3.6 ng/L, a majority (90.99%) of measured PFOS concentrations in the current literature fall below 300 ng/L with fewer (9.01%) observed concentrations greater than 300 ng/L (Figure 3). As mentioned in the sources of PFOS section above, in contrast with other contaminants commonly found in aquatic ecosystems PFOS is a synthetic compound with no natural source. Thus, the occurrence of PFOS in surface water is an indication of anthropogenic sources, including consumer and industrial use, long-range transport, atmospheric deposition, surface water runoff, and general persistence in the environment (Ahrens 2011).The higher frequency of PFOS concentrations below 300 ng/L can likely be attributed to: (1) the increased tendency of study designs found in the current literature to include sites with no known previous exposures to PFAS in order to compare results to sites with known previous exposure to PFAS, which is depicted in Figure 4 and (2) it is likely that sites in which the study authors did not specify potential exposure to PFAS could be classified as relatively pristine sites with no known PFOS inputs since the reported measured concentrations indicate that the PFOS concentrations are generally similar to those observed in sites noted to have no known PFOS inputs (Table 2).

Numerous available studies report measured PFOS concentrations in surface waters across the U.S. (Figure 2 and Table 1), some of which are summarized below; however, more detailed information on PFOS occurrence in areas not previously sampled and spatial and temporal variability of PFOS remain limited. Prior to this review, there were few current analyses of spatial variability of PFOS concentrations in surface water across the U.S. (Remucal 2019). This review indicates that PFOS occurrence is widely reported in areas where sampling has been conducted. And that the presence and measured concentrations of PFOS in surface waters are similar between both lotic and lentic and freshwater and estuarine/marine systems, based on the limited data available (Supplemental Data Table S2). Higher PFOS concentrations in surface water tend to be dependent on the presence of a nearby source and generally increase with levels of urbanization. Across the Great Lakes region, PFOS concentrations were higher in the more southern lakes of Erie and Ontario compared to the upstream lakes of Superior, Michigan, and Huron (Table 1; Remucal 2019). Similarly, Zhang et al. (2016) observed that measured PFOS surface water concentrations in urban areas (with mean PFOS concentrations of 4.21 ng/L in urban sites within Rhode Island, New York, and New Jersey) were an order of magnitude higher than those at rural sites (with an mean PFOS concentration of 0.42 ng/L).

Currently, there are insufficient data to quantitatively evaluate temporal trends of PFOS in surface waters across the U.S. (Remucal 2019). However, recent studies have suggested that PFOS concentrations in surface waters with limited sampling sites in northeastern states appear to have generally decreased since the voluntary phase out of PFOS in 2002 (Zhang et al. 2016; Pan et al. 2018). While these more recent studies observed lower measured PFOS concentrations in surface waters compared to those reported in earlier reports (Hansen et al. 2002; Nakayama et al. 2007), few studies have measured PFOS concentrations from the same sampling locations over time. However, it appears that eight studies (six focused on the Great Lakes and two in New York on the Hudson River; Table 1) measured PFOS in the same waterbody over time (Supplemental Data, Table S2 and Figure S1). Thus, the observed lower concentrations reported in recent literature could be due to trends of

PFOS concentrations decreasing since the 2002 PFOS phaseout, differences in sampling site locations, and/or advances in analytical methods for detecting PFOS that reduced detection limits. If the dataset is limited to Lake Ontario, which is one of the most well-studied waterbodies for PFOS occurrence in the U.S., data from 2002 to 2010 indicates an apparent decrease in PFOS concentrations over time. This and any future decreases would be likely due to the reduction in PFOS use in manufacturing. However, this downward trend of PFOS concentrations in Lake Ontario surface waters was not statistically significant ($p > 0.05$; Remucal 2019).

PFOS occurrence and concentrations in the Great Lakes region

The Great Lakes are among the most widely studied waterbodies in the U.S. for PFOS occurrence. However, occurrence data are still relatively limited for this system and were largely collected between 2003 and 2010. Comparisons across the Great Lake system indicate PFOS concentrations are higher in Lakes Erie and Ontario, ranging between 2.8 and 38.5 ng/L and 2.9 and 85.5 ng/L, respectively (Figure 5; Sinclair et al. 2006; Boulanger et al. 2004; De Silva et al. 2011; and Furdui et al. 2009), compared to the more northern Great Lakes. These northern Great Lakes have a maximum reported concentration of 5.46 ng/L in Lake Huron. However, current measured PFOS concentrations in Lakes Huron, Michigan, and Superior were not from sampling sites around urbanized areas (such as Chicago and Detroit) and may not be representative of the potential sources of PFOS related to these areas. The measured concentrations of PFOS in the surface waters of Lakes Huron and Michigan, range between 0.24 and 5.46 ng/L (Table 1; Remucal 2019; Furdui et al. 2008; De Silva et al. 2011) and 0.93 and 3.13 ng/L (De Silva et al. 2011; Simcik and Dorweiler 2013), respectively. In contrast measured PFOS concentrations observed in Lake Superior were considerably lower and range between 0.074 and 0.996 ng/L (Scott et al. 2010; De Silva et al. 2011; Furdui et al. 2011). The higher PFOS concentrations in Lakes Erie and Ontario are likely due to higher levels of industrial activities and urbanization around these lakes (Boulanger et al. 2004; Remucal 2019), and could also be associated with the sampling locations. A mass balance constructed for Lake Ontario by Boulanger et al. (2004) indicated wastewater effluent was the major source of PFOS to the lake. In contrast, inputs from Canadian tributaries and atmospheric deposition of PFOS, and other PFAS that may be transformed into PFOS, were the major contributing sources of PFOS to Lake Superior. Inputs from Canadian tributaries and atmospheric deposition were estimated to contribute 57 and 32% of PFOS inputs into Lake Superior respectively (Scott et al. 2010).

PFOS occurrence and concentrations in the southeastern U.S.

Measured PFOS concentrations in southeastern U.S. surface waters were similar to those measured in Lakes Erie and Ontario, with the exception of some of the highest concentrations detected in waterbodies near areas with PFOS manufacturing (Figure 6 and Table 1). In 1999, the 3M Company conducted a multi-city study measuring PFOS concentrations across waterbodies with known manufacturing and/or industrial uses of PFOS (3M Company 2001). In the 3M Company's 2001 report, PFOS concentrations from sites with known PFOS discharges were compared to PFOS concentrations measured in waterbodies with no known sources of any PFAS chemical (3M Company 2001). In this comparison study, cities with known PFOS exposure were Mobile and Decatur, Alabama;

Columbus, Georgia; and Pensacola, Florida. Measured PFOS concentrations ranged from not detected (reported detection limit of 2.5 ng/L; 3M Company 2001) to 41.5 ng/L in the cities with known PFOS discharges (Table 1). These PFOS concentrations were compared to those measured in control cities. These study control cities were Cleveland, Tennessee and Port St. Lucie, Florida and PFOS concentrations ranged from not detected to 137.5 ng/L (3M Company 2001). The PFOS concentrations measured in Cleveland, Tennessee were below the limit of detection (2.5 ng/L[1]) and were lower than the PFOS concentrations observed in the cities with known PFOS exposure, as was expected in the report for the control cities. However, PFOS concentrations around Port St. Lucie, Florida, the other control city, were unexpectedly similar to and at times higher than the waterbodies with known PFOS discharges. The sources of PFOS near Port St. Lucie, Florida remain unknown; however, observed PFOS concentrations suggest the presence of a potential manufacturing/industrial source or the use of AFFF in this area (3M Company 2001).

Water samples were collected from ponds near all of the sampling sites except those in Cleveland, Tennesse. As reported in Table 1, PFOS concentrations in these additional pond sites were similar to those measured in Mobile, Alabama (ranging between 32 and 33 ng/L), lower than those observed in Columbus, Georgia (as PFOS was not detected with a detection limit of 2.5 ng/L), and higher than those measured in Decatur, Alabama (ranging between 108 and 111 ng/L) and in Port St. Lucie, Florida (ranging between 1,830 and 48,200 ng/L). Samples collected from the pond site near Port St. Lucie, Florida had some of the highest measured PFOS concentrations in publicly available literature with the maximum concentration of 48,200 ng/L. In the report, the 3M Company conducted additional sampling at the pond site in Port St. Lucie, Florida and determined that the measured PFOS concentrations at this site were more variable than the initial measurements alone indicated and were lower than the previous measurements, ranging between below detection (i.e., < 2.5 ng/L) and 2,340 ng/L. Aside from the samples collected in Port St. Lucie, Florida, this report demonstrated that measured PFOS concentrations in surface waters tend to be higher in areas with PFOS manufacturing and/or industrial use (3M Company 2001).

In separate studies, PFOS and PFOA concentrations were measured in surface waters by Hansen et al. (2002) near Decatur, Alabama and Konwick et al. (2008) in Georgia. Hansen et al. (2002) studied a stretch of the Tennessee River near Decatur, Alabama and Konwick et al. (2008) focused on the Conasauga River in Georgia, both areas with known PFOS discharge and use. In Hansen et al. (2002), discharge from a fluorochemical manufacturing facility entered the Tennessee River towards the middle of the study area. In contrast, Konwick et al. (2008) compared the PFOS concentrations measured in the Conasauga River with those from sites with no known exposure along the Altamaha River. In both studies, mean PFOS concentrations were higher in the study areas with PFOS sources. Specifically, Hansen et al. (2002) observed mean PFOS concentrations upstream of the fluorochemical manufacturing facility were 30.85 ng/L (ranging between 16.0 and 52.6 ng/L) and were 103.9 ng/L

[1]Limits of detection and quantification differ across the PFOS occurrence literature and is dependent on the specific lab and the analytical methods used in a particular study. Specific limits of detection and quantification provided in the subsequent text for each individual study.

(ranging between 30.3 and 144 ng/L) downstream of the fluorochemical manufacturing facility. Similarly, Konwick et al. (2008) observed higher measured PFOS concentrations in the Conasauga River, which ranged from below limit of detection (with a limit of detection of 1.5 ng/L) to 321 ng/L, compared to those in the Altamaha River, ranging between 2.6 and 2.7 ng/L. Consistent with the report from the 3M Company summarized above, effluents from manufacturing facilities, WWTP, and carpet mill effluents were determined to be the source of increased PFOS concentrations in both the Tennessee and Conasauga Rivers (Hansen et al. 2002 and Konwick et al. 2008, respectively). These PFOS concentrations are relatively consistent with those measured in Alabama and Georgia as reported by the 3M Company (3M Company 2001).

Nakayama et al. (2007) and Cochran (2015) measured PFAS, including PFOS, in the Cape Fear Drainage Basin in North Carolina and waterbodies on Barksdale Air Force Base in Bossier City, Louisiana, respectively. PFOA and PFOS were found to be the dominant PFAS detected in both studies. Nakayama et al. (2007) detected PFOS in 97.5% of all samples above the limit of quantification of 1 ng/L. PFOS concentrations in the Cape Fear Drainage Basin ranged between < 1 (the lower limit of quantification) and 132 ng/L with a mean concentration of 31.2 ng/L. As in other studies summarized above, lower PFAS concentrations, including PFOS, were found in the upland tributaries and concentrations were highest in the middle reaches of the Cape Fear Drainage Basin, near expected sources. Municipal wastewater treatment plant effluents were identified as a source of PFAS to the study area. AFFF usage at the Department of Defense base in Fayetteville, North Carolina and the land application of biosolids likely contributed as well (Nakayama et al. 2007). Cochran (2015) detected PFOS in 79% of all water samples collected and concentrations ranged between below the limit of quantification (i.e., 10 ng/L) and 7,070 ng/L, with an average concentration of 776.7 ng/L. PFOS concentrations varied in samples collected in Barksdale Air Force Base based on proximity to fire training areas. Cochran (2015) attributed the evaluated PFOS concentrations to run off and ground infiltration of AFFF formally used on the base during firefighting and/or training.

PFOS occurrence and concentrations in the midwestern U.S.

Similar PFOS concentrations were reported in the publicly available literature for waterbodies in urban areas across the midwestern U.S. Lower PFOS concentrations were reported in areas with no previous PFAS exposure (identified as remote areas by the individual study authors) in the same states (Simcik and Dorweiler 2005; Newsted et al. 2017). In Minnesota, Simcik and Dorweiler (2005) observed PFOS concentrations ranging between 2.4 and 50.4 ng/L in urban areas near Minneapolis and concentrations ranging between less than the limit of quantification of 0.29 ng/L and 1.2 ng/L were observed in remote areas in northern Minnesota (Table 1). Additionally, Newsted et al. (2017) reported an average PFOS concentration of 528.9 ng/L (ranging between below limit of quantification and 18,200 ng/L; limit of quantification not provided) in surface waters collected from the Upper Mississippi River near the Minneapolis/St. Paul, Minnesota metropolitan area with a maximum concentration of 18,200 ng/L. The occurrence of PFOS at these urban sites was attributed to the presence of manufacturing source, runoff, and wastewater discharge (Simcik and Dorweiler 2005 and Newsted et al. 2017).

PFOS occurrence and concentrations in the northeastern U.S.

Several studies measured PFOS concentrations in surface waters in the northeastern U.S that are comparable to those reported in Minnesota (Sinclair et al. 2006; NJ DEP 2019). Sinclair et al. (2006) measured PFOS in various waterbodies across New York state and observed a median concentration of 756 ng/L in surface waters collected from a Superfund site at Lake Onondaga (ranging between 198 and 1,090 ng/L; Table 1) and attributed these elevated concentrations to several industries located along Lake Onondaga. All other observed concentrations of PFOS in New York, including sites along the Niagara River, the Finger Lakes, Lakes Oneida and Champlain, the Erie Canal, and Hudson River, had lower median PFOS concentrations ranging between 0.8 and 13 ng/L (Table 1; Sinclair et al. 2006).

New Jersey Department of Environmental Protection (NJ DEP) measured PFOS in surface water samples collected from 14 different sites across New Jersey. PFOS concentrations ranged from below the detection limit of 1.0 ng/L to 102 ng/L (NJ DEP 2019). Individual samples collected along Pine, Little Pine, and Mirror Lakes had measured PFOS concentrations of 102, 100, and 72.9 ng/L, respectively. All other observed concentrations of PFOS in New Jersey freshwaters were below 15 ng/L (Table 1). NJ DEP attributed the elevated concentrations of PFOS observed at Pine, Little Pine, and Mirror Lakes to the use of AFFF in training and/or firefighting on the Department of Defense (DoD) Joint Base McGuire-Dix-Lakehurst (NJ DEP 2019).

PFOS occurrence and concentrations in the western U.S.

PFOS concentrations in surface waters of western U.S. states are generally consistent with the lower-end concentrations (less than 100 ng/L) measured in eastern states; however, the monitoring data for PFOS was limited in the western U.S. Plumlee et al. (2008) measured PFOS and PFOA in Coyote Creek and a tributary of Upper Silver Creek in San Jose, California and determined PFOS concentrations in both Coyote and Upper Silver Creeks to be similar to those measured in eastern states, which are summarized above (Figure 6). Concentrations of PFOS in Coyote Creek ranged from 4.8 to 25 ng/L and concentrations in Upper Silver Creek ranged from 27 to 56 ng/L. The source of PFOS to these aquatic systems was unknown, however, Plumlee et al. (2008) stated that a combination of atmospheric deposition of volatile precursors and surface runoff were likely sources of PFOS to both Coyote and Upper Silver Creeks.

State level data are currently available for Colorado and New Mexico. In particular Colorado Department of Public Health and the Environment (2020) measured PFOS in surface water samples collected from 71 different sampling locations across Colorado. PFOS concentrations ranged from below the detection limit (which varied between 0.42 and 2.50 ng/L across sites) to 54 ng/L. New Mexico Environment Department (2021) also measured PFOS concentrations collected from 67 surface water sampling sites across the state. These PFOS concentrations ranged from below detection limit (which varied between 0.86 and 1.9 ng/L across sites with some limits not reported) to 5,900 ng/L. The elevated concentrations of PFOS were observed in sampling locations near Holloman Air Force Base (New Mexico Environment Department 2021).

Lastly, Dinglasan-Panlilio et al. (2014) measured PFOS concentrations in surface waters along the Puget Sound in Washington, as well as Clayoquot and Barkley Sounds in British Columbia, Canada. PFOS concentrations measured by Dinglasan-Panlilio et al. (2014) were lower than those observed from sites in eastern states (such as those summarized above for Alabama, Florida, and North Carolina with known manufacturing and/or industrial use of PFOS (Table 1) Concentrations ranging from 0.2 to 5.9 ng/L in Puget Sound and 0.25 to 0.7 ng/L in Clayoquot and Barkley Sound, British Columbia. These concentrations are consistent with those reported in the publicly available literature for areas identified as remote by individual study authors, such as in Minnesota (Simcik and Dorweiler 2005) and in New York (Sinclair et al. 2006), as summarized above. The study authors indicated specific regional sources and atmospheric deposition were likely PFOS sources to these areas with no previous PFAS exposure (Dinglasan-Panlilio et al. 2014).

Summary of PFOS occurrence and concentrations across the U.S.

Despite the wide use and persistence of PFOS in aquatic ecosystems and unlike the sampling of PFOS in drinking water sources[2], groundwater, and fish tissue monitoring[3], current information on the environmental distribution of PFOS in ambient surface waters across the U.S. remains very limited. Additionally, sampling efforts in other media corroborate that PFOS occurrence in aquatic ecosystems is relatively widespread, particularly in urban areas with detection frequencies of 73% in fish tissue samples from urban rivers and 100% in fish tissue samples from the Great Lakes (Stahl et al. 2014). Similar conclusions were reached in a sampling effort across Canada by Gewurtz et al. (2013), which demonstrated that distribution of PFOS detected in multiple media types (i.e., air, water, sediment, and fish and bird tissue) generally related to urbanization with PFOS concentrations reported in surface water for 23 out of 31 sampling locations.

Present surface water occurrence data are largely collected from freshwater systems in eastern states and in the upper midwest and focused on a handful of study areas with known manufacturing or industrial uses of PFAS or use of AFFF. Current data indicate that PFOS concentrations measured in U.S. surface waters vary widely, across eight orders of magnitude (Table 1). PFOS concentrations in areas with little to no PFAS manufacturing and/or industrial use) range between 0.074 to 23.23 ng/L (Figure 4 and Table 1). This contrasts with PFOS concentrations measured in areas with known PFAS manufacturing, industrial use, and/or application of AFFF, which vary widely and reach up the maximum observed concentration of 8,970,000 ng/L at a site impacted by AFFF (Figure 6 and Table 1). While current PFAS occurrence data illustrate the prevalence and quantify concentrations of PFOS in ambient surface waters across the U.S., additional data, particularly in central, southwestern, and western freshwaters as well as saltwater systems, is needed to better understand PFOS occurrence in aquatic ecosystems across the U.S.

[2]EPA's database for the Unregulated Contaminant Monitoring Rule (UCMR) that includes data for treated surface waters, (https://www.epa.gov/dwucmr)

[3]EPA's National Rivers and Streams Assessment (NRSA; https://www.epa.gov/national-aquatic-resourcesurveys/ncca) and the Great Lakes Human Health Fish Tissue Study component of the EPA National Coastal Condition Assessment (NCCA/GL)

COMPARISON OF PFOS OCCURRENCE IN THE U.S. TO GLOBAL AMBIENT SURFACE WATERS

Similar surface waters in the U.S., generally PFOS and PFOA were the most commonly detected PFAS in surface waters around the world (Ahrens 2011). However, it should be noted that the frequency of PFOS and PFOA detection may be an artifact resulting from regulations of these compounds and the lack of available standardized analytical methods for other PFAS. Generally, on a global scale PFOS concentrations in surface waters generally range between picogram/liter and nanogram/liter with some concentrations in the milligram/liter range. PFOS concentrations in the U.S. were comparable to those reported in studies with sampling sites in other countries. Global surface water PFOS concentrations reported in the public literature ranged between not detected and 2,100,000 ng/L. These global surface water concentrations are summarized below to provide a comparison with those observed in the U.S.

In Canada elevated PFOS concentrations in surface waters generally occurred in urbanized areas, suggesting urban areas with high population densities contributed to the elevated PFOS concentrations, similar to indications from U.S. data (Gewurtz et al. 2013; Scott et al. 2009). And PFOS was monitored and assessed for locations across Canada from 2006 through 2011, and it was concluded that PFOS posed a risk to the aquatic environment (ECCC 2018). PFOS concentrations measured by Gewurtz et al. (2013) ranged between not detected (with a detection limit of 2 ng/L) and 10 ng/L in surface waters across Canada. PFOS was rarely detected in surface water samples collected from non-urban areas (Gewurtz et al. 2013). In a systematic, cross-Canada study of PFAS in surface waters, Scott et al. (2009) observed PFOS and PFOA as the dominant PFAS detected and that generally PFOS concentrations were higher, overall ranging between < 0.02 and 34.6 ng/L, than PFOA concentrations, which ranged between 0.044 and 9.9 ng/L. These studies indicated PFOS concentrations in Canadian surface waters were lower than those in the U.S., Europe and Asia (Scott et al. 2009). However, PFOS ranged from not detected (with a detection limit of 4 ng/L) to 2,100,000 ng/L in Etobicoke Creek, a tributary to Lake Ontario, after an accidental spill of a fire-retardant foam containing perfluorinated surfactants was released at L.B. Pearson International Airport in Toronto, Ontario in June 2000 (Moody et al. 2002). The elevated concentrations of PFOS measured by Moody et al. (2002) were higher than those measured in U.S. surface waters and are consistent with the presence of elevated concentrations in surface waters near a source of PFOS (Ahrens 2011).

PFOS concentrations measured in surface waters across Europe were similar to those observed in the U.S. Specifically, in a European Union (EU)-wide study of polar organic persistent pollutants, Loos et al. (2009) observed a median PFOS concentration of 6 ng/L in surface waters sampled across a wide range of sampling sites (including contaminated and pristine rivers and streams of various sizes). However, relatively high median PFOS concentrations between 32 (from the Rhine River in Germany) and 1,371 ng/L (from Krka River in Slovenia) were also observed. Mean PFOS concentrations observed by Pan et al. (2018) were similar to those reported in Loos et al. (2009) and across the U.S., with mean surface water concentrations from waterbodies across western Europe, specifically the

Thames River, Mälaren Lake, and Rhine River, ranging between 3.15 and 13.8 ng/L and a maximum concentration of 18.8 ng/L measured in the Thames River. Kwadijk et al. (2010) reported PFOS concentrations between 4.7 and 32 ng/L in surface water samples collected from 20 sampling locations across the Netherlands. Lastly, similar and some slightly higher PFOS concentrations, with averages ranging between 16 and 449 ng/L, were observed by Huset et al. (2008) in the Glatt Valley Watershed in Switzerland. Like in the U.S. and Canada, concentrations of PFOS in surface waters across Europe were higher in urbanized areas and sources have been attributed to municipal waste water treatment plant effluent, AFFF spills, and fluorochemical manufacturing facilities (Loos et al. 2007 and 2009; Huset et al. 2008; Kwadijk et al. 2010; Ahrens 2011; Pan et al. 2018).

Lastly, like PFOS concentrations observed in Canada and Europe, PFOS occurrence in surface waters across Asia were generally similar to those reported in the U.S., with lower reported maximum concentrations being observed in Asia compared to the U.S. (Xu et al. 2013). In Japan, Saito et al. (2003) observed PFOS concentrations ranging between 0.3 and 157 ng/L with a median of 1.68 ng/L in 142 surface water samples collected from various locations. Similarly, Nguyen et al. (2011) reported PFOS concentrations ranging between 1 and 156 ng/L collected from an urbanized section of the Marina catchment in Singapore. However, PFOS concentrations in more recently collected surface water samples reported by Pan et al. (2018) were lower than those previously reported in Asia from publicly available literature. Median surface water PFOS concentrations from samples collected from the Yangtze (sample size (n) = 35), Yellow (n = 15), Pearl (n = 13), Liao (n = 6), Han (n = 6), and Huai (n = 9) Rivers and Chao (n = 13) and Tai (n = 15) Lakes ranged between 1.41 and 8.56 ng/L with the an overall maximum PFOS concentration of 29.7 ng/L in Choa Lake (Pan et al. 2018). Overall, the PFOS concentrations observed in Asia were similar to the lower end of those reported in the U.S.

Overall, these studies show the widespread distribution and variability of PFOS concentrations in surface waters around the world and demonstrate that surrounding land use has a large influence on PFOS concentrations in ambient surface waters. Urbanized areas with high population densities tended to have elevated PFOS concentrations in surface waters (Loos et al. 2007 and 2009; Scott et al. 2009; Ahrens 2011; Gewurts et al. 2013). Like in the U.S., PFOS concentrations in surface waters around the world vary widely and current information on the environmental distribution of PFOS in ambient surface waters around the world is relatively limited.

CONCLUSIONS

Currently PFOS is one of the most commonly observed PFAS detected in surface waters (Ahrens 2011; Benskin et al. 2012; Zareitalabad et al. 2013; Dinglasan-Panlilio et al. 2014; Nakayama et al. 2017; Remucal 2019). As demonstrated in this review, PFOS has been detected in a number of ambient surface waters across the U.S. and concentrations of PFOS vary widely (over eight orders of magnitude). The occurrence of PFOS in surface waters indicates the presence of an anthropogenic source, such as consumer and/or industrial use and/or atmospheric deposition, surface water runoff or groundwater discharge, and results from the general persistence and mobility of these chemicals in the environment.(Ahrens

2011). This review indicates that elevated PFOS concentrations are generally associated with a nearby source or urbanization (Table 2). PFOS concentrations measured in areas with known PFAS sources varied widely with a maximum observed concentration of 8,970,000 ng/L (Table 1 and Figure 6A) in comparison to detected PFOS concentrations measured in areas with little or no PFAS sources, which ranged between 0.074 and 23.23 ng/L (Figure 6B). Additionally, some ambient surface water concentrations in the U.S. are within the range of observed toxicity values reported in current literature (with effect concentrations ranging between 28 and 5000,000,000 ng/L; https://cfpub.epa.gov/ecotox/).

As restrictions of PFOS have gone into place, concentrations in ambient surface waters are expected to decrease. Several studies have suggested that PFOS concentrations in U.S. surface waters have decreased since 2002 (Zhang et al. 2016; Pan et al. 2018). Although these studies observed lower measured PFOS concentrations in surface waters compared to those reported in earlier literature (Hansen et al. 2002; Nakayama et al. 2007), to the authors' knowledge, there has not been a systematic sampling effort to measure PFOS concentrations in sites previously sampled or a comparison between analytical methods to confirm the decrease in PFOS concentrations. Recent studies have reported a shift in the PFAS compounds reported in the aquatic environment. Concentrations of shorter-chained PFAS, particularly PFSAs and perfluoroalkyl carboxylic acids (PFCAs), have increased compared to those of PFOS and PFOA (Möller et al. 2009; Pan et al. 2018) as use has switched to shorter chain PFAS. While the shift in PFAS use and manufacturing may result in a decrease in PFOS concentrations entering aquatic environments it will likely take decades or longer for existing sources of PFOS to be reduced to the point that they do not impact water quality in aquatic systems.

Despite the historical wide use, high persistence, and increased public interest in PFAS generally (including PFOS, which is one of the most commonly detected PFAS in surface waters) current information on the distribution of PFOS in aquatic environments across the U.S. is fairly limited and is largely based on studies that have targeted sites where PFAS were known to have been used. However, it should be noted that many states in the U.S. have conducted monitoring of PFOS and most of the state monitoring data for PFOS are currently not publicly available. Thus, the current dataset contains sampling location and study design bias (Figures 1, 3, and 4). Currently, PFOS occurrence data are limited in western states across the U.S., with the existing dataset only including data for California and Washington (Figure 1). Additionally, PFOS occurrence data are limited in marine and estuarine environments. Therefore, both of these areas continue to be PFOS occurrence data gaps in the U.S. Future PFOS sampling efforts should include consideration of filling these data gaps. Additionally, many of the current PFOS occurrence studies in the public literature include numerous sites with no known PFAS exposure in order to compare PFOS concentrations to exposed sites. This general study design may result in a higher frequency of measured PFOS concentrations below 300 ng/L compared to middle, high, and very high concentrations. This tendency may skew the median and mean concentrations of PFOS in ambient surface waters across the U.S. to the lower end of the concentrations. However, based on the great difference between the arithmetic mean concentration of 786.77 ng/L, a geometric mean concentration of 5.468 ng/L, and the median concentration of 3.6 ng/L for measured PFOS concentrations across the U.S. in the currently available public literature,

the measured PFOS concentrations in ambient surface waters occur over a wide range. Also, the increased frequency of measured PFOS concentrations at sites with no known previous PFAS exposure or sites that were not identified by the individual study authors as either sites with or without previous exposure (Table 2 and Figure 4), may skew the measured PFOS concentrations in U.S. ambient surface waters toward the lower end of the wide range of measured concentrations. To better understand the occurrence of PFOS in ambient surface waters across the U.S, additional data from previously exposed sites and in urban areas, particularly those in areas where little or no data are available, are needed. Specifically, a systematic study focused on measuring PFAS (including PFOS, PFOA and their precursors and shorter chain PFAS) in ambient surface waters across the U.S. is needed to eliminate potential bias from differences in analytical methods, sample collection, and/or location, and to fill in existing data gaps. Filling these data gaps would provide a more robust understanding of the spatial and temporal variability of PFOS occurrence in U.S. ambient surface waters. Given the widespread use of PFAS and the persistence of PFOS in the aquatic environment, a thorough understanding of the total environmental distribution of PFAS in surface waters (particularly of PFOS itself and the volatile compounds that can transform PFSA) is needed to fully understand the occurrence of PFOS in the environment and any potential risks it may pose in aquatic ecosystems.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Data availability statement—

Data, associated metadata, and calculation tools are available from the corresponding author (jarvis.amanda@epa.gov).

REFERENCES

3M Environmental Laboratory. 2011. Environmental monitoring – multi-city study water, sludge, sediment, POTW effluent and landfill leachate samples. Executive Summary. https://static.ewg.org/files/MultiCity_execsum.pdf

Ahrens L 2011. Polyfluoroalkyl compounds in the aquatic environment: a review of their occurrence and fate. J. Environ. Monit 13:20–31. [PubMed: 21031178]

Ahrens L Yeung LWY, Taniyasu S, Lam PKS, Yamashita N 2011. Partitioning of perfluorooctanoate (PFOA), perfluorooctane sulfonate (PFOS) and perfluorooctane sulfonamide (PFOSA) between water and sediment. Chemosphere. 85:731–737. [PubMed: 21742366]

Ahrens L and Bundschuh M. 2014. Fate and effects of poly- and perfluoroalkyl substances in the aquatic environment: a review. Environ. Toxicol. Chem 33:1921–1929. [PubMed: 24924660]

Allred BM, Lang JR, Barlaz MA, Field JA 2015. Physical and biological release of poly- and perfluoroalkyl substances (PFASs) from municipal solid waste in anaerobic model landfill reactors. Environ Sci Technol. 49:768–7656.

Anderson RH, Long GC, Porter RC, Anderson JK. 2016. Occurrence of select perfluoroalkyl substances at U.S. Air Force aqueous film-forming foams release sites other than fire-training areas: field-validation of critical fate and transport properties. Chemosphere. 150: 678–685. [PubMed: 26786021]

Ankley GT, Cureton P, Hoke RA, Houde M, Kumar A, Kurias J, Lanno R, McCarthy C, Newsted J, Salice CJ, Sample BE, Sepúlveda Steevens J, Valsecchi S 2020. Assessing the ecological risks of Per- and Polyfluoroalkyl substances: current state-of-the science and a proposed path forward. Environ. Toxicol. Chem 40: 564–605. [PubMed: 32897586]

Beach SA, Newsted JL, Coady K, Giesy JP. 2006. Ecotoxicological evaluation of perfluorooctanesultonate (PFOS). Rev Environ Contam Toxicol. 186:133–174. [PubMed: 16676904]

Benskin PJ, Mui DCG, Scott BF, Spencer C, De Silva AO, Kylin H, Martin JW, Morris A, Lohmann R, Tomy G, Rosenberg B, Taniyasu S, Yamashita N. 2012. Perfluoroalkyl acids in the Atlantic and Canadian Arctic oceans. Environ Sci Technol. 46:5815–5823. [PubMed: 22548373]

Benskin JP. Holt A, Martin JW 2009. Isomer-specific biotransformation rates of a perfluoruooctane sulfonate (PFOS)-precursor by cytochrome P450 isozymes and human liver miscrosomes. Environ Sci Technol 43: 8566–8572. [PubMed: 20028053]

Boulanger B, Vargo JD, Schnoor JL, Hornbuckle KI. 2005. Evaluation of perfluorooctane surfactants in wastewater treatment system and in commercial surface protection product. Environ Sci Technol. 39: 5524–5530. [PubMed: 16124283]

Boulanger B, Vargo J, Schnoor JL, Hornbuckle KC 2004. Detection of perfluorooctane surfactants in Great Lakes Water. Enrion Sci Technol 38:4064–4070.

Buck RC, Franklin J, Berger U, Conder JM, Cousins IT, De Voogt P, Jensen AA, Kannan K, Mabury SA, Van Leeuwen SPJ. 2011. Perfluoroalkyl and polyfluoroalkyl substances in the environment: Terminology, classification, and origins. Integr Environ Assess Manag. 7:513–541. [PubMed: 21793199]

Burkhard LP. 2021. Evaluation of published bioconcentration factor (BCF) and bioaccumulation factor (BAF) data for Per- and Polyfluoroalkyl substances across aquatic species. Environ. Toxicol. Chem 40: 1530–1543. [PubMed: 33605484]

Cochran RS. 2015. Evaluation of perfluorinated compounds in sediment, water, and passive samplers collected from the Barksdale Air Force Base. MS Thesis. Texas Tech University, Lubbock, Texas United States.

Colorado Department of Public Health and the Environment. 2020. PFAS sampling project results: surface water. https://cohealthviz.dphe.state.co.us/t/EnvironmentalEpidemiologyPublic/views/PFAS_results_DRAFT/Samplelocationsdash?%3AshowAppBanner=false&%3Adisplay_count=n&%3AshowVizHome=n&%3Aorigin=viz_share_link&%3AisGuestRedirectFromVizportal=y&%3Aembed=y

De Silva AO, Spencer C, Scott BF, Backus S, Muir DCG. 2011. Detection of a cyclic perfluorinated acid, perfluoroethylcyclohexane sulfonate, in the Great Lakes of North America. Environ Sci Technol. 45:8060–8066. [PubMed: 21528907]

Dinglasan-Panlilio M, Prakash SS, Baker JE 2014. Perfluorinated compounds in the surface waters of Puget Sound, Washington and Clayoquot and Barkley Sounds, British Columbia. Marine Pollut Bullet. 78:173–180.

Environment and Climate Change Canada (ECCC). 2018. Federal Environmental Quality Guidelines Perfluorooctance Sulfonate (PFOS). https://www.canada.ca/en/environment-climate-change/services/evaluatingexisting-substances/federal-environmental-quality-guidelines-perfluorooctanesulfonate.html

Furdui VI, Crozier PW, Reiner EJ, Mabury SA. 2008. Trace level determination of perfluorinated compounds in water by direct injection. Chemosphere. 73:524–530.

Gewurtz SB, Backus SM, De Silva AO, Ahrens L, Armellin A, Evans M, Fraser S, Gledhill M, Guerra P, Harner T, Helm PA, Hung H, Khera N, Kim MG, King M, Lee SC, Letcher RJ, Martin P, Marvin C, McGoldrick DJ, Myers AL, Pelletier M, Pomeroy J, Reiner EJ, Rondeau M, Sauve MC, Sekela M, Shoeib M, Smith DW, Smyth SA, Struger J, Spry D, Syrgiannis J, Waltho J. 2013. Perfluoroalkyl acids in the Canadian environment: Multi-media assessment of current status and trends. Environ Int 59:183–200. [PubMed: 23831544]

Giesy JP, Naile JE, Khim JS, Jones PD, Newsted JL. 2010. Aquatic toxicology of perfluorinated chemicals. Rev Environ Contam Toxicol 202:1–52. [PubMed: 19898760]

Hansen KJ, Johnson HO, Eldridge JS, Butenhoff JL, Dick LA. 2002. Quantitative characterization of trace levels of PFOS and PFOA in the Tennessee River. Environ Sci Technol. 36:1681–1685. [PubMed: 11993863]

Houde M, Martin JW, Letcher RJ, Solomon KR, Muir DCG. 2006. Biological monitoring of polyfluoroalkyl substances: A review. Environ Sci Technol 40:3463–3473. [PubMed: 16786681]

Houde M, De Silva AO, Muir DCG, Letcher RJ. 2011. Monitoring or perfluorinated compounds in aquatic biota: an updated review. Environ Sci Technol. 45:7962–7973. [PubMed: 21542574]

Houtz EF, Sutton R, Park J, Sedlak M. 2016. Poly- and perfluoroalkyl substances in wastewater: significance of unknown precursors, manufacturing shifts, and likely AFFF impacts. Water Res. 95: 142–149. [PubMed: 26990839]

Huset CA, Chiaia AC, Barfosky DF, Jonkers N, Kohler H- PE, Ort C, Giger W, Field JA. 2008. Occurrence and mass flows of fluorochemicals in the Glatt Valley watershed, Switzerland. Environ Sci Technol 42:6369–6377. [PubMed: 18800503]

Kannan K, Tao L, Sinclair E, Pastva SD, Jude DJ, Giesy JP. 2005. Perfluorinated compounds in aquatic organisms at various trophic levels in a Great Lakes food chain. Arch Environ Contam Toxicol. 48: 559–566. [PubMed: 15883668]

Kannan K 2011. Perfluoroalkyl and polyfluoroalkyl substances: current and future perspectives. Environ Chem. 8:333–338.

Kim S and Kannan K. 2007. Perfluorinated acids in air, rain, snow, surface runoff, and lakes: relative importance of pathways to contamination of urban lakes. Environ Sci Technol. 41:8328–8334. [PubMed: 18200859]

Konwick BJ, Tomy GT, Ismail N, Peterson JT, Fauver RJ, Higginbotham D, Fisk AT. 2008. Concentrations and patterns of perfluoroalkyl acids in Georgia, USA surface waters near and distant to a major use source. Environ Toxicol Chem. 27:2011–2018. [PubMed: 18419175]

Kwadijk CJAF, Korytar P, Koelmans AA. 2010. Distribution of perfluorinated compounds in aquatic systems in The Netherlands. Environ Sci Technol 44:3746–3751. [PubMed: 20426438]

Lang JR, Allred BM, Peaslee GF, Field JA, Barlaz MA. 2016. Release of per- and polyfluoroalkyl substances (PFASs) from carpet and clothing in model anaerobic landfill reactors. Envrion Sci Technol 50: 5024–5032.

Lange CC. 2000. The aerobic biodegradation of N-EtFOSE alcohol by the microbial activity present in municipal wastewater treatment sludge. The 3M Company. St. Paul, MN.

Lasier PJ, Wahsington JW, Hassan SM, Jenkins TM. 2011. Perfluorinated chemicals in surface water and sediments from northwest Georgia, USA, and their bioaccumulation Lumbriculus variegatus. Environ Toxicol Chem. 20(10): 2194–2201.

Lau C, Anitole K, Hodes C, Lai D, Pfahles-Hutchens A, Seed J. 2007. Perfluoroalkyl acids: a review of monitoring and toxicological findings. Toxicol Sci. 99:366–394. [PubMed: 17519394]

Lehmler H 2005. Synthesis of environmentally relevant fluorinated surfactants – a review. Chemosphere. 58:1471–1496. [PubMed: 15694468]

Lindstrom A, Strynar M, Libelo EL. 2011. Polyfluorinated compounds: past, present, and future. Environ. Sci. Technol 45:7954–7961. [PubMed: 21866930]

Liu X, Guo Z, Krebs KA, Pope RH, Roache NF. 2014. Concentrations and trends of perfluorinated chemicals in potential indoor sources from 2007 through 2011 in the US. Chemosphere 98: 51–57. [PubMed: 24268172]

Liu J and Avendaño SM 2013. Microbial degradation of polyfluoroalkyl chemicals in the environment: a review. Environ Intern 61:98–114.

Loos R, Wollgast J, Huber T, Hanke G 2007. Polar herbicides, pharmaceutical products, perfluorooctanesulfonate (PFOS), perfluorooctanoate (PFOA), and nonylphenol and its carboxylates and ethoxylates in surface and tap waters around Lake Maggiore in northern Italy. Annul Bioanal Chem. 387:1469–1478.

Loos R, Gawlik BM, Locoro G, Rimaviciute E, Contini S, Bidoglio G. 2009. EU-wide survey of polar organic persistent pollutants in European river waters. Environ Pollut 157:561–568. [PubMed: 18952330]

Möller A, Ahren L, Sturm R, Ebinghaus R. 2009. Identification of point sources of polyfluoroalkyl compounds (PFCs) along the River Rhine watershed and their transportation into the North Sea. Coastline Reports. 13–143–154.

Moody CA, Martin JW, Kwan WC, Muir DCG, Mabury SA. 2002. Monitoring perfluorinated surfactants in biota and surface water samples following an accidental release of fire-fighting foam into Etobicoke Creek. Environ Sci Technol 36:545–551. [PubMed: 11883418]

Nakayama S, Strynar MJ, Helfant L, Egeghy P, Ye X, Lindstrom AB. 2007. Perfluorinated compounds in the Cape Fear Drainage Basin in North Carolina. Environ Sci Technol. 41:5271–5276. [PubMed: 17822090]

New Jersey Department of Environmental Protection Divison of Science, Research, and Environmental Health. 2019. Investigation of levels of perfluorinated compounds in New Jersey fish, surface water, and sediment. SR15–010. https://www.nj.gov/dep/dsr/publications/Investigation%20of%20Levels%20of%20Perfluorinated%20Compounds%20in%20New%20Jersey%20Fish,%20Surface%20Water,%20and%20Sediment.pdf

New Mexico Environment Department. 2020–2021. https://www.env.nm.gov/pfas/data/

Newsted JL, Holem R, Hohenstein G, Lange C, Ellefson M, Reagen W, Wolf S. 2017. Spatial and temporal trends of poly- and perfluoroalkyl substances in fish fillets and water collected from pool 2 of the upper Mississippi River. Environ Toxicol Chem. 36:3138–3147. [PubMed: 28628233]

Nguyen VT, Reinhard M, Karina GY. 2011. Occurrence and source characterization of perfluorochemicals in urban watershed. Chemosphere. 82: 1277–1285. [PubMed: 21208640]

[OECD] Organisation for Economic Co-operation and Development. 2002. Hazard assessment of perfluorooctane sulfonate (PFOS) and its salts. Report ENV/JM/RD(2002)17/FINAL.

Oliaei F Kriens D Weber R, Watson A 2013. PFOS and PFC releases and associated pollution from a PFC production plant in Minnesota (USA). Environ Sci Pollut Res. 20: 1977–1992.

Pan Y, Zhang H, Cui Q, Sheng N, Yeung LWY, Sun Y, Guo Y, Dai J. 2018. Worldwide distribution of novel perfluoroether carboxylic and sulfonic acids in surface water. Environ Sci Technol. 52:7621–7629. [PubMed: 29749740]

Paul AG, Jones KC, Sweetman AJ. 2009. A first global production, emission, and environmental inventory for perfluorooctane sulfonate. Environ Sci Technol 43:386–392. [PubMed: 19238969]

Plumlee MH, McNeil K, Reinhard M. 2009. Indirect photolysis of perfluorochemicals: hydroxyl radical-initiated oxidation of N-ethyl perfluorooctane sulfonamido acetate (N-EtFOSAA) and other perfluoroalkanesulfonamides. Environ Sci Technol. 43: 3662–3668. [PubMed: 19544870]

Plumlee MH, Larabee J, Reinhard M. 2008. Perfluorochemicals in water reuse. Chemosphere. 72:1541–1547. [PubMed: 18547612]

Prevedouros K, Cousins IT, Buck RC, Korzeniowski SH. 2006. Sources, fate and transport of perfluorocarboxylates. Environ Sci Technol 40:32–44. [PubMed: 16433330]

Remucal CK. 2019. Spatial and temporal variability of perfluoroalkyl substances in the Laurentian Great Lakes. Environ Sci Processes Impacts. 11:1816–1834.

Rankin K, Mabury SA, Jenkins TM, Washington JW. 2016. A North American and global survey of perfluoroalkyl substances in surface soils: distribution patterns and mode of occurrence. 161: 331–341.

Rhoads KR, Janssen EML, Luthy RG, Criddle CS. 2008. Aerobic biotransformation and fate of N-ethyl perfluorooctane sulfonamidoethanol (N-EtFOS) in activated sludge. Environ Sci Technol. 42: 2873–2878. [PubMed: 18497137]

Saito N, Sasaki K, Nakatome K, Harada K, Yoshinaga T, Koizumi A. 2003. Perfluorooctane sulfonate concentrations in surface water in Japan. Arch Environ Contam Toxicol. 45:149–158. [PubMed: 14565571]

Scott BF, Spencer C, Lopez E, Muir DCG. 2009. Perfluorinated alkyl acid concentrations in Canadian rivers and creeks. Water Qual Res J Can. 44:263–277.

Scott BF, De Silva AO, Spencer C, Lopez E, Backus SM, Muir DCG. 2010. Perfluoroalkyl acids in Lake Superior water: trends and sources. J. Great Lake Res 36:277–284.

Schultz MM, Higgins CP, Huset CA, Luthy RG, Barofsky DF, Field JA 2006. Fluorochemical mass flows in a municipal wastewater treatment facility. Envion Sci Technol. 40–7350–7357.

Sedlak MD. Benskin JP, Wong A, Grace R, Greig DJ 2017. Per- and polyfluoroalkyl substances (PFASs) in San Francisco Bay wildlife: temporal trends, exposure pathways, and notal presence of precursor compounds. Chemosphere. 185:1217–1226. [PubMed: 28477851]

Simcik MF and Dorweiler KJ. 2005. Ratio of perfluorochemical concentrations as a tracer of atmospheric deposition to surface water. Environ Sci Technol. 39:8678–8683. [PubMed: 16323762]

Sinclair E and Kannan K. 2006. Mass loading and fate of perfluoroalkyl surfactants in wastewater treatment plans. Environ Sci Technol. 40:1408–1414 [PubMed: 16568749]

Sinclair E, Mayack DT, Roblee K, Yamashita N, Kannan K. 2006. Occurrence of perfluoroalkyl surfactants in water, fish, and birds from New York State. Arch Environ Contam Toxciol. 50:398–410.

Smithwick M, Norstrom RJ, Mabury SA, Solomon K, Evans TJ, Stirling I, Taylor MK, Muir DCG. 2006. Temporal trends of perfluoroalkyl contaminants in polar bears Ursus maritimus from two locations in the North American arctic. Environ. Sci. Technol 40:1139–1143. [PubMed: 16572767]

Stahl LL. Snyder BD, Olsen AR, Kincaid TM, Wathen JB, McCarty HB 2014. Perfluorinated compounds in fish from U.S. urban rivers and the Great Lakes. Sci Total Environ. 499: 185–195. [PubMed: 25190044]

U.S. EPA. National Coastal Condition Assessment. https://www.epa.gov/nationalaquatic-resource-surveys/ncca

U.S. EPA. Monitoring the occurrence of unregulated drinking water contaminants. https://www.epa.gov/dwucmr.

U.S. EPA. ECOTOX Knowledgebase. https://cfpub.epa.gov/ecotox/

U.S. EPA. 2020. Long-Chain Perfluoroalkyl Carboxylate and Perfluoroalkyl Sulfonate Chemical Substances; Significant New Use Rule. Office of Pollution Prevention and Toxics. EPA-HQ-OPPT-2013–0225; FRL-10010–44. https://www.epa.gov/sites/production/files/2020-06/documents/10010-44_prepubcopy_fr_doc_epa_admin_esign_2020-06-22final.pdf

Wang Z, DeWitt JC, Higgins CP, Cousins IT. 2017. A never-ending story of per- and polyfluoroalkyl substances (PFASs)? Environ. Sci. Technol 51:2508–2518. [PubMed: 28224793]

Wang Z, Cousins IT, Scheringer M, Buck RC, Hungerbühler K. 2014. Global emission inventories C4 – C14 perfluoroaklyl carboxylic acid (PFCA) homologues from 1951 to 2030, part I: production and emission from quantifiable sources. Environ Internat 60: 62–75.

Wang Z, Cousins IT, Scheringer M, Buck RC, Hungerbühler K. 2014. Global emission inventories for C4 – C14 perfluoroaklyl carboxylic acid (PFCA) homologues from 1951 to 2030, part II: the remaining pieces of the puzzle. Environ Internat 60:166–176.

Wang Z, Cousins IT, Scheringer M, Hungerbühler K 2013. Fluorinated alternatives to long-chain perfluoroalkyla carboxylic acids (PFCAs), perfluoroalkane sulfonic acids (PFSAs) and their potential percursors. Environ Internat. 60: 242–248.

Washington JW, and Jenkins TM. 2015. Abiotic hydrolysis of fluorotelomer polymers as a source of perfluorocarboxylates at the global scale. Environ Sci Technol 49:14129–14135.

Washington JW, Jenkins TM, Rankin K, Naile JE. 2015. Decades-Scale Degradation of Commercial, Side-Chain, Fluorotelomer-based Polymers in Soils & Water. Environ Sci Tech 49(2):915–923.

Xu X Xu H, Zhang D, Shen X, Tong G, Lu Y, Wang W 2013. Occurrence of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in aquatic systems from Asia. Appl Mechan and Materials. 295–298:513–519.

Zareitalabad P, Siemens J, Hamer M, Amelung W. 2013. Perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) in surface waters, sediments, soils and wastewater – a review on concentrations and distribution coefficients. Chemosphere. 91:725–732. [PubMed: 23498059]

Zhang X, Lohmann R, Dassuncao C, Hu XC, Weber AK, Vecitis CD, Sunderland EM. 2016. Source attribution of poly- and perfluoroalkyl substances (PFASs) in surface waters from Rhode Island and the New York Metropolitan Area. Environ. Sci. Technol. Lett 3:316–321. [PubMed: 28217711]

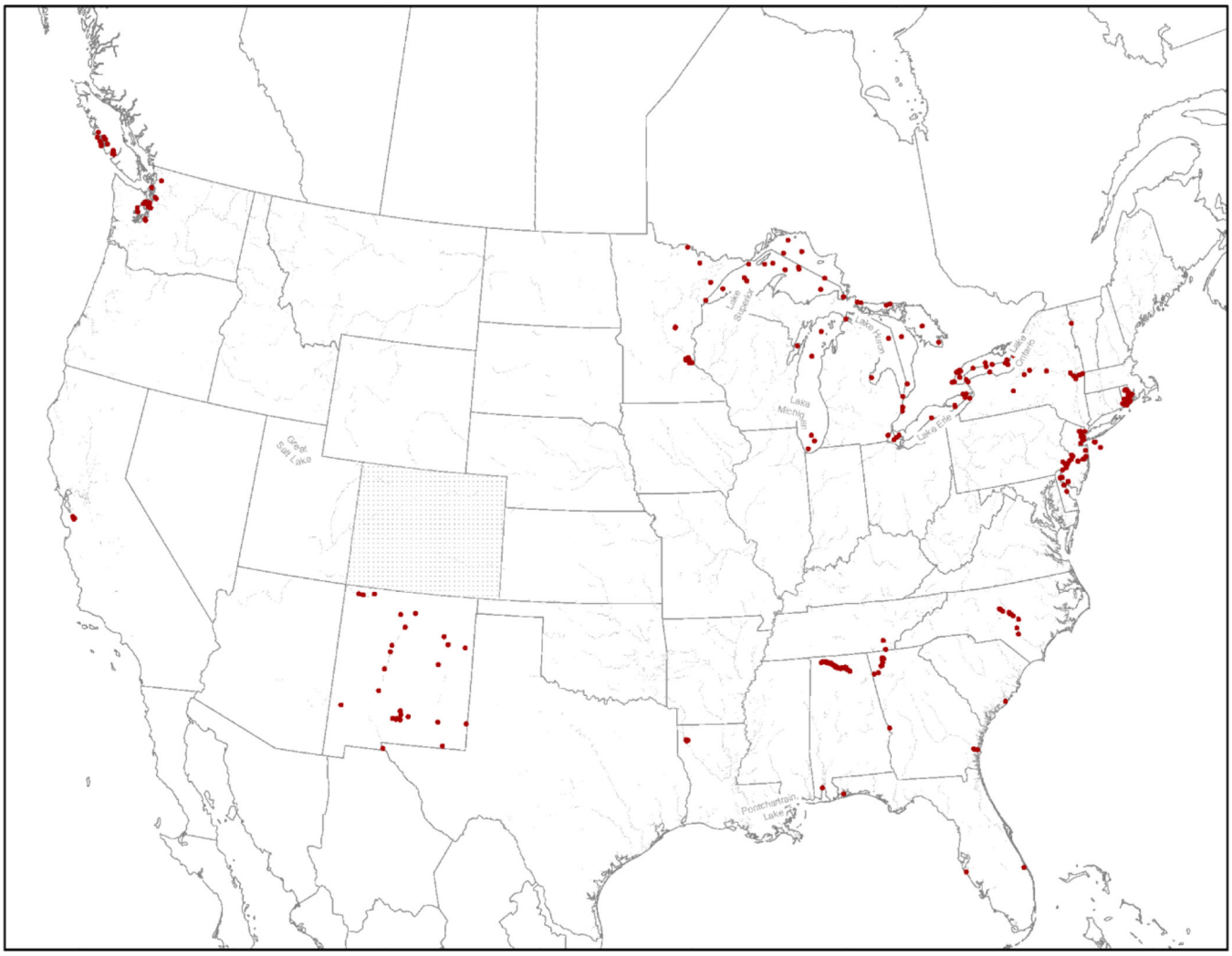

Figure 1.

Map indicating sampling locations for perfluorooctane sulfonate (PFOS) measured in surface waters across the United States (U.S.) based on data reported in the current, publicly available literature. Sampling locations for the Colorado data were not available and these data are represented by the dash marks to indicate measured PFOS surface water concentrations are available. Detailed information on sampling locations, including references, coordinates, and sampling site identification numbers and names, provided in Supplemental Data, Figure S1 and Table S1.

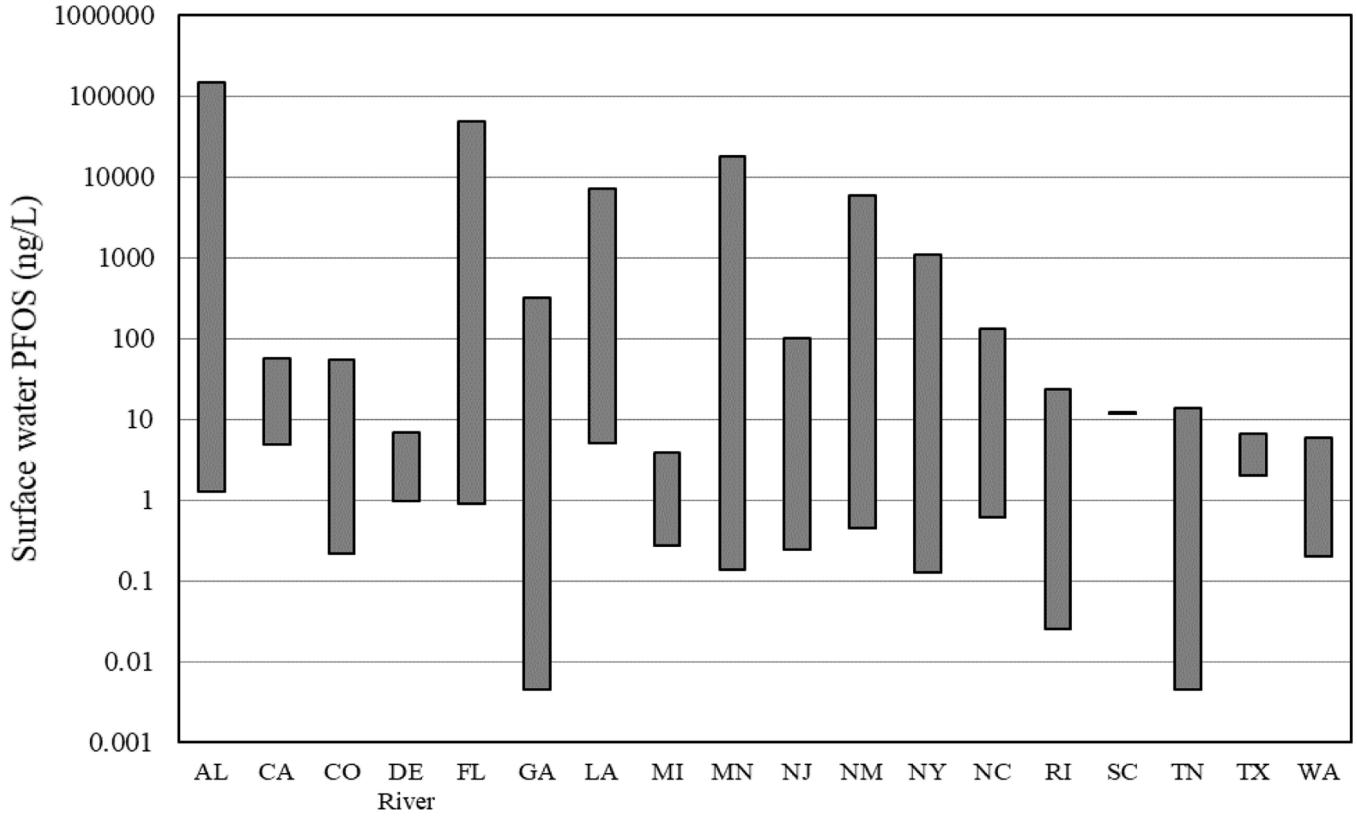

Figure 2.

Distribution of the minimum and maximum concentrations (ng/L) of perfluorooctane sulfonate (PFOS) measured in surface waters for each state or waterbody (excluding the Great Lakes) with reported data in the current, publicly available literature and is not necessarily comprehensive of PFOS concentrations in surface waters across each state. The distribution is arranged alphabetically by state and waterbody.

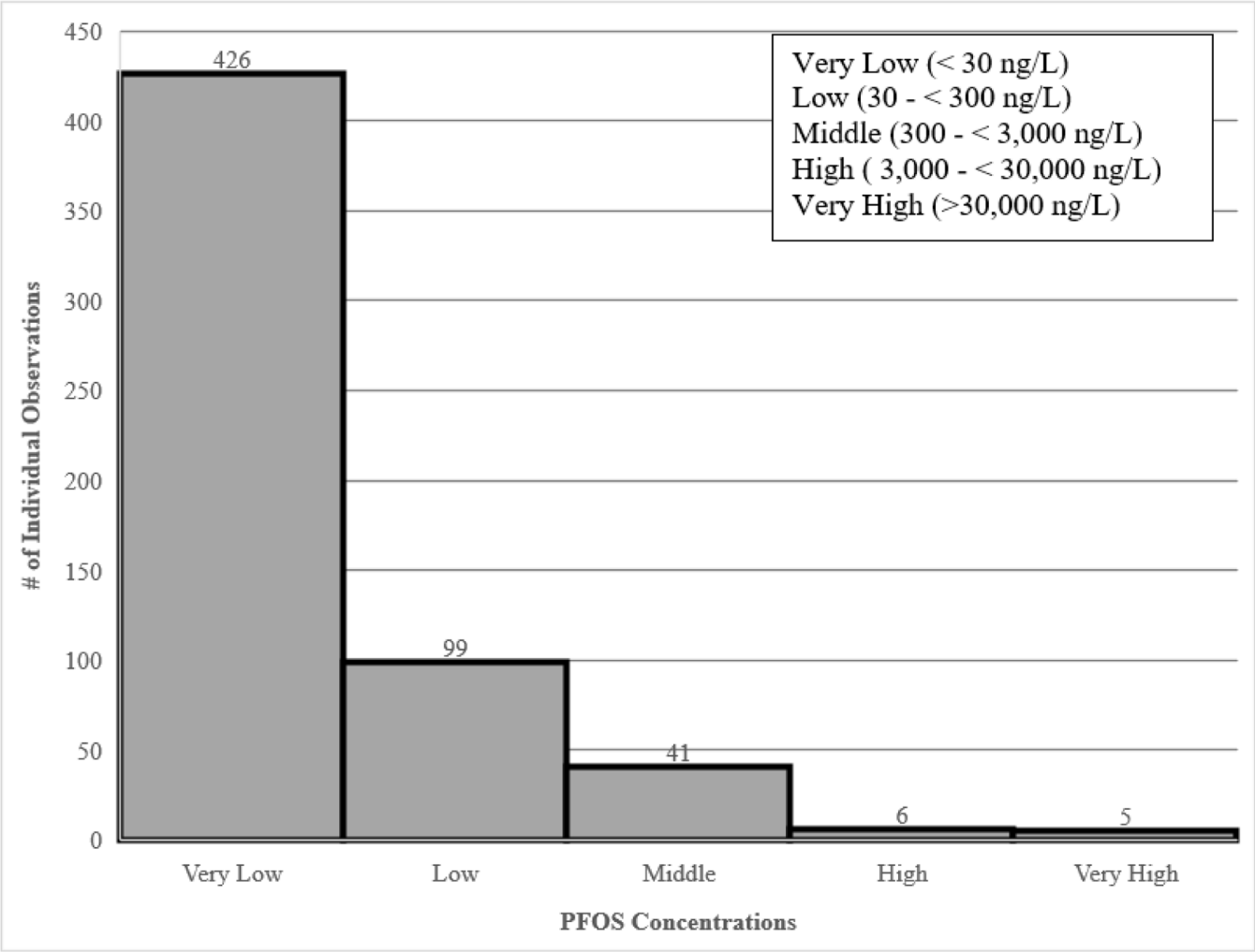

Figure 3.

Number of individual observations of measured perfluorooctane sulfonate (PFOS) concentrations that are very low (< 30 ng/L), low (30 – < 300 ng/L), middle (300 – < 3,000 ng/L), high (3,000 – < 30, 0000 ng/L), and very high (> 30,000 ng/L) in ambient surface waters across the United States. The bins of PFOS concentrations were determined from the currently available toxicity literature for PFOS.

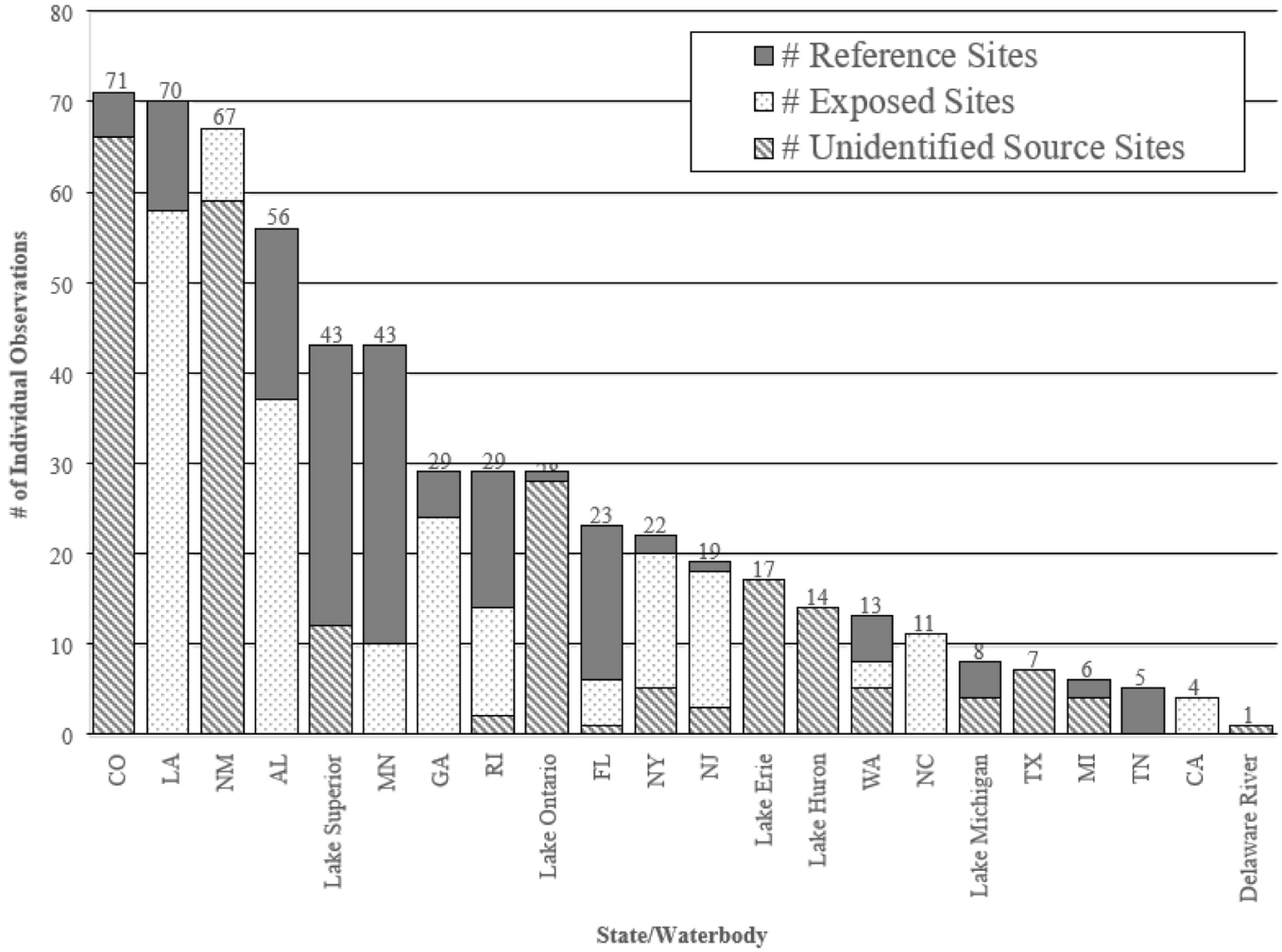

Figure 4.

Distribution of individual observations among sites with no known previous exposure to PFASs (identified as Reference Sites by individual study authors, Exposed (sites with known previous exposure to PFASs and also identified as such by individual study authors), and Unidentified (sites in which the study authors did not specify sites as reference or potential exposure to PFASs) ambient surface water sites in the current, publicly available for PFOS occurrence data and is not necessarily comprehensive of PFOS concentrations in surface waters across each state.. The distribution is arranged by highest to lowest number of individual observations and grouped by state or waterbody.

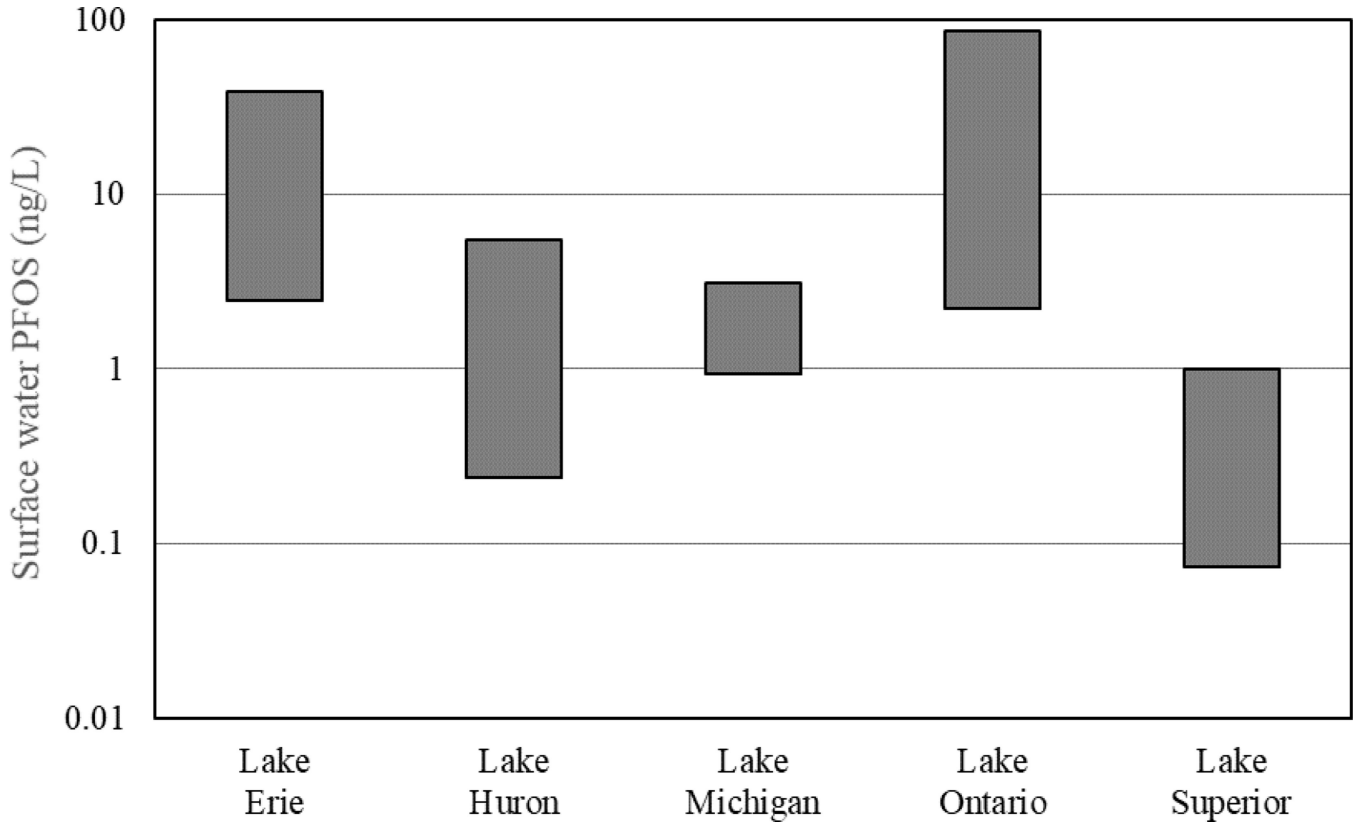

Figure 5.

Distribution of the minimum and maximum concentrations (ng/L) of perfluorooctane sulfonate (PFOS) measured in surface water samples collected from the Great Lakes as reported in the current, publicly available literature and is not necessarily comprehensive of PFOS concentrations in surface waters across each state.. This distribution is arranged alphabetically by waterbody.

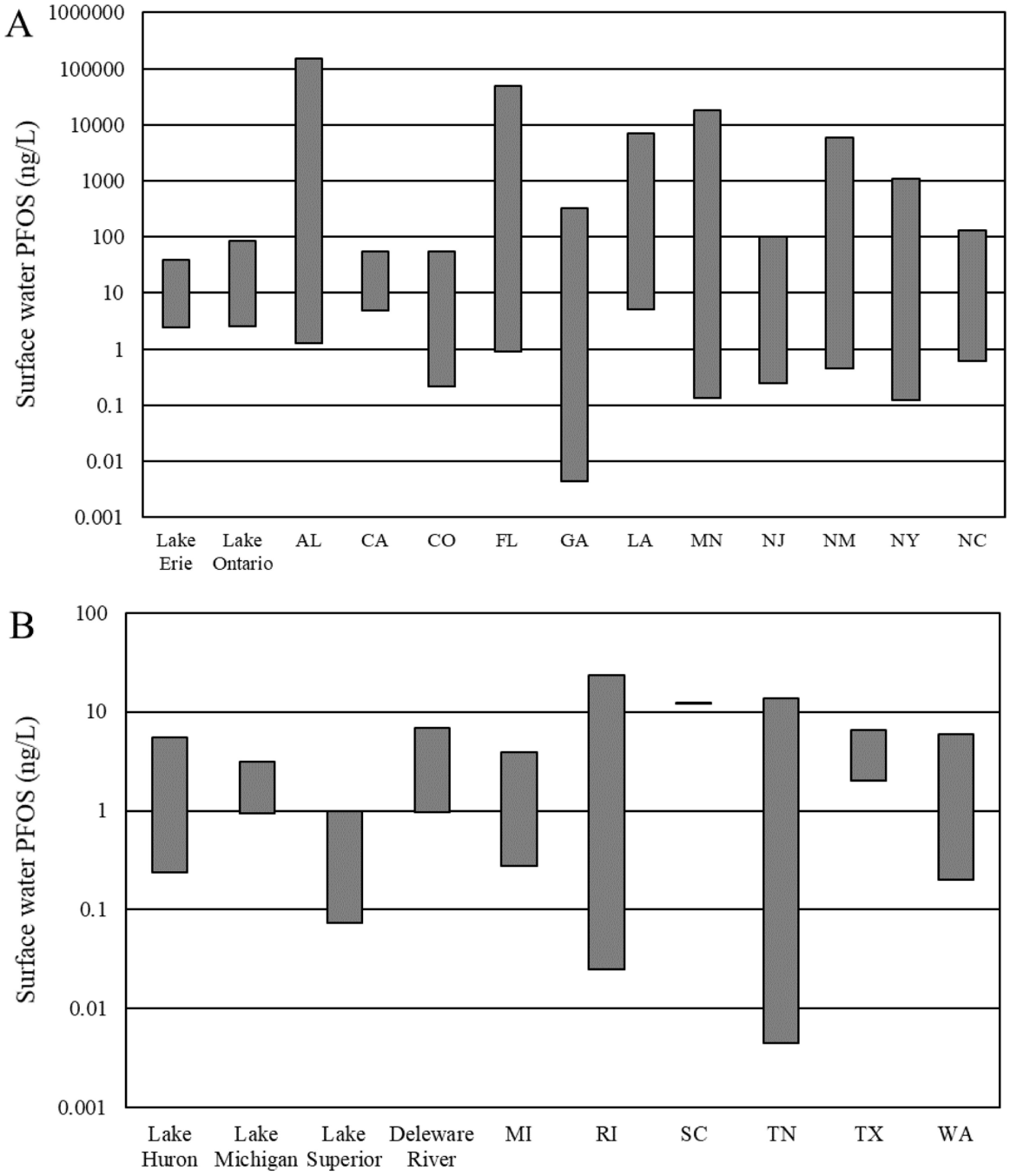

Figure 6.

Comparison of relatively high (A; greater than 30 ng/L) and low (B; less than 30 ng/L) maximum perfluorooctane sulfonate (PFOS) concentrations (ng/L) measured in surface water samples collected across the United States (U.S.) as reported in the current, publicly available literature and is not necessarily comprehensive of PFOS concentrations in surface waters across each state.. The relatively high PFOS concentrations were associated

with specific nearby consumer and/or industrial source. Both distributions are arranged alphabetically by waterbody or state.

Table 1.

Current Publicly Available Measured Perfluorooctane Sulfonate (PFOS) Concentrations in Surface Waters Across the United States (U.S.). Additional details, including study specific sampling dates, number of measurements, and limits of detection and quantification, provided in Supplemental Data, Table S2.

State	Waterbody[1]	Arithmetic Mean PFOS Concentration (ng/L)[2]	Median PFOS Concentration (ng/L)[2]	Range of PFOS Concentration (ng/L)	Reference
	Lake Erie	3.77	3	2.8 – 5.5	Sinclair et al. (2006)
		31.3	32.5	21.5 – 38.5	Boulanger et al. (2004)
		2.84	2.63	2.49 – 3.41	De Silva et al. (2011)
	Lake Huron	4.5	4.2	4.0 – 5.3	Furdui et al. (2008)
		2.25	1.96	0.239 – 5.46	De Silva et al. (2011)
		1.73	1.5	1.2 – 2.7	Furdui et al. (2007)
	Lake Michigan	2.03	2.03	0.93 – 3.13	Simcik and Dorweiler (2005)
		2.00	1.96	1.73 – 2.36	De Silva et al. (2011)
		not provided	4.9	2.9 – 30	(Sinclair et al.2006)
	Lake Ontario	55.4	59.8	16.5 – 85.5	Boulanger et al. (2004)
		5.96	5.63	2.60 – 9.48	De Silva et al. (2011)
		8.69	6.6	3.6 – 37.6	Furdui et al. (2008)
		2.20	not provided	not provided	Houde et al. 2008
	Lake Superior	0.255	0.236	0.095 – 0.395	De Silva et al. (2011)
		0.233	0.3	0.1 – 0.3	Furdui et al. (2008)
		0.246	0.124	0.074 – 0.996	Scott et al. (2010)
Alabama	Waterbody near Decatur	58,016	41,027	9 – 150,000	OECD (2002)
	Waterbody in Decatur	2.5 < x < 25	2.5 < x < 25	2.5 < x < 25	3MCompan y (2001)
	Pond in Decatur	111	111	111	
	Waterbody in Mobile	30.3	35.5	< 25 – 41.5	3MCompan y (2001)
	Pond in Mobile	32.5	32.5	32.5	
	Tennessee River (upstream of Baker's Creek)	30.85	29.80	16.0 – 52.6	Hansen et al. (2002)
	Tennessee River (downstream of Baker's Creek)	103.9	107.0	30.3 – 144	Hansen et al. (2002)
California	Upper Silver Creek	not provided	not provided	27 – 56	Plumlee et al. (2008)
	Coyote Creek	not provided	not provided	4.8 – 25	
	Animas River	< 0.48	< 0.48	< 0.48	
	Arkansas River	1.96	0.62	0.23 – 5.00	

State	Waterbody[1]	Arithmetic Mean PFOS Concentration (ng/L)[2]	Median PFOS Concentration (ng/L)[2]	Range of PFOS Concentration (ng/L)	Reference
Colorado	Arvada Blunn Reservoir	0.77	0.77	0.77	Colorado Department of Public Health and the Environment 2020
	Barker Reservoir	< 0.49	< 0.49	< 0.49	
	Bessemer Ditch	14.0	14.0	14.0	
	Big Thompson River	3.90	3.90	3.90	
	Blue River	1.20	1.20	1.20	
	Boulder Feeder Canal	< 0.45	< 0.45	< 0.45	
	Boyd Lake	1.00	1.00	1.00	
	Cache la Poudre River	5.61	5.61	< 0.45 – 11.0	
	Clear Creek	7.95	7.95	7.20 – 8.70	
	Colorado River	0.67	0.66	0.65 – 0.69	
	Coon Creek	< 0.48	< 0.48	< 0.48	
	Eagle River	0.68	0.68	0.68	
	East Plum Creek	< 0.43	< 0.43	< 0.43	
	Erie Lake	3.70	3.70	3.70	
	Fairmount Reservoir	< 2.50	< 2.50	< 2.50	
	Fountain Creek	16.9	20.0	3.50 −24.0	
	Fraser River	1.00	1.00	1.00	
	Gore Creek	0.98	0.98	0.98	
	Gunnison River	0.71	0.71	0.71	
	Horsetooth Reservoir	0.51	0.51	0.51	
	Jackson Creek	< 0.44	< 0.44	< 0.44	
	Jerry Creek	< 0.485	< 0.485	< 0.48 - < 0.49	
	Kannah Creek Flowline	< 0.49	< 0.49	< 0.49	
	Lakewood Reservoir	< 0.45	< 0.45	< 0.45	
	Little Fountain Creek	< 0.46	< 0.46	< 0.46	
	Maple Grove Reservoir	10.0	10.0	10.0	
	Marstron Reservoir	0.48	0.48	0.48	
	McBroom Ditch	4.90	4.90	4.90	
	Mclellen Reservoir	1.30	1.30	1.30	
	Mesa Creek	< 0.49	< 0.49	< 0.49	
	Michigan River	< 0.46	< 0.46	< 0.46	
	Molina Power Plant Tail	< 0.50	< 0.50	< 0.50	
	North Fork Gunnison River	< 0.47	< 0.47	< 0.47	
	Purdy Mesa Flowline	< 0.49	< 0.49	< 0.49	
	Purgatoire River	0.47	0.47	0.47	
	Ralston Reservoir	< 0.46	< 0.46	< 0.46	
	Rio Grande	< 0.47	< 0.47	< 0.47	
	Roaring Fork River	< 0.50	< 0.50	< 0.50	

State	Waterbody[1]	Arithmetic Mean PFOS Concentration (ng/L)[2]	Median PFOS Concentration (ng/L)[2]	Range of PFOS Concentration (ng/L)	Reference
	San Juan River	< 0.44	< 0.44	< 0.44	
	Sand Creek	30.3	30.3	6.50 – 54.0	
	Severy Creek	< 0.47	< 0.47	< 0.47	
	Somerville Flowline	< 0.48	< 0.48	< 0.48	
	South Boulder Creek	0.50	0.50	0.50	
	South Platte River	10.5	11.5	3.80 – 16.0	
	St. Vrain River	3.90	3.90	3.90	
	Strontia Springs	< 0.51	< 0.51	< 0.51	
	Taylor River	< 0.45	< 0.45	< 0.45	
	Uncompahgr e River (delta)	0.54	0.54	0.54	
	Welton Reservoir	2.60	2.60	2.60	
	White River	< 0.46	< 0.46	< 0.46	
	Yampa River	< 0.47	< 0.47	< 0.47	
Delaware, New Jersey, Pennsylvania	Delaware River	3.98	3.5	0.97 – 6.92	Pan et al. (2018)
	Waterbody in Pensacola	16.29	2.5 < x < 25	<25 – 29	
	Pond in Pensacola	2.5 < x < 25	2.5 < x < 25	2.5 < x < 25	
	Waterbody in Port St.	50.83	2.5 < x < 25	< 2.5 – 137.5	3MCompan y (2001)
Florida	Lucie Small pond in Port St. Lucie[3]	9,784	1,945	1,830 – 48,200	
	Sarasota Bay	0.90	not provided	not provided	Houde et al. 2006
	Waterbody in Columbus	59.9	55	44.6 – 80	3Mcompan y (2001)
	Pond in Columbus	< 2.5	< 2.5	< 2.5	
	Conasauga River	162.1	192	< 1.5 – 321	
	Altamaha River	2.63	2.6	2.6 – 2.7	Konwick et al. (2008)
Georgia	Streams and ponds in Dalton	70.36	70.73	10.5–119.5	
	Oostanaula River	150.3	151	148 – 152	Lasier et al. (2011)
Louisiana	Waterbodies (locations of concern) near Barksdale A.F.B.	776.7	195.0	< 10 – 7,070	Cochran (2015); Lanza et al. (2017)
	Reference waterbodies near Barksdale A.F.B.	< 10	< 10	< 10	
	Raisin River	3.5	3.5	3.5	Kannan et al. (2005)
Michigan	St Clair River	2.6	2	1.9 – 3.9	
	Siskiwit Lake	0.283	0.283	0.277 – 0.289	Scott et al. (2010)
	Upper Mississippi River	528l.9	< 2	< 2 – 18,200	Newsted et al. (2017)
	Lake of the Isles	2.47	2.47	2.47	

State	Waterbody[1]	Arithmetic Mean PFOS Concentration (ng/L)[2]	Median PFOS Concentration (ng/L)[2]	Range of PFOS Concentration (ng/L)	Reference
Minnesota	Lake Calhoun	50.4	50.4	50.4	Simcik and Dorweiler (2005)
	Lake Harriet	22.1	22.1	22.1	
	Minnesota River	9.21	9.21	9.21	
	Lake Tettegouche	0.23	0.23	0.23	
	Lake Nipisiquit	< 0.27	< 0.27	< 0.27	
	Lake Loiten	< 0.27	< 0.27	< 0.27	
	Little Trout Lake	1.2	1.2	1.2	
	Echo Lake Reservoir	< 2	< 2	< 2	
New Jersey	Passaic River	13.1	13.1	13.0 – 13.2	NJDEP (2019)
	Raritan River	6.9	6.9	6.9	Zhang et al. (2016)
	Metedeconk River	1.65	1.65	< 2 – 2.8	
	Pine Lake	102	102	102	
	Horicon Lake	10	10	10	
	Little Pine Lake	100	100	100	
	Mirror Lake	72.9	72.9	72.9	
	Woodbury Creek	6.4	6.4	6.4	
	Fenwick Creek	3.1	3.1	3.1	
	Cohansey River	< 2	< 2	< 2	
	Harbortown Road	1.93	1.93	1.93	
	Passaic River	4.59	4.07	0.244 – 9.99	
New Mexico	Alamogordo Domestic Water Sys.	< 1	< 1	< 1	New Mexico Environment Department 2020–2021
	Animas River	0.799	0.625	< 0.89 – 1.5	
	Canadian River	0.848	0.9	< 0.89 – 1.2	
	Cloud Country Estates WUA	< 0.93	< 0.93	< 0.93	
	Gila River	< 0.93	< 0.93	< 0.93	
	Holloman AFB Golf Course Pond 1	1,220	1,220	1,220	
	Holloman AFB Golf Course Pond 2	878	878	878	
	Holloman AFB Lagoon G	310	310	310	
	Holloman AFB Outfall	951	951	951	
	Holloman AFB Sewage Lagoon	2,200	2,200	2,200	
	Karr Canyon Estates	< 0.93	< 0.93	< 0.93	
	La Luz MDWCA	< 1.3	< 1.3	< 1.3	
	Lake Holloman	4,033	4,500	1,700 – 5,900	
	Mountain Orchard MDWCA	< 0.93	< 0.93	< 0.93	
	Pecos River	1.223	1.50	<0.94 – 1.70	

State	Waterbody[1]	Arithmetic Mean PFOS Concentration (ng/L)[2]	Median PFOS Concentration (ng/L)[2]	Range of PFOS Concentration (ng/L)	Reference
New York	Rio Chama	< 0.98	< 0.98	< 0.96 – < 1	Kim and Kannan (2007)
	Rio Grande	1.052	0.474	< 0.465 – 2.90	
	Rio Puerco	4.35	4.35	3.10 – 5.60	
	San Juan River	< 1.15	< 1.15	< 1.06 – < 1.24	
	Tularosa Water System	0.723	0.723	< 0.89 – 1.0	
	Washington Park Lake	1.67	1.77	< 0.25 – 2.88	
	Rensselaer Lake	7.11	6.58	5.85 – 9.3	
	Iroquois Lake	not provided	not provided	not provided	
	Unnamed lake 1 outside Albany, NY	not provided	not provided	not provided	
	Unnamed lake 2 outside Albany, NY	not provided	not provided	not provided	
	Niagara River	5.17	5.5	3.3 – 6.7	Sinclair et al. (2006)
	Finger Lakes	not provided	1.6	1.3 – 2.6	
	Lake Onondaga	681	756	198 – 1,090	
	Lake Oneida	3.5	3.5	3.5	
	Erie Canal	8.37	6.4	5.7 – 13	
	Hudson River	not provided	1.7	1.5 – 3.4	
	Lake Champlain	not provided	2.7	0.8 – 7.7	
	Lower NY Harbor	0.755	0.755	0.755	Zhang et al.(2016)
	Staten Island	1.66	1.66	1.66	
	Hudson River	1.81	1.81	0.79 – 2.84	
North Carolina	Cape Fear River	31.2	28.9	< 1 – 132	Nakayama et al. (2007)
Rhode Island	Narragansett Bay	2.2	2.2	2.2	Benskin et al. (2012)
	Allen Cove Inflow	1.20	1.20	1.20	Zhang et al.(2016)
	Bristol Harbor	0.508	0.46	0.437 – 0.626	
	Brook at Mill Cove	9.80	9.80	9.80	
	Buckeye Brook	4.13	4.13	4.13	
	Chickasheen Brook	< 0.05	< 0.05	< 0.05	
	EG Town Dock	0.735	0.735	0.735	
	Fall River	0.238	0.238	0.238	
	Green Falls River	0.291	0.291	0.29 – 0.292	
	Hunt River	1.48	1.48	1.48	
	Mill Brook	3.94	3.94	3.94	
	Narrow River	0.298	0.264	0.176 – 0.488	
	Pawcatuck River	0.561	0.561	0.509 – 0.612	
	Pawtuxet River	2.19	2.19	2.19	
	Queens River	0.334	0.334	0.334	

State	Waterbody[1]	Arithmetic Mean PFOS Concentration (ng/L)[2]	Median PFOS Concentration (ng/L)[2]	Range of PFOS Concentration (ng/L)	Reference
	Sand Hill Brook	1.82	1.82	1.82	
	Secret Lake - Oak Hill Brook	< 0.05	< 0.05	< 0.05	
	Slack's Tributary	0.777	0.777	0.777	
	South Ferry Road Pier	0.161	0.161	0.161	
	Southern Creek	3.74	3.74	3.74	
	Woonasquat ucket River	14.6	14.6	5.87 – 23.2	
South Carolina	Charleston Harbor	12.0	not provided	not provided	Houde et al. 2006
Tennessee	Waterbody near Cleveland	2.5 < x < 25	2.5 < x < 25	< 2.5 – < 25	3MCompan y (2001)
	Conasauga River	<0.009[4]	<0.009[4]	<0.009[4]	Laiser et al.2011
Texas	Rio Grande	4.17	4.1	2.0 – 6.5	New Mexico Environment Department 2020
	Puget Sound	2.3	1.45	0.2 – 5.9	
Washington	Clayoquot Sound	0.32	0.3	0.25 – 0.4	Dinglasan- Panlilio et al. (2014)
	Barkley Sound	0.7	0.7	0.7	
Multiple States (10 Air Force Bases across the continental U.S.)	Surface waters impacted by aqueous film forming foam use	not provided	2,170	8,970,000 (maximum)	Anderson et al. (2016)

Less than (<) values based on study specific LOD and LOQ values that the study authors reported, LOD = limit of detection and LOQ = limit of quantitation

[1]Name of Waterbody Sampled for PFOS. Name or description of waterbody above is consistent with that provided in cited reference.

[2]Calculation of arithmetic mean and median includes lower of ½ LOD or ½ LOQ, depending on information provided. See full occurrence table in Appendix N for waterbody-specific details.

[3]. Study authors conducted additional sampling of this waterbody but were unable to detect the initial high PFOS concentrations in any of the additional samples.

[4]. Reported as ng/g by the study authors.

Table 2.

Measured Perfluorooctane Sulfonate (PFOS) Concentrations in Reference, Exposed, and Unidentified Surface Waters Sites Across the United States (U.S.). Additional details, including site type based on classification provided in the individual paper, number of measurements, and limits of detection and quantification, provided in Supplemental Data, Table S2.

Site Classification	Mean PFOS Concentration (ng/L)	Median PFOS Concentration (ng/L)	Range of PFOS Concentration (ng/L)
Exposed	1,746	76	< LOD – 8,970,000
Reference	776.94 (7.43)[1]	1	< LOD – 51,100 (< LOD – 138)[2]
Unidentified	5.91	1.28	< LOD – 121

LOD = limit of detection

[1]Mean including concentrations for pond in Port St. Lucie, Florida, which was classified as a reference site by the study authors. The mean concentration excluding these concentrations (total of 13 individual observations excluded) presented in parentheses.

[2]Range including concentrations for pond in Port St. Lucie, Florida, which as noted under footnote 1 above was classified as a reference site by the study authors. The range excluding these concentrations (total of 13 individual observations excluded presented in parentheses.

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Review

Poly- and Perfluoroalkyl Substances in Municipal Wastewater Treatment Plants in the United States: Seasonal Patterns and Meta-Analysis of Long-Term Trends and Average Concentrations

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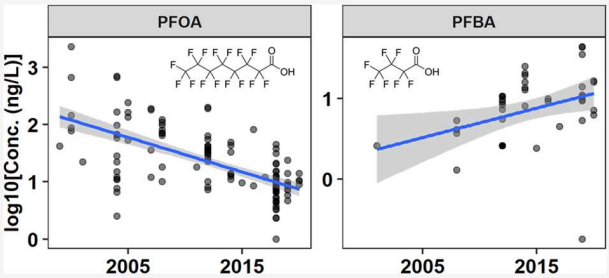
ABSTRACT: This paper presents an up-to-date meta-analysis assessing per- and polyfluoroalkyl substance (PFAS) concentrations at wastewater treatment plants (WWTPs) as well as changes over time. PFAS concentrations were compiled for WWTPs in the United States from peer-reviewed studies, technical reports, and original data. Perfluorooctanoic acid (PFOA) increased by an average of 6.0 ± 1.6 ng/L from the influents to the effluents of WWTPs, but perfluorosulfonic acid (PFOS) did not significantly change, indicating sorption to sludge is offset by biotransformation of precursor compounds. The occurrence of individual PFAS may vary temporally; for example, perfluoropentanoic acid correlated weakly with seasonal temperatures at a site in Virginia. Wastewater effluent PFOA concentrations decreased at a site in Nevada from 2012 to February 2020 but appeared to increase during the COVID-19 pandemic. Effluent PFOA also declined nationally from 1999 to 2020 by ∼13% per year. Nevertheless, the national mean PFOA concentration was 8.4 ± 0.4 ng/L in data collected from 2013 to 2020 with outliers omitted, indicating persisting low-level occurrence. This would equate to 383 ± 20 kg of PFOA per year continuing to enter the environment via WWTP effluents.



1. INTRODUCTION

Poly- and perfluoroalkyl substances (PFAS) are ubiquitous in municipal wastewater and biosolids. Major point sources include PFAS-producing or -using industrial sites, such as papermaking, textile mills, and electroplating.[1−4] However, PFAS have been detected in wastewater even without direct industrial sources, such as in septic tanks and office buildings.[5,6] Similarly, PFAS have been detected in the biosolids of small municipal wastewater treatment plants (WWTPs) without known direct industrial sources.[7] PFAS detected in wastewater and biosolids include not only the two most studied PFAS, perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), but also short-chain PFAS and polyfluorinated compounds.[5,8−10] It is suspected that PFAS in non-industrial wastewater may occur in part due to environmental degradation of polyfluorinated microfibers released by water-resistant clothing during laundry.[11] Another plausible non-industrial source of PFAS in municipal wastewater is human excretion after oral exposure.[12,13] Often, a portion of the PFAS in wastewater effluent can be ascribed to PFAS in the community's tap water.[14,15]

Certain PFAS have been under increasing regulatory scrutiny due to their persistence, bioaccumulation, and toxicity. PFOA has probable links with several toxicological end points, including kidney cancer, testicular cancer, and high cholesterol.[16] PFOS has been linked with reproductive toxicity, immune effects, and kidney toxicity in laboratory animals.[17,18] Biomarker studies have indicated similar effects in humans.[18] PFOA and PFOS have been phased out in the United States under the 2010/2015 PFOA Environmental Protection Agency (EPA) Stewardship Program and replaced with short-chain PFAS, polyfluorinated compounds, and perfluoroethers.[19,20] Shorter-chain alternatives have been utilized due to their shorter half-lives in the human body [e.g., 2.9 years for PFOS but 0.12 year for perfluorobutanesulfonic acid (PFBS)].[21] Less is known about the toxicity of short-chain PFAS, but precaution is warranted on the basis of the similarity of their persistence to that of PFOA and PFOS.[19] PFAS in wastewater effluent may also pose a hazard to aquatic ecosystems. The Australian PFAS National Environmental Management Plan recommends a guideline of 0.23 ng/L PFOS for 99% species protection in aquatic ecosystems.[1022]

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PFAS are poorly removed by WWTPs and conventional drinking water treatment plants.[8,23] In many cases, the concentrations of perfluoroalkyl acids (PFAAs) increase due to the biotransformation or oxidation of polyfluoroalkyl precursor compounds.[23,24] Without appropriate mitigation methods, human exposure could occur from wastewater through de facto and planned potable reuse, or through bioaccumulation into food or leaching into groundwater from biosolid application sites.[2,25−28]

Many studies have quantified PFAS in wastewater effluent[8,23,29,30] or biosolids.[7,31,32] Venkatesan and Halden[9] measured PFAS in five composites of 110 biosolid samples collected from 94 WWTPs in the United States in 2001. However, because of the way the samples were composited, distributions could not be analyzed and sources could not be identified. Furthermore, the types of PFAS observed in biosolids have changed since 2001 due to shifts in manufacturing toward short-chain PFAS.

Armstrong et al.[31] found no significant decline in PFAAs in biosolids at a WWTP in the mid-Atlantic region of the United States from 2005 to 2013.[33] However, because PFOS and PFOA were gradually phased out by different producers from 2002 to 2015, these two studies may have occurred too soon to observe any resulting changes in PFAS in biosolids.[34,35] Houtz et al.[36] found two short-chain PFAS, perfluorobutanoic acid (PFBA) and perfluorohexanoic acid (PFHxA), increased in the effluent of WWTPs in the San Francisco Bay Area between 2009 and 2014, but the sample size was limited (n = 6).[37] Arvaniti and Stasinakis[38] thoroughly reviewed occurrence studies for PFAS in wastewater and biosolids in 2015, but new data have emerged in the six years since.[1,25,39−48]

A comprehensive estimate of national average PFAS concentrations in WWTPs would have several uses and benefits. For one, WWTPs could compare their influent PFAS concentrations to the average to determine whether they are representative of baseline residential wastewater concentrations, or likely coming from an industrial source. PFAS WWTP effluent occurrence data could be compared with de facto reuse data to estimate PFAS exposure via drinking water due to PFAS in effluent.[49] PFAS biosolid concentrations could be used as inputs to models to estimate human exposure through agricultural products from land application sites.[50,51] The historical rate at which PFAS concentrations have changed at WWTPs should be quantified, so older PFAS data can be appropriately omitted or adjusted when conducting literature reviews or meta-analyses. Whether PFAS concentrations in wastewater effluent vary seasonally should also be investigated. This would inform the sample size and sampling frequency in future PFAS monitoring campaigns.

The goal of this meta-analysis was to generate an updated, comprehensive estimate of PFAS in U.S. WWTP influent, effluent, and biosolids. For effluent, which had the most data and over the widest time range, long-term trends were also examined. Seasonal patterns were also investigated at an individual WWTP with a relatively large sample size. To explore the impact of industrial wastewater on the overall average, outliers or WWTPs with stated industrial sources were sometimes omitted.

2. METHODS

2.1. Long-Term Trends (a single WWTP). Wastewater effluent concentrations of perfluoropentanoic acid (PFPeA), PFHxA, perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluorohexanesulfonic acid (PFHxS), and PFOS were compiled from a WWTP in Nevada from 2012 to 2021 (n = 36 total, n = 22 from 2012 to February 2020). Details about the Nevada WWTP and its sample locations are provided in Text S1 and Figure S1. Some of the PFAS data evaluated for the Nevada site have been published previously,[1,52,53] while some have been collected for ongoing research projects using published methods.[54,55] Linear modeling was conducted in R with the lm() function. To investigate whether the PFOA phaseout was causing on overall trend toward shorter-chain PFAS in effluent, the average PFCA chain length was calculated as

$$L_{av} = \frac{5[PFPeA] + 6[PFHxA] + 7[PFHpA] + 8[PFOA] + 9[PFNA] + 10[PFDA]}{[PFPeA] + [PFHxA] + [PFHpA] + [PFOA] + [PFNA] + [PFDA]} \quad (1)$$

where square brackets denote concentrations in nanomolar.

2.2. Seasonal Patterns. Twenty-seven PFAS were monitored every 2 weeks from May 2019 to May 2020 in the secondary effluent of a WWTP in Virginia using published methods (n = 24).[48,55] This WWTP has a design flow of 110000 m^3/day (30 MGD) with a five-stage Bardenpho process, and its sewershed has a landfill as a known PFAS source.

Certain PFAS such as PFPeA, PFHxA, PFHpA, and PFOA biotransform from polyfluorinated precursors, such as n:2 fluorotelomer alcohols and 6:2 fluorotelomer sulfonic acid (FTSA), which have been observed in wastewater.[39,48,56,57] 6:2 FTSA is known to biotransform from polyfluorinated substances in aqueous film forming foam (AFFF).[38,55,58] The Virginia WWTP has a zero discharge policy toward AFFF, and discharges of AFFF to sewers in general would be unlikely. Nevertheless, it was suspected that 6:2 FTSA might biodegrade from related higher-molecular weight polyfluorinated substances in wastewater. PFBA can also biotransform from the same polyfluorinated precursors as PFPeA and PFHxA[57] but was not evaluated due to the low detection frequency at the WWTP (12.5%). The impact of temperature on polyfluorinated precursor biotransformation rates has not been studied in controlled wastewater microcosms to the best of our knowledge. However, it is well-known that higher temperatures generally facilitate biological activity in WWTPs. Higher summer temperatures have been shown to increase the removal of acesulfame[59] and pharmaceuticals.[60] Tavasoli et al. found higher total measured PFAS concentrations in July than in March.[46]

Thus, it was hypothesized that these five PFAS would have a seasonal pattern caused by variables such as temperature accelerating biotransformation during certain times of year. Single-factor analysis of variance (ANOVA) was conducted with data points categorized into four seasons (winter, spring, summer, and fall). The normality of the PFAS concentrations at the Virginia WWTP was tested using normal probability plots, histograms, skewness, and kurtosis. Single and multiple

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Table 1. Slopes of PFAAs and Average PFCA Chain Lengths over Time (2012 to February 2020) in Wastewater Effluent at a WWTP in Nevada

parameter	n	predicted concentration in 2012 (ng/L)	slope (ng L^{-1} $year^{-1}$)	SE[a] (ng L^{-1} $year^{-1}$)	R^2	p value
PFPeA	22	17	0.15	0.44	0.006	0.73
PFHxA	22	8.8	0.19	0.22	0.034	0.41
PFHpA	22	2.2	−0.10	0.033	0.30	0.0084
PFOA	22	9.4	−0.73	0.079	0.81	1.2×10^{-8}
PFNA	22	2.7	−0.30	0.041	0.73	4.6×10^{-7}
PFDA	22	1.8	−0.14	0.022	0.65	5.4×10^{-6}
PFHxS	22	1.5	0.033	0.13	0.0033	0.80
PFOS	22	5.4	−0.52	0.37	0.088	0.18
average PFCA length[b]	22	6.2	−0.064	0.0094	0.70	1.4×10^{-6}

[a]SE is the standard error of the slope. [b]The units for average PFCA length are numbers of carbons.

linear correlations between PFAS concentrations and temperature, aerobic solid retention time, and percentage of flow from landfill leachate were investigated in R with the lm() function.

2.3. National Estimates. PFAS wastewater influent, effluent, and biosolid data were compiled from peer-reviewed articles searched using Web of Science. Statistical analyses were limited to studies sampling WWTPs in the United States to control for different PFAS practices among countries and so the data could be used for national PFAS emission estimates. Analyses were also limited to studies that reported data for individual WWTPs. Otherwise, (1) WWTPs within studies that reported averages only would be underweighted and (2) the distribution of PFAS concentrations would appear to be narrower than the true distribution. Three studies for wastewater influent or effluent and one study for biosolids were omitted on the basis of this criterion.[5,61−63] If multiple stages of wastewater treatment were sampled, only the final, most treated sample (e.g., disinfected tertiary effluent but not the preceding secondary effluent) was included to avoid double-counting WWTPs.

Sludge, digested sludge, and biosolids were analyzed as a group due to the relatively small sample size and because studies indicate that current full-scale solid handling practices have little impact on PFAS concentrations except for dilution or incineration.[32,64] The protocols if multiple solid samples were collected from different locations in the same WWTP or if the same solid samples were analyzed in multiple studies are described in Text S2.[65−67]

On the basis of this scope, aqueous data were analyzed from 20 peer-reviewed articles,[7,8,25,30,39−41,43−48,68−74] one Water Research Foundation (WRF) technical report,[1] one 3M report,[75] and one state government report.[42] Original data were also included for six WWTPs managed by a single utility in Virginia. The data for biosolids were analyzed from 10 peer-reviewed articles and one 3M report. The combined data set is provided in the second Supporting Information file. Concentrations below the method detection limit (MDL) were assumed to be half of the MDL. Alternatively, concentrations were assumed to be half of the method reporting limit (MRL) or limit of quantitation (LOQ) if the MDL was not reported. The half-MDL approach is considered adequate for summary statistics with <70% censoring and group comparisons with <25% censoring.[76,77] Statistical analyses were conducted for each PFAS for which the sample size was ≥30 and the detection frequency was ≥30% under each set of assumptions. Results for PFOA are shown in the text, and results for other PFAS with sufficient sample size are shown in the Supporting Information. Total U.S. wastewater flow was assumed to be 4.6 $\times 10^{13}$ L/year.[78] The total mass of biosolids was assumed to be 6.51 $\times$ 10^9 kg/year, of which approximately 55% is land applied for agriculture.[79]

Juxtaposing the mean PFAS concentrations with and without major point sources would indicate the relative importance of such sources. This would in turn reveal the limit to which industrial source control could reduce PFAS in wastewater effluent. PFAS concentrations in WWTPs without major point sources were investigated through two approaches. First, WWTPs that were clearly and specifically stated in the cited studies to have industrial PFAS sources were omitted. Due to the limited information available about WWTPs' sewersheds in published studies, industrial PFAS sources were not strictly defined on the basis of industrial categories or confirmed PFAS usage. Rather, the cited references' criteria for defining and reporting industrial PFAS sources to the WWTPs were taken at face value. However, studies might not consistently and clearly report on point sources to the WWTPs, and point sources might be highly dilute or emit PFAS other than the analyte in question. Therefore, a second, data-driven approach was also explored for omitting WWTPs with major industrial point sources. Using Tukey's outlier test (values greater than the 75th percentile plus 1.5 times the interquartile range), the highest outlier was iteratively omitted and the percentiles were recalculated until no high outliers remained.[80−82]

The simplest approach for estimating the mean of concentrations in wastewater or biosolids would be to treat all of the literature data as a simple random sample (SRS). The mean and standard error of the sample would then be treated as a direct estimate of the likely range of the mean of the population (i.e., all U.S. WWTPs). However, this approach would ignore the possibility that certain subpopulations of WWTPs—most critically, those with or without major PFAS point sources—might be over- or underrepresented in the literature data.

Stratified sampling is a statistical method for correcting for differences between the sample and the population in terms of the relative frequency of key subpopulations. For stratified samples, the mean is calculated as

$$\bar{x}_{st} = \sum_{i=1}^{j} w_i \bar{x}_i \tag{2}$$

where $\bar{x}_{st}$ is the mean of the stratified sample, $\bar{x}_i$ is the mean within each stratum, w_i is the weight of each stratum, j is the number of strata, and w_i is the weight assigned to each stratum.

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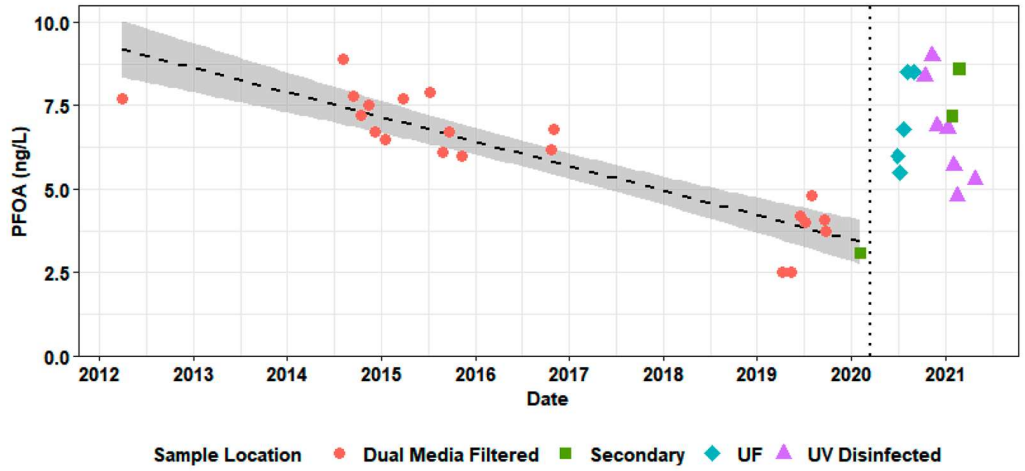

Figure 1. Concentrations of PFOA from 2012 to 2021 at the Nevada WWTP (n =36). The dashed line represents the line of best fit of the correlation with time from 2012 to February 2020 (n = 22). The shaded gray area is its 95% confidence band. The dotted vertical line indicates March 17th, 2020, the day closures of non-essential business went into effect in Nevada due to the COVID-19 pandemic. UF is ultrafiltration.

The standard error of the mean of stratified samples is calculated as

$$\mathrm{SE} = \left(\sum_{i=1}^{j} \frac{{w_i}^2 \times \mathrm{Var}_i}{n_i} \right)^{1/2} \quad (3)$$

where Var_i is the variance within each stratum.[83] However, a limitation of this approach is that it requires knowledge of the proportion of each subpopulation within both the sample and the population. Such information is not always consistently available when conducting meta-analyses.

3. RESULTS AND DISCUSSION

3.1. Long-Term Trends (a single WWTP). From 2012 to February 2020, PFOA concentrations in wastewater effluent at the Nevadan WWTP significantly declined at a rate of -0.73 ± 0.079 ng L^{-1} $year^{-1}$ (n = 22; $p = 7.8 \times 10^{-8}$) (Table 1 and Figure 1). PFHpA, PFNA, and PFDA significantly declined, as well (Table 1 and Figure S2). The average PFCA chain length decreased by -0.064 ± 0.0094 carbon/year (n = 22; $p = 1.4 \times 10^{-6}$) (Figure S3). PFOS would have been significantly decreasing if not for a high outlier in late 2014 (Figure S4).

However, PFOA concentrations at this WWTP increased during 2020 and then returned closer to 2019 levels by early 2021. This spike in PFOA concentrations coincided with the early stages of the societal response to the COVID-19 pandemic in the United States. The maximum PFOA concentration, 9 ng/L, was measured in November 2020, more than double the expected value of ~4 ng/L based on the 2012−2019 trendline. PFOS (Figure S2) and PFDA (Figure S3) also approximately doubled in 2020 before returning to values similar to those of 2019 by 2021. Flow at this WWTP decreased by 17% in March 2020 due to reduced flow from the hospitality industry,[85] which would not be expected to contribute substantially to PFAS loads. Therefore, the 2-fold increase in long-chain PFAS could be explained partially, but not fully, by reduced dilution from that sector. Changes in consumer product use (e.g., more usage of home cookware, more take-out or drive-through instead of dine-in restaurant experiences, and more washing of informal clothing) may also have contributed to the divergence from the previous trend. Effluent at this WWTP was sampled at different locations within its tertiary treatment after 2019, which could appear to confound this observation. However, the processes between these sample points are known to have a negligible effect on PFAS based on studies at this WWTP (Text S1).[1]

3.2. Seasonal Patterns (a single WWTP). Visually, PFPeA had the most plausible seasonal pattern among the PFAS evaluated, with the highest concentrations in the summer (Figure S5). PFPeA correlated with raw influent temperature with a significant p value of 7.97×10^{-3} but a low R^2 of 0.28 (Figure 2 and Table S1). A single-factor ANOVA

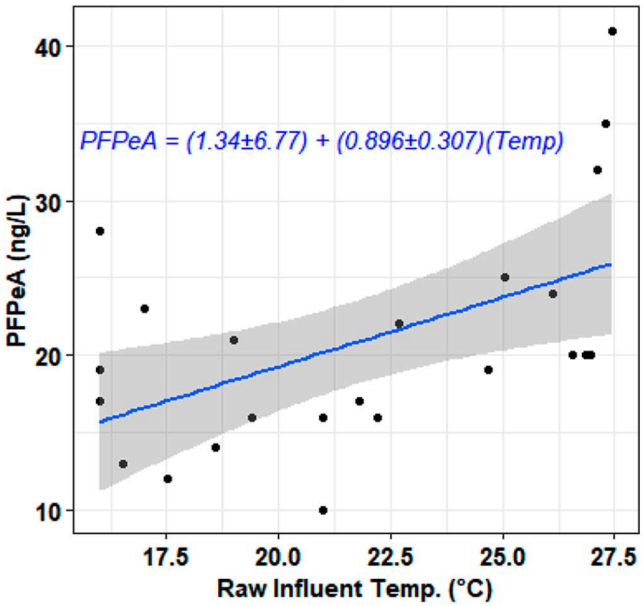

Figure 2. Correlation between effluent PFPeA concentration and raw influent temperature at the Virginia WWTP.

was conducted to test whether the mean PFPeA concentration likely differed among the seasons. However, the p value of this ANOVA was not quite significant (p = 0.0501). Histograms and normal probability plots indicated PFPeA concentrations were somewhat right-skewed (Figures S6 and S7). However, the kurtosis and skewness of PFPeA had absolute values of <2, indicating acceptable normality for ANOVA.[86] Further testing

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Table 2. Estimates of PFOA Concentrations and National Emissions in Wastewater Effluent[a]

method	n	mean concentration (ng/L)	mean concentration SE (ng/L)	estimated emissions (kg/year)	estimated emissions SE (kg/year)
SRS, all studies	129	68.9	19.7	3141	899
SRS, no stated industrial source	112	50.5	10.7	2303	486
SRS, no outliers	70	8.6	0.401	391	18.3
SRS, 2013−2020	68	12.8	1.58	585	72.0
SRS, 2013−2020, no outliers	58	8.41	0.444	383	20.2
stratified, 0.1% stated industrial source	129	50.7	10.7	2309	486
stratified, 20% stated industrial source	129	78.4	27.7	3575	1263

[a]SE is the standard error. SRS is a simple random sample. See Tables S2−S14 for similar tables of other PFAS. The sample year range is 1998−2020 unless otherwise noted.

at monthly or greater frequencies at other WWTPs is needed to conclude whether the temperature correlation observed here is generalizable, not site-specific or coincidental.

3.3. National Estimates. *3.3.1. Wastewater Effluent. 3.3.1.1. All Data.* Considering all literature data, the mean PFOA concentration was 69 ± 20 ng/L (Table 2). This concentration corresponds to total emissions of 3141 ± 899 kg/year, which is within reason compared to the estimated range of 5000−40000 kg/year of total PFOA emissions to water in the United States, Japan, and western Europe prior to 2003 according to Wang et al.[89] However, the PFOA data were not normally distributed, with an extreme right skew and an outlier 165 times higher than the median (Figure S8A). This non-normality indicates a confidence interval might not reflect the true likely range of the mean concentration. Furthermore, the outliers are presumably from WWTPs with major industrial PFAS sources.

3.3.1.2. Stated Industrial Point Sources Omitted. Omitting WWTPs with stated industrial sources decreased the mean PFOA concentration to 51 ± 11 ng/L. However, the PFOA concentration distribution remained right skewed with multiple outliers (Figure S8B). This was at least partly because this data subset still contained some WWTPs with major industrial sources. For example, EGLE[42] stated six of 42 sampled WWTPs had industrial PFAS sources but did not state which six.

3.3.1.3. Tukey Outliers Omitted. After Tukey outliers had been omitted, the mean PFOA concentration was only 8.6 ± 0.4 ng/L, 6 times lower than the mean relying on references to state industrial influence accurately. Thus, it is apparent that industrially impacted WWTPs are not consistently identified in the literature. After outliers had been omitted, the remaining PFOA data appeared to be normally distributed (Figure S8C). However, this was not the case for PFBS, or PFDA, possibly due to a greater proportion of the data for these analytes being below their MDLs (Figure S11).

3.3.1.4. Long-Term Trends and Recent Data. When all WWTPs were included, the PFOA concentration significantly correlated with sample year with a slope of −14 ± 3 ng L^{-1} $year^{-1}$ ($p = 1.2 \times 10^{-5}$) (Figure S14A). PFOS (−18 ± 6 ng L^{-1} $year^{-1}$; $p = 0.0060$) and FOSA (−0.75 ± 0.30 ng L^{-1} $year^{-1}$; $p = 0.016$) also showed significant negative trends (Figure S15). In contrast, PFBA, a short-chain PFAS, was significantly increasing at a rate of 0.98 ± 0.33 ng L^{-1} $year^{-1}$. However, the true long-term trends could be confounded if industrially impacted WWTPs were more likely to be sampled in older or more recent years. When WWTPs with stated industrial sources were omitted, only PFOA had a significant downward trend, declining at a rate of −8.7 ± 1.7 ng L^{-1} $year^{-1}$ ($p = 1.1 \times 10^{-6}$) (Figures S14C and S16). Data subsets with outliers omitted were not analyzed for long-term trends because a data point could plausibly be an outlier due to being relatively old rather than being industrially impacted.

Because the concentration of PFOA in U.S. wastewater effluent is decreasing over time, older studies may no longer be representative of current emissions. Considering the confidence band of the correlation between PFOA and sample year, 2013 was selected as the most distant past year in which mean PFOA concentrations would be expected to be like 2020 (Figure S14C). Therefore, "recent" studies were herein defined as those sampling wastewater effluent in 2013−2020. When the data set was restricted to only recent studies, the mean PFOA concentration was only 13 ± 2 ng/L (Table 2). PFOA and PFNA were significantly decreasing over this more limited time frame (Figure S17).

When Tukey outliers were omitted from the recent data, the mean PFOA concentration was 8.4 ± 0.4 ng/L, significantly lower than with outliers (Table 2). This indicates PFOA industrial sources were still important after 2013. The maximum PFOA concentration during 2013−2020 after omitting outliers was 15 ng/L. Therefore, PFOA concentrations above this level would be considered atypical for recent samples from WWTPs without major PFAS point sources.

Linear regression on log-transformed concentrations showed PFOA significantly declined at an annual rate of 13% (i.e., half-life of 5.0 years) when all data were analyzed and 12% (i.e., half-life of 5.3 years) when WWTPs with stated industrial sources were omitted (Figure S14B and Figure 3). Using the log-transformed correlation without stated industrial sources, the mean PFOA concentration would reach the Illinois drinking water health advisory level of 2 ng/L around 2030.[91]

3.3.1.5. Stratified Analyses for Industrially Impacted WWTPs. It would be useful to conduct a stratified analysis to correct for differences in the proportion of industrially impacted WWTPs between the sample and the true population. However, this would require knowledge of the percentage of WWTPs in the United States that are substantially impacted by PFAS-producing or -using industries. Unfortunately, this is difficult to estimate. EGLE[42] sampled PFAS at 42 WWTPs in Michigan, of which six (14%) were industrially impacted. However, that study took place in a relatively industrial state and intentionally focused on larger WWTPs that are simultaneously more likely to contain industrial point sources yet also have a high degree of dilution for any point sources. Hu et al.[2] identified 16 PFAS-producing

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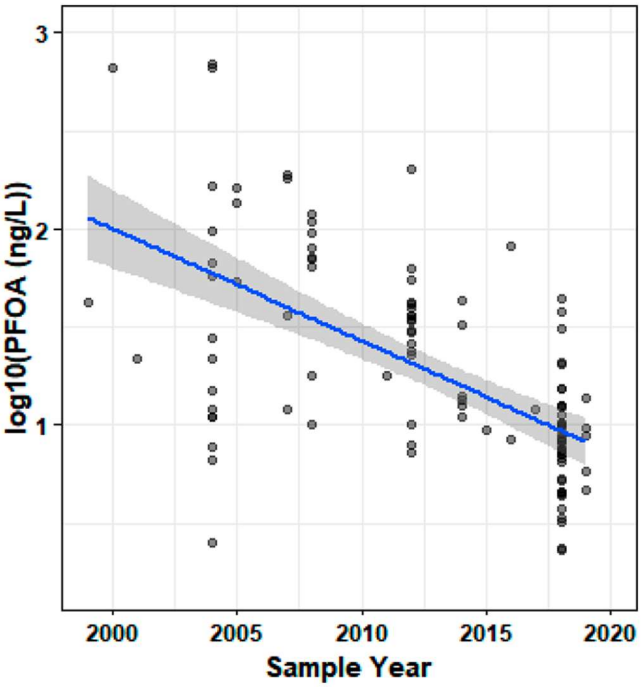

Figure 3. Correlations between log-transformed wastewater effluent PFOA concentration and sample year in the United States without stated industrial sources. The blue line represents the line of best fit, and the shaded gray area represents its 95% confidence band.

facilities in the United States, but it is unclear which, if any, of these facilities discharge PFAS to municipal sewers. Hu et al.[2] also identified 533 AFFF-certified airports, which may emit AFFF to the sewer intermittently if at all. The Environmental Working Group (EWG) identified 2444 facilities suspected of using PFAS based on their industrial categories.[92] However, this is likely an overestimate because, for example, not all textile-manufacturing facilities may produce water- or stain-resistant fabric. Furthermore, it is unclear how many of these industrial sites discharge to municipal sewers or directly to the aquatic environment, or how many clusters of sites discharge to the same few sewer systems. The U.S. EPA Chemical Data Reporting Rule lists 28 facilities known to produce or use large amounts of PFAS, but this may be an underestimate because it is based on voluntary reporting.[92]

There were at least 2600 active municipal landfills in the United States in 2020 according to the U.S. EPA.[93] With a few exceptions in arid regions, the leachate from these landfills is discharged to municipal sewers. The total PFOA emission from landfills in the United States in 2013 was estimated to be 52 kg/year.[94] This would be <2% compared to the wastewater effluent estimate using all data (3141 ± 899 kg/year) or <10% compared to the estimate for 2013−2020 (585 ± 72 kg/year) (Table 2). Estimated emissions of other PFAAs from landfills in the same study would also be ≤10% compared to estimated wastewater effluent emissions (Tables S2−S13). The measured PFAS with the highest landfill emission was the polyfluorinated compound 5:3 fluorotelomer carboxylic acid.[94] However, few data were available for this analyte in U.S. wastewater effluent. Thus, landfill leachate is likely a minor contributor to total, national wastewater PFAS emissions. Nonetheless, landfills could cause high PFAS concentrations in site-specific situations, particularly considering landfills may discharge to small WWTPs because they tend to be located outside city centers. An example of a small WWTP with PFAS concentrations meaningfully impacted by landfill leachate was documented by Tavasoli et al.[46]

On the basis of the considerations mentioned above, a sensitivity analysis was conducted assuming the true proportion of the 16000 WWTPs in the United States with substantial industrial impact was as low as 0.1% or as high as 20%. The literature data were treated as a stratified sample with two strata: stated industrial PFAS source(s) or no stated industrial PFAS source. WWTPs with industrial PFAS sources were defined as those clearly stated to be so in the references (i.e., the same assumption as the second row of Table 2). On the basis of these assumptions and weights, the mean PFOA concentration in wastewater effluent would be as low as 51 ± 11 ng/L or as high as 78 ± 28 ng/L (Table 2). These end cases are not put forth as best estimates of the true, current mean PFOA concentration in U.S. wastewater effluent, considering they incorporate data collected before 2013 and the true proportion of industrially impacted WWTPs is likely somewhere in the middle. Rather, these extreme possibilities demonstrated the necessity of better data on the use and disposal of PFAS by industries, such as including some PFAS in the federal Toxic Release Inventory.

3.3.1.6. Relative Concentrations. Which PFAA had the highest mean concentration depended on the set of assumptions. PFPeA was the highest including all data at 87 ± 51 ng/L (Table S3). PFOA was the highest omitting known industrial sources at 51 ± 11 ng/L (Table 2). PFHxA was the highest when omitting suspected industrial outliers at 23 ± 2 ng/L (Table S4). PFHxA was also highest when limiting to 2013−2020 data at 48 ± 11 ng/L.

Knowing the typical relative proportions of different PFAS analytes in effluent would have multiple benefits. For one, effluents that significantly deviate from these proportions could be identified as likely industrially impacted. For another, surface waters that match these proportions with equal or lower total concentrations could be identified as likely impacted by de facto reuse. To this end, data were filtered to only WWTPs at which all of the following common PFAAs were analyzed: PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, and PFOS (n = 23). Proportions were calculated on a molar basis and are shown in Figure 4. PFPeA, PFHxA, and PFOA each accounted for ∼25% of the commonly measured PFAAs in wastewater effluent without stated industrial sources.

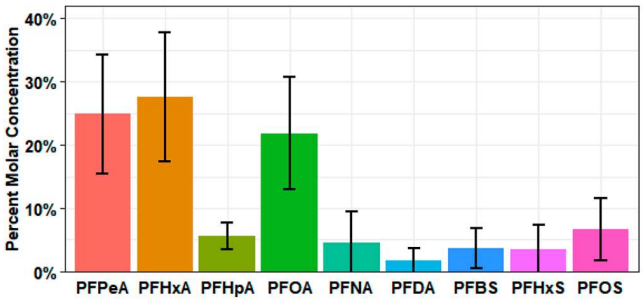

Figure 4. Mean molar proportions of PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, and PFOS in wastewater effluents without stated industrial sources (n = 23). Error bars are standard deviations.

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3.3.1.7. Downstream Surface Water Implications. For context, PFOA and PFOS concentrations were divided by their Michigan surface water standards, or water quality values (WQVs) (Table S14). Michigan has set lower WQVs for water bodies used for drinking water and higher WQVs for water bodies not used for drinking water. For PFOA, the drinking water and nondrinking water WQVs are 420 and 12000 ng/L, respectively. For PFOS, the drinking water and nondrinking water WQVs are 11 and 12 ng/L, respectively. Under none of the sets of assumptions explored in this study would the mean effluent PFOA concentration exceed its drinking water WQV (Figure 5 and Figure S18). In contrast, mean effluent PFOS would exceed its WQVs by a factor of ≥2 considering all data, data without stated industrial sources, and data for 2013−2020 (Figure 5 and Figure S18). Without Tukey outliers, mean PFOS is 10.0 ± 0.7 ng/L, indicating some non-industrially impacted WWTPs could have effluent PFOS above these WQVs. However, de facto reuse of >50% is rare in the United States.[49] Therefore, considering dilution, it is unlikely municipal wastewater effluent would cause an exceedance of these WQVs without an industrial source.

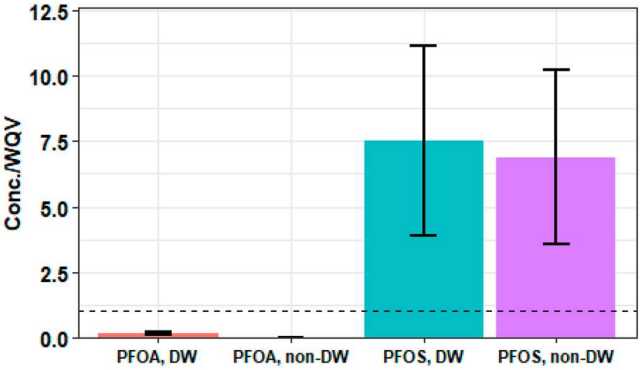

Figure 5. Mean concentration divided by Michigan water quality values (WQVs) for drinking water (DW) and nondrinking water (non-DW) water bodies considering all literature data. Error bars are standard errors of the mean. The dashed horizontal line represents 1 or a concentration equal to the WQV.

3.3.2. Wastewater Influent. A smaller sample size was available in the literature for PFAS in U.S. wastewater influent, with only PFOA and PFOS having $n \geq 30$ (Table S16). Furthermore, a majority of these PFOA and PFOS data were from a single study in Michigan, so it may not be nationally representative.[42] When all data were included, the mean PFOS influent concentration was 35 ± 14 ng/L, which corresponded to a loading of 1596 ± 638 kg/year (n = 67). When all data were included, the mean PFOA influent concentration was 11 ± 2 ng/L, which corresponded to a loading of 495 ± 101 kg/year (n = 62). This mean influent PFOA concentration was ∼7 times lower than the effluent concentration when all data were included.

The lower mean PFAS in influent may have been partly due to the influent data set having fewer industrially impacted WWTPs and a more recent average sampling year, but it could also be partly from PFOA precursor transformation. The increase in average PFOA in WWTPs with both influent and effluent data was 6.0 ± 1.6 ng/L (n = 62). The increases in PFOA were not normally distributed (Figure S19A), but both a t test (p = 0.00019) and a Wilcoxon signed rank test ($p = 1.5 \times 10^{-7}$) indicated the mean difference was greater than zero. The true PFOA formation could be somewhat greater considering ∼14% of influent PFOA is expected to sorb to sludge.[98] In WWTPs with both influent and effluent data available, PFOS did not significantly increase or decrease from influent to effluent (t test p = 0.73, and Wilcoxon p = 0.94). Considering ∼50% of influent PFOS would be expected to sorb to sludge,[98] it could be PFOS formation and sorption roughly offset each other. Alternatively, the true trend in PFOS concentrations changes through WWTPs may have been obscured by extreme outliers in both directions (Figure S19B).

3.3.3. Biosolids and Sludge. Within the scope of this meta-analysis, only PFOS (n = 36) and PFOA (n = 36) had $n \geq 30$ in sludge and biosolids. Due to this smaller sample size, data for solids were not subsetted by industrial impact or sample year like was done for WWTP effluent. However, data were stratified assuming 0.1% or 20% of the known industrial sources, as was done with influent and effluent. Assuming 20% of WWTPs nationwide were industrially impacted increased the estimated mean concentration in influent and effluent. However, for solids, this assumption resulted in a similar estimated mean concentration as treating the data as a simple random sample (Table 3). This was because samples of industrially impacted solids were overrepresented in the database at 22%.

3.3.4. Limitations. Differences in analytical methods among the cited studies may have contributed to uncertainty in the estimates of long-term trends and national averages. For example, most studies employed solid phase extraction[8,96] but some used direct injection.[30] Isotope dilution with internal mass-labeled standards is an effective method for correcting for extraction efficiency. One study used external calibration only;[96] some studies used mass-labeled internal standards for a subset of PFAS analyzed,[69] while others used mass-labeled standards for all PFAS.[8] Overall method recoveries varied among studies.[8,96] Generally, sources of imprecision like these would tend to obscure a true correlation rather cause a false positive.[97]

Studies differed in terms of whether they censored data based on MDL,[71] MRL,[8] or LOQ.[69] MRLs and LOQs also differed within and among studies.[8,44,69] Less than 1% of WWTP effluent samples included in this meta-analysis had PFOA below the censored level, so nondetections and different

Table 3. Estimates of PFOA Concentrations and National Emissions in Biosolids and Sludge[a]

method	n	mean concentration (ng/g)	mean concentration SE (ng/g)	estimated load (kg/year)	estimated load SE (kg/year)
SRS, all studies	36	23.8	6.84	155	44.5
stratified, 0.1% stated industrial source	36	15.3	4.30	99.7	28.0
stratified, 20% stated industrial source	36	22.9	6.12	149	39.8

[a]SE is the standard error. SRS is the simple random sample. See Table S17 for a similar table of PFOS.

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censored levels would have little impact on the PFOA statistics discussed in the text. However, nondetections may have increased the uncertainty for other PFAS featured in the Supporting Information; e.g., 25% of PFNA data points were below their censored level.

Insufficient data were available for emerging PFAS [e.g., perfluoroethers and polyfluorinated substances except for *N*-ethyl perfluorooctane sulfonamido acetic acid (EtFOSAA) or 6:2 FTSA] to make statistically sound inferences about long-term trends, seasonal patterns, or national average concentrations in the United States. Furthermore, none of the cited studies stated whether the sampled WWTPs had combined sewers or sanitary sewers only. Therefore, the impact of intentional urban runoff mixing on PFAS concentrations could not be investigated with the literature data.[84,87,88,90,95]

4. CONCLUSIONS

Concentrations of long-chain PFAS, particularly PFOA, declined over time in wastewater effluent. Concentrations of short-chain PFAAs, such as PFBA, may be increasing. The long-term trend for PFOA was observed both nationally and at a specific WWTP in Nevada with a relatively high sample size from 2012 to February 2020. However, the COVID-19 pandemic may have caused a divergence from the prior long-term trend at the Nevadan WWTP. Seasonal analysis of PFPeA at a WWTP in Virginia showed a weak correlation with temperature. Mean PFAS concentrations in the literature are dominated by industrially impacted outliers. Historically, industrial discharges to sewers likely caused the majority of the PFAS emissions from WWTPs; more recently, it appears PFAS from domestic wastewater accounts for most of the load. A precise, confident estimate of the total WWTP PFAS emissions, including industrial contributions, would require more information about the location, number, and emissions of PFAS users. PFOA and PFOS may be formed from transformation of precursors and partition to solids within WWTPs. For PFOS, these processes roughly offset each other, while PFOA had a mean net increase of 6.0 ± 1.6 ng/L.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestwater.1c00377.

Additional meta-analysis methodological details as well as tables and figures particularly for PFAS other than PFOA (PDF)

Data for cited and original wastewater influent, effluent, and biosolids used for this meta-analysis (XLS)

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Notes

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■ REFERENCES

(1) Dickenson, E. R. V.; Pisarenko, A. N.; Marti, E. J.; Gerrity, D.; Vanderford, B. J. Formation of Nitrosamines and Perfluoroalkyl Acids During Ozonation in Water Reuse Applications. Project 11-08; WateReuse Research Foundation: Alexandria, VA, 2015.

(2) Hu, X. C.; Andrews, D. Q.; Lindstrom, A. B.; Bruton, T. A.; Schaider, L. A.; Grandjean, P.; Lohmann, R.; Carignan, C. C.; Blum, A.; Balan, S. A.; Higgins, C. P.; Sunderland, E. M. Detection of Poly- and Perfluoroalkyl Substances (PFASs) in U.S. Drinking Water Linked to Industrial Sites, Military Fire Training Areas, and Wastewater Treatment Plants. *Environmental Science and Technology Letters* **2016**, *3* (10), 344−350.

(3) Clara, M.; Scheffknecht, C.; Scharf, S.; Weiss, S.; Gans, O. Emissions of Perfluorinated Alkylated Substances (PFAS) from Point Sources - Identification of Relevant Branches. *Water Sci. Technol.* **2008**, *58* (1), 59−66.

(4) Ebersbach, I.; Ludwig, S. M.; Constapel, M.; Kling, H. W. An Alternative Treatment Method for Fluorosurfactant-Containing Wastewater by Aerosol-Mediated Separation. *Water Res.* **2016**, *101*, 333−340.

(5) Subedi, B.; Codru, N.; Dziewulski, D. M.; Wilson, L. R.; Xue, J.; Yun, S.; Braun-Howland, E.; Minihane, C.; Kannan, K. A Pilot Study on the Assessment of Trace Organic Contaminants Including Pharmaceuticals and Personal Care Products from On-Site Wastewater Treatment Systems along Skaneateles Lake in New York State, USA. *Water Res.* **2015**, *72*, 28−39.

(6) Kothari, M.; Triolo, S.; Weeks, B.; Salveson, A. *Building Scale Potable Water Reuse Demonstration Project*; Water Research Foundation: Denver, 2020.

(7) Sinclair, E.; Kannan, K. Mass Loading and Fate of Perfluoroalkyl Surfactants in Wastewater Treatment Plants. *Environ. Sci. Technol.* **2006**, *40* (5), 1408−1414.

(8) Appleman, T. D.; Higgins, C. P.; Quiñones, O.; Vanderford, B. J.; Kolstad, C.; Zeigler-Holady, J. C.; Dickenson, E. R. V. Treatment of Poly- and Perfluoroalkyl Substances in U.S. Full-Scale Water Treatment Systems. *Water Res.* **2014**, *51*, 246−255.
(9) Venkatesan, A. K.; Halden, R. U. National Inventory of Perfluoroalkyl Substances in Archived U.S. Biosolids from the 2001 EPA National Sewage Sludge Survey. *Journal of Hazardous Materials* **2013**, *252*−*253*, 413−418.
(10) Coggan, T. L.; Moodie, D.; Kolobaric, A.; Szabo, D.; Shimeta, J.; Crosbie, N. D.; Lee, E.; Fernandes, M.; Clarke, B. O. An Investigation into Per- and Polyfluoroalkyl Substances (PFAS) in Nineteen Australian Wastewater Treatment Plants (WWTPs). *Heliyon* **2019**, *5* (8), e02316.
(11) Schellenberger, S.; Jönsson, C.; Mellin, P.; Levenstam, O. A.; Liagkouridis, I.; Ribbenstedt, A.; Hanning, A. C.; Schultes, L.; Plassmann, M. M.; Persson, C.; Cousins, I. T.; Benskin, J. P. Release of Side-Chain Fluorinated Polymer-Containing Microplastic Fibers from Functional Textiles during Washing and First Estimates of Perfluoroalkyl Acid Emissions. *Environ. Sci. Technol.* **2019**, *53* (24), 14329−14338.
(12) Worley, R. R.; McAfee Moore, S.; Tierney, B. C.; Ye, X.; Calafat, A. M.; Campbell, S.; Woudneh, M. B.; Fisher, J. Per- and Polyfluoroalkyl Substances in Human Serum and Urine Samples from a Residentially Exposed Community. *Environ. Int.* **2017**, *106*, 135−143.
(13) Ma, J.; Zhu, H.; Kannan, K. Fecal Excretion of Perfluoroalkyl and Polyfluoroalkyl Substances in Pets from New York State, United States. *Environmental Science and Technology Letters* **2020**, *7* (3), 135−142.
(14) Andrews, D. Q.; Naidenko, O. V. Population-Wide Exposure to Per- and Polyfluoroalkyl Substances from Drinking Water in the United States. *Environmental Science & Technology Letters* **2020**, *7* (12), 931−936.
(15) Filipovic, M.; Berger, U. Are Perfluoroalkyl Acids in Waste Water Treatment Plant Effluents the Result of Primary Emissions from the Technosphere or of Environmental Recirculation? *Chemosphere* **2015**, *129*, 74−80.
(16) Betts, K. S. PFOA and High Cholesterol: Basis for the Finding of a Probable Link. *Environ. Health Perspect.* **2014**, *122* (12), 2014.
(17) *Drinking Water Health Advisory for Perfluorooctane Sulfonate (PFOS)*; U.S. Environmental Protection Agency: Washington, DC, 2016.
(18) Zeng, Z.; Song, B.; Xiao, R.; Zeng, G.; Gong, J.; Chen, M.; Xu, P.; Zhang, P.; Shen, M.; Yi, H. Assessing the Human Health Risks of Perfluorooctane Sulfonate by in Vivo and in Vitro Studies. *Environ. Int.* **2019**, *126*, 598−610.
(19) Brendel, S.; Fetter, É.; Staude, C.; Vierke, L.; Biegel-Engler, A. Short-Chain Perfluoroalkyl Acids: Environmental Concerns and a Regulatory Strategy under REACH. *Environ. Sci. Eur.* **2018**, *30* (1), 9 DOI: 10.1186/s12302-018-0134-4.
(20) Hopkins, Z. R.; Sun, M.; DeWitt, J. C.; Knappe, D. R. U. Recently Detected Drinking Water Contaminants: GenX and Other Per- and Polyfluoroalkyl Ether Acids. *Journal - American Water Works Association* **2018**, *110* (7), 13−28.
(21) Xu, Y.; Fletcher, T.; Pineda, D.; Lindh, C. H.; Nilsson, C.; Glynn, A.; Vogs, C.; Norström, K.; Lilja, K.; Jakobsson, K.; Li, Y. Serum Half-Lives for Short- and Long-Chain Perfluoroalkyl Acids after Ceasing Exposure from Drinking Water Contaminated by Firefighting Foam. *Environ. Health Perspect.* **2020**, *128* (July), 077004.
(22) Phillips, M. M.; Dinglasan-Panlilio, M. J. A.; Mabury, S. A.; Solomon, K. R.; Sibley, P. K. Fluorotelomer Acids Are More Toxic than Perfluorinated Acids. *Environ. Sci. Technol.* **2007**, *41* (20), 7159−7163.
(23) Arvaniti, O. S.; Ventouri, E. I.; Stasinakis, A. S.; Thomaidis, N. S. Occurrence of Different Classes of Perfluorinated Compounds in Greek Wastewater Treatment Plants and Determination of Their Solid-Water Distribution Coefficients. *Journal of Hazardous Materials* **2012**, *239*−*240*, 24−31.
(24) Pisarenko, A. N.; Marti, E. J.; Gerrity, D.; Peller, J. R.; Dickenson, E. R. V. Effects of Molecular Ozone and Hydroxyl Radical on Formation of N-Nitrosamines and Perfluoroalkyl Acids during Ozonation of Treated Wastewaters. *Environmental Science: Water Research and Technology* **2015**, *1* (5), 668−678.
(25) Glover, C. M.; Quiñones, O.; Dickenson, E. R. V. Removal of Perfluoroalkyl and Polyfluoroalkyl Substances in Potable Reuse Systems. *Water Res.* **2018**, *144*, 454−461.
(26) Lindstrom, A. B.; Strynar, M. J.; Delinsky, A. D.; Nakayama, S. F.; McMillan, L.; Libelo, E. L.; Neill, M.; Thomas, L. Application of WWTP Biosolids and Resulting Perfluorinated Compound Contamination of Surface and Well Water in Decatur, Alabama, USA. *Environ. Sci. Technol.* **2011**, *45* (19), 8015−8021.
(27) Ahrens, L.; Shoeib, M.; Harner, T.; Lee, S. C.; Guo, R.; Reiner, E. J. Wastewater Treatment Plant and Landfills as Sources of Polyfluoroalkyl Compounds to the Atmosphere. *Environ. Sci. Technol.* **2011**, *45* (19), 8098−8105.
(28) Erickson, J. PFAS Wastewater Testing: Most Utilities Decline to Participate. https://www.nbc26.com/news/local-news/pfas-wastewater-testing-most-utilities-decline-to-participate (accessed 2021-10-08).
(29) Ahrens, L.; Felizeter, S.; Sturm, R.; Xie, Z.; Ebinghaus, R. Polyfluorinated Compounds in Waste Water Treatment Plant Effluents and Surface Waters along the River Elbe, Germany. *Mar. Pollut. Bull.* **2009**, *58* (9), 1326−1333.
(30) Schultz, M. M.; Higgins, C. P.; Huset, C. A.; Luthy, R. G.; Barofsky, D. F.; Field, J. A. Fluorochemical Mass Flows in a Municipal Wastewater Treatment Facility. *Environ. Sci. Technol.* **2006**, *40* (23), 7350−7357.
(31) Armstrong, D. L.; Lozano, N.; Rice, C. P.; Ramirez, M.; Torrents, A. Temporal Trends of Perfluoroalkyl Substances in Limed Biosolids from a Large Municipal Water Resource Recovery Facility. *Journal of Environmental Management* **2016**, *165*, 88−95.
(32) Loganathan, B. G.; Sajwan, K. S.; Sinclair, E.; Senthil Kumar, K.; Kannan, K. Perfluoroalkyl Sulfonates and Perfluorocarboxylates in Two Wastewater Treatment Facilities in Kentucky and Georgia. *Water Res.* **2007**, *41* (20), 4611−4620.
(33) Zennegg, M.; Munoz, M.; Schmid, P.; Gerecke, A. C. Temporal Trends of Persistent Organic Pollutants in Digested Sewage Sludge (1993−2012). *Environ. Int.* **2013**, *60*, 202−208.
(34) Ozekin, K.; Fulmer, A. *Per- and Polyfluoroalkyl Substances: Background and Technical Information*; The Water Research Foundation: Alexandria, VA, 2019.
(35) Ulrich, H.; Freier, K. P.; Gierig, M. Getting on with Persistent Pollutants: Decreasing Trends of Perfluoroalkyl Acids (PFAAs) in Sewage Sludge. *Chemosphere* **2016**, *161*, 527−535.
(36) Houtz, E. F.; Sutton, R.; Park, J. S.; Sedlak, M. Poly- and Perfluoroalkyl Substances in Wastewater: Significance of Unknown Precursors, Manufacturing Shifts, and Likely AFFF Impacts. *Water Res.* **2016**, *95*, 142−149.
(37) Oyetade, O. A.; Varadwaj, G. B. B.; Nyamori, V. O.; Jonnalagadda, S. B.; Martincigh, B. S. *A Critical Review of the Occurrence of Perfluoroalkyl Acids in Aqueous Environments and Their Removal by Adsorption onto Carbon Nanotubes*; Springer: Dordrecht, The Netherlands, 2018; Vol. *17*.
(38) Arvaniti, O. S.; Stasinakis, A. S. Review on the Occurrence, Fate and Removal of Perfluorinated Compounds during Wastewater Treatment. *Sci. Total Environ.* **2015**, *524*−*525*, 81−92.
(39) Dixon-Anderson, E.; Lohmann, R. Field-Testing Polyethylene Passive Samplers for the Detection of Neutral Polyfluorinated Alkyl Substances in Air and Water. *Environ. Toxicol. Chem.* **2018**, *37* (12), 3002−3010.
(40) Elmoznino, J.; Vlahos, P.; Whitney, M. Occurrence and Partitioning Behavior of Perfluoroalkyl Acids in Wastewater Effluent Discharging into the Long Island Sound. *Environ. Pollut.* **2018**, *243*, 453−461.
(41) Ateia, M.; Arifuzzaman, M.; Pellizzeri, S.; Attia, M. F.; Tharayil, N.; Anker, J. N.; Karanfil, T. Cationic Polymer for Selective Removal

of GenX and Short-Chain PFAS from Surface Waters and Wastewaters at Ng/L Levels. *Water Res.* **2019**, *163*, 114874.
(42) Summary Report: Initiatives to Evaluate the Presence of PFAS in Municipal Wastewater and Associated Residuals (Sludge/Biosolids) in Michigan. Michigan Department of Environment, Great Lakes, and Energy: Lansing, MI, 2020.
(43) Houtz, E. F.; Sutton, R.; Park, J. S.; Sedlak, M. Poly- and Perfluoroalkyl Substances in Wastewater: Significance of Unknown Precursors, Manufacturing Shifts, and Likely AFFF Impacts. *Water Res.* **2016**, *95*, 142−149.
(44) Masoner, J. R.; Kolpin, D. W.; Cozzarelli, I. M.; Smalling, K. L.; Bolyard, S. C.; Field, J. A.; Furlong, E. T.; Gray, J. L.; Lozinski, D.; Reinhart, D.; Rodowa, A.; Bradley, P. M. Landfill Leachate Contributes Per-/Poly-Fluoroalkyl Substances (PFAS) and Pharmaceuticals to Municipal Wastewater. *Environmental Science: Water Research and Technology* **2020**, *6* (5), 1300−1311.
(45) Sgroi, M.; Anumol, T.; Roccaro, P.; Vagliasindi, F. G. A.; Snyder, S. A. Modeling Emerging Contaminants Breakthrough in Packed Bed Adsorption Columns by UV Absorbance and Fluorescing Components of Dissolved Organic Matter. *Water Res.* **2018**, *145*, 667−677.
(46) Tavasoli, E.; Luek, J. L.; Malley, J. P.; Mouser, P. J. Distribution and Fate of Per- And Polyfluoroalkyl Substances (PFAS) in Wastewater Treatment Facilities. *Environmental Science: Processes and Impacts* **2021**, *23* (6), 903−913.
(47) Yanala, S. R.; Pagilla, K. R. Use of Biochar to Produce Reclaimed Water for Irrigation Use. *Chemosphere* **2020**, *251*, 126403.
(48) Gonzalez, D.; Thompson, K.; Quiñones, O.; Dickenson, E.; Bott, C. Granular Activated Carbon-Based Treatment and Mobility of per- and Polyfluoroalkyl Substances in Potable Reuse for Aquifer Recharge. *AWWA Water Science* **2021**, *3* (5), e1247.
(49) Rice, J.; Westerhoff, P. Spatial and Temporal Variation in de Facto Wastewater Reuse in Drinking Water Systems across the U.S.A. *Environ. Sci. Technol.* **2015**, *49* (2), 982−989.
(50) Brown, J. B.; Conder, J. M.; Arblaster, J. A.; Higgins, C. P. Assessing Human Health Risks from Per- And Polyfluoroalkyl Substance (PFAS)-Impacted Vegetable Consumption: A Tiered Modeling Approach. *Environ. Sci. Technol.* **2020**, *54* (23), 15202−15214.
(51) Pepper, I. L.; Brusseau, M. L.; Prevatt, F. J.; Escobar, B. A. Incidence of PFAS in Soil Following Long-Term Application of Class B Biosolids. *Science of The Total Environment* **2021**, *793*, 148449.
(52) Stanford, B. D.; Dickenson, E. R. V.; Wert, E.; Inyang, M. *Controlling Trace Organic Compounds Using Alternative, Non-FAT Technology for Potable Water Reuse*; Water Environment & Reuse Foundation: Alexandria, VA, 2017.
(53) Salveson, A.; Dickenson, E.; Soller, J.; Angelotti, B.; Parker, A. *Pathogen Risk Evaluation of Treatment and Monitoring System Performance for Potable Reuse*; Water Environment & Reuse Foundation: Alexandria, VA, 2018.
(54) Quiñones, O.; Snyder, S. A. Occurrence of Perfluoroalkyl Carboxylates and Sulfonates in Drinking Water Utilities and Related Waters from the United States. *Environ. Sci. Technol.* **2009**, *43* (24), 9089−9095.
(55) Gonzalez, D.; Thompson, K.; Quiñones, O.; Dickenson, E.; Bott, C. Assessment of PFAS Fate, Transport, and Treatment Inhibition Associated with a Simulated AFFF Release within a WASTEWATER Treatment Plant. *Chemosphere* **2021**, *262*, 127900.
(56) Zhang, S.; Szostek, B.; McCausland, P. K.; Wolstenholme, B. W.; Lu, X.; Wang, N.; Buck, R. C. 6:2 and 8:2 Fluorotelomer Alcohol Anaerobic Biotransformation in Digester Sludge from a WWTP under Methanogenic Conditions. *Environ. Sci. Technol.* **2013**, *47* (9), 4227−4235.
(57) Hamid, H.; Li, L. Y.; Grace, J. R. Aerobic Biotransformation of Fluorotelomer Compounds in Landfill Leachate-Sediment. *Sci. Total Environ.* **2020**, *713*, 136547.
(58) Houtz, E.; Wang, M.; Park, J. S. Identification and Fate of Aqueous Film Forming Foam Derived Per- and Polyfluoroalkyl Substances in a Wastewater Treatment Plant. *Environ. Sci. Technol.* **2018**, *52* (22), 13212−13221.
(59) Kahl, S.; Kleinsteuber, S.; Nivala, J.; van Afferden, M.; Reemtsma, T. Emerging Biodegradation of the Previously Persistent Artificial Sweetener Acesulfame in Biological Wastewater Treatment. *Environ. Sci. Technol.* **2018**, *52* (5), 2717−2725.
(60) Vieno, N. M.; Tuhkanen, T.; Kronberg, L. Seasonal Variation in the Occurrence of Pharmaceuticals in Effluents from a Sewage Treatment Plant and in the Recipient Water. *Environ. Sci. Technol.* **2005**, *39* (21), 8220−8226.
(61) Procopio, N. A.; Karl, R.; Goodrow, S. M.; Maggio, J.; Louis, J. B.; Atherholt, T. B. Occurrence and Source Identification of Perfluoroalkyl Acids (PFAAs) in the Metedeconk River Watershed, New Jersey. *Environmental Science and Pollution Research* **2017**, *24* (35), 27125−27135.
(62) Sedlak, M. D.; Benskin, J. P.; Wong, A.; Grace, R.; Greig, D. J. Per- and Polyfluoroalkyl Substances (PFASs) in San Francisco Bay Wildlife: Temporal Trends, Exposure Pathways, and Notable Presence of Precursor Compounds. *Chemosphere* **2017**, *185*, 1217−1226.
(63) Venkatesan, A. K.; Halden, R. U. National Inventory of Perfluoroalkyl Substances in Archived U.S. Biosolids from the 2001 EPA National Sewage Sludge Survey. *Journal of Hazardous Materials* **2013**, *252*−*253*, 413−418.
(64) Kim Lazcano, R.; de Perre, C.; Mashtare, M. L.; Lee, L. S. Per- and Polyfluoroalkyl Substances in Commercially Available Biosolid-Based Products: The Effect of Treatment Processes. *Water Environ. Res.* **2019**, *91* (12), 1669−1677.
(65) Higgins, C. P.; Field, J. A.; Criddle, C. S.; Luthy, R. G. Quantitative Determination of Perfluorochemicals in Sediments and Domestic Sludge. *Environ. Sci. Technol.* **2005**, *39* (11), 3946−3956.
(66) Reiner, J. L.; Blaine, A. C.; Higgins, C. P.; Huset, C.; Jenkins, T. M.; Kwadijk, C. J. A. F.; Lange, C. C.; Muir, D. C. G.; Reagen, W. K.; Rich, C.; Small, J. M.; Strynar, M. J.; Washington, J. W.; Yoo, H.; Keller, J. M. Polyfluorinated Substances in Abiotic Standard Reference Materials. *Anal. Bioanal. Chem.* **2015**, *407* (11), 2975−2983.
(67) Yoo, H.; Washington, J. W.; Jenkins, T. M.; Laurence Libelo, E. Analysis of Perfluorinated Chemicals in Sludge: Method Development and Initial Results. *Journal of Chromatography A* **2009**, *1216* (45), 7831−7839.
(68) Boulanger, B.; Vargo, J. D.; Schnoor, J. L.; Hornbuckle, K. C. Evaluation of Perfluorooctane Surfactants in a Wastewater Treatment System and in a Commercial Surface Protection Product. *Environ. Sci. Technol.* **2005**, *39* (15), 5524−5530.
(69) Furl, C. v.; Meredith, C. A.; Strynar, M. J.; Nakayama, S. F. Relative Importance of Wastewater Treatment Plants and Non-Point Sources of Perfluorinated Compounds to Washington State Rivers. *Sci. Total Environ.* **2011**, *409* (15), 2902−2907.
(70) Loganathan, B. G.; Sajwan, K. S.; Sinclair, E.; Senthil Kumar, K.; Kannan, K. Perfluoroalkyl Sulfonates and Perfluorocarboxylates in Two Wastewater Treatment Facilities in Kentucky and Georgia. *Water Res.* **2007**, *41* (20), 4611−4620.
(71) Plumlee, M. H.; Larabee, J.; Reinhard, M. Perfluorochemicals in Water Reuse. *Chemosphere* **2008**, *72* (10), 1541−1547.
(72) Quiñones, O.; Snyder, S. A. Occurrence of Perfluoroalkyl Carboxylates and Sulfonates in Drinking Water Utilities and Related Waters from the United States. *Environ. Sci. Technol.* **2009**, *43* (24), 9089−9095.
(73) Schultz, M. M.; Barofsky, D. F.; Field, J. A. Quantitative Determination of Fluorinated Alkyl Substances by Large-Volume-Injection Liquid Chromatography Tandem Mass Spectrometry - Characterization of Municipal Wastewaters. *Environ. Sci. Technol.* **2006**, *40* (1), 289−295.
(74) Scott, B. F.; De Silva, A. O.; Spencer, C.; Lopez, E.; Backus, S. M.; Muir, D. C. G. Perfluoroalkyl Acids in Lake Superior Water: Trends and Sources. *J. Great Lakes Res.* **2010**, *36* (2), 277−284.
(75) 3M Environmental Laboratory. *Environmental Monitoring - Multi-City Study: Water, Sludge, Sediment, POTW Effluent and Landfill Leachate Samples*; 3M: Minneapolis, MN, 2001.

(76) Antweiler, R. C. Evaluation of Statistical Treatments of Left-Censored Environmental Data Using Coincident Uncensored Data Sets. II. Group Comparisons. *Environ. Sci. Technol.* **2015**, *49* (22), 13439−13446.
(77) Antweiler, R. C.; Taylor, H. E. Evaluation of Statistical Treatments of Left-Censored Environmental Data Using Coincident Uncensored Data Sets: I. Summary Statistics. *Environ. Sci. Technol.* **2008**, *42* (10), 3732−3738.
(78) USEPA. National Water Reuse Action Plan. US Environmental Protection Agency: Washington, DC, USA, 2020.
(79) Beecher, N.; Crawford, K.; Goldstein, N.; Kester, G.; Lono-Batura, M.; Dziezyk, E. *A National Siosolids Regulation, Quality End Use and Disposal Survey*; North East Biosolids and Residuals Association: Tamworth, NH, 2007.
(80) Tukey, J. W. *Exploratory Data Analysis*; Addison-Wesley: Reading, MA, 1977.
(81) Brant, R. Comparing Classical and Resistant Outlier Rules. *Journal of the American Statistical Association* **1990**, *85* (412), 1083−1090.
(82) Zijlstra, W. P.; van der Ark, L. A.; Sijtsma, K. Outlier Detection in Test and Questionnaire Data. *Multivariate Behavioral Research* **2007**, *42* (3), 531−555.
(83) Stehman, S. v. Estimating Area and Map Accuracy for Stratified Random Sampling When the Strata Are Different from the Map Classes. *International Journal of Remote Sensing* **2014**, *35* (13), 4923−4939.
(84) Schaefer, C. E.; Guelfo, J. L.; Dickenson, E. R. V.; Knappe, D. R. U.; Hooper, J. L.; RamMohan, G.; Drennan, D. Occurrence of PFAS Compounds in U.S. Wastewater Treatment Plants. Project 5031; The Water Research Foundation: Denver, CO, 2022.
(85) Gerrity, D.; Papp, K.; Stoker, M.; Sims, A.; Frehner, W. Early-Pandemic Wastewater Surveillance of SARS-CoV-2 in Southern Nevada: Methodology, Occurrence, and Incidence/Prevalence Considerations. *Water Research X* **2021**, *10*, 100086.
(86) George, D.; Mallery, M. *SPSS for Windows Step by Step: A Simple Guide and Reference*, 10th ed.; Pearson: Boston, 2010.
(87) Wang, N.; Liu, J.; Buck, R. C.; Korzeniowski, S. H.; Wolstenholme, B. W.; Folsom, P. W.; Sulecki, L. M. 6:2 Fluorotelomer Sulfonate Aerobic Biotransformation in Activated Sludge of Waste Water Treatment Plants. *Chemosphere* **2011**, *82* (6), 853−858.
(88) Zhao, L.; Mccausland, P. K.; Folsom, P. W.; Wolstenholme, B. W.; Sun, H.; Wang, N.; Buck, R. C. 6:2 Fluorotelomer Alcohol Aerobic Biotransformation in Activated Sludge from Two Domestic Wastewater Treatment Plants. *Chemosphere* **2013**, *92* (4), 464−470.
(89) Wang, Z.; Cousins, I. T.; Scheringer, M.; Buck, R. C.; Hungerbühler, K. Global Emission Inventories for C4-C14 Perfluoroalkyl Carboxylic Acid (PFCA) Homologues from 1951 to 2030, Part I: Production and Emissions from Quantifiable Sources. *Environ. Int.* **2014**, *70*, 62−75.
(90) Gluge, J.; Scheringer, M.; Cousins, I. T.; DeWitt, J. C.; Goldenman, G.; Herzke, D.; Lohmann, R.; Ng, C. A.; Trier, X.; Wang, Z. An Overview of the Uses of Per- and Polyfluoroalkyl Substances (PFAS). *Environmental Science: Processes & Impacts* **2020**, *22* (12), 2345−2373.
(91) Health Advisory for Perfluorooctanoic Acid (PFOA) Chemical Abstract Service Registry Number (CASRN) 335-67-1. Illinois Environmental Protection Agency: Springfield, IL, 2021.
(92) EWG. UPDATE: Thousands of Industrial Facilities Likely Discharging Toxic 'Forever Chemicals' into Air and Water. https://www.ewg.org/news-insights/news/update-thousands-industrial-facilities-likely-discharging-toxic-forever (accessed 2021-10-08).
(93) USEPA. Project and Landfill Data by State. https://www.epa.gov/lmop/project-and-landfill-data-state (accessed 2020-09-24).
(94) Lang, J. R.; Allred, B. M. K.; Field, J. A.; Levis, J. W.; Barlaz, M. A. National Estimate of Per- and Polyfluoroalkyl Substance (PFAS) Release to U.S. Municipal Landfill Leachate. *Environ. Sci. Technol.* **2017**, *51* (4), 2197−2205.
(95) Grandjean, P.; Timmermann, C. A. G.; Kruse, M.; Nielsen, F.; Vinholt, P. J.; Boding, L.; Heilmann, C.; Molbak, K. Severity of COVID-19 at Elevated Exposure to Perfluorinated Alkylates. *PLoS One* **2020**, *15* (12), e0244815.
(96) Boulanger, B.; Vargo, J. D.; Schnoor, J. L.; Hornbuckle, K. C. Evaluation of Perfluorooctane Surfactants in a Wastewater Treatment System and in a Commercial Surface Protection Product. *Environ. Sci. Technol.* **2005**, *39* (15), 5524−5530.
(97) Grandjean, P.; Budtz-Jørgensen, E. An Ignored Risk Factor in Toxicology: The Total Imprecision of Exposure Assessment. *Pure Appl. Chem.* **2010**, *82* (2), 383−391.
(98) Arvaniti, O. S.; Andersen, H. R.; Thomaidis, N. S.; Stasinakis, A. S. Sorption of Perfluorinated Compounds onto Different Types of Sewage Sludge and Assessment of Its Importance during Wastewater Treatment. *Chemosphere* **2014**, *111*, 405−411.

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ACS EST Water 2022, 2, 690−700

If the subject property has ever been a part or used for the following, there is a potential that significant PFAS contamination is present at the subject property. If your site meets any of the conditions below, then PFAS should be discussed in the Phase I Environmental Site Assessment (if a discussion of PFAS is approved by the client as a non-scope item).

On-Site Sources

GOVERNMENT & MILITARY INSTALLATIONS USE OF PFAS (High Risk)	
Government/military facilities that utilized AFFF for fire training	☐ No ☐ Yes
Electroplating operations at federal facilities and government-owned, contractor-operated (GOCO) research and development plants	☐ No ☐ Yes
Wastewater treatment plants (WWTP) at federal installations	☐ No ☐ Yes
COMMERCIAL & INDUSTRIAL USES OF PFAS (High Risk)	
Electroplating Note: Electroplating (specifically hard chromium plating) is an industrial activity where PFAS-containing mist suppressants may have been used	☐ No ☐ Yes
Landfill operations, waste disposal areas, and wastewater treatment plants	☐ No ☐ Yes
Open burning / open detonation of munitions	☐ No ☐ Yes
Agricultural uses (1970s-present) that included application of biosolids/WWTP sludge (that potentially contained PFAS) on bare soils	☐ No ☐ Yes
Refineries and petrochemical industries that utilized AFFF fire training areas (1960s-present)	☐ No ☐ Yes
Public- and private-sector airports (any AFFF utilized on-site for fire training) (1960s-present)	☐ No ☐ Yes
PFAS manufacturer or any chemical industry with a special focus on processing aids in the polymerization of fluoropolymers	☐ No ☐ Yes
Any large past fire events, or any known or suspected use of AFFF on site (1960s-present)	☐ No ☐ Yes
Was the property a fire station (with a fire training area) or have any fire training areas (1960s-present)	☐ No ☐ Yes
Large industrial facility with an AFFF firefighting system that requires yearly testing and discharge of AFFF during testing (1960s-present)	☐ No ☐ Yes

COMMERCIAL & INDUSTRIAL USES OF PFAS High to Moderate Risk (if there are known exposure pathways)	
Section A	
Surface protection of textile, apparel, leather, carpets, and paper	☐ No ☐ Yes
Electronics industry	☐ No ☐ Yes
Plastics and rubber production	☐ No ☐ Yes
Coatings, paints, and varnishes	☐ No ☐ Yes
Munitions and explosives production	☐ No ☐ Yes
Aircraft and heavy equipment manufacturing	☐ No ☐ Yes
Carwash, Laundry, Dry cleaner	☐ No ☐ Yes
Industry listed in the PFAS NAICS/SIC Code Reference Table below	☐ No ☐ Yes
Section B: If "Yes" was checked for any of the items in Section A, were the following identified:	
Drains, sumps, pits, and/or subsurface conduits that potentially contained/discharged PFAS	☐ No ☐ Yes
Large-scale operation	☐ No ☐ Yes
Smokestacks or aerial dispersion that potentially contained/discharged PFAS	☐ No ☐ Yes
Permitted (NPDES) and potentially unpermitted wastewater discharges that potentially contained/discharged PFAS (e.g. NPDES listing in the database for industrial waste water discharges) water discharges)	☐ No ☐ Yes
Any large containers or drums containing of PFAS (raw or mixture) that may have potentially leaked or discharged to the subsurface	☐ No ☐ Yes
Any known or suspected conditions that would result in a potential release of raw materials or wastes from the site operations	☐ No ☐ Yes

Index Codes

This table provides the first three digits of industries that may have used compounds containing PFAS in their manufacturing or production processes.

To determine three digit code(s) associated with current/former on-site operations, review RCRA listing details in the regulatory database. Take the first three or four digits of the NAICS code(s) listed and look for them in the table below. For example:

NAICS Code: 331316 (Aluminum extruded product manufacturing)

NAICS Code: 332112 (Nonferrous forging)

Search 331 and 332 in the table below.

In this case, since both codes (331 & 332) are listed, it indicates the business may have used PFAS.

Index Codes				
3&4-Digit	**Industry**		**6-digit**	**Sector**
313	Textiles			
314	Textile Product			
315	Apparel			
316	Leather			
321	Wood Products			
322	Paper			
323	Printing			
324	Petroleum			
325	Chemicals			
326	Plastics and Rubber			
327	Nonmetallic Mineral Product			
331	Primary Metals			
332	Fabricated Metals			
			332992	Small Arms Ammunition
			332993	Ammunition-except small arms
333	Machinery			
334	Computers/Electronic Products			
335	Electrical Equipment			
336	Transportation Equipment			
3&4-Digit	**Industry**		**6-digit**	**Sector**
337	Furniture			
339	Miscellaneous Manufacturing			
511	Publishing			
562	Hazardous Waste			
	Other		111998	All Other Miscellaneous Crop Farming
	Other		113310	Logging
	Other		211112	Natural Gas Liquid Extraction
	Other		212324	Kaolin and Ball Clay Mining
	Other		212325	Clay and Ceramic and Refractory Minerals Mining
	Other		212393	Other Chemical and Fertilizer Mineral Mining
	Other		212399	All Other Nonmetallic Mineral Mining

Index Codes				
	Other		221330	Steam and Air-Conditioning Supply
	Other		488390	Other Support Activities for Water Transportation
	Other		512220	Integrated Record Production/Distribution
	Other		541710	R&D in the Physical, Engineering, and Life Sciences
	Other		541712	R&D in the Physical, Engineering, and Life Sciences (except Biotechnology)
	Other		541713	Research and Development in Nanotechnology
	Other		541715	R&D in the Physical, Engineering, and Life Sciences (except Nanotechnology and Biotechnology)
	Other		811490	Other Personal and Household Goods Repair and Maintenance
2121	Coal Mining		212111	Bituminous Coal and Lignite Surface Mining
2121	Coal Mining		212112	Bituminous Coal Underground Mining
2121	Coal Mining		212113	Anthracite Mining
2122	Metal Mining		212221	Gold Ore Mining
2122	Metal Mining		212222	Silver Ore Mining
2122	Metal Mining		212230	Copper, Nickel, Lead and Zinc Mining
2122	Metal Mining		212231	Lead Ore and Zinc Ore Mining
2122	Metal Mining		212234	Copper Ore and Nickel Ore Mining
2122	Metal Mining		212299	All Other Metal Ore Mining
2211	Electric Utilities		221111	Hydroelectric Power Generation
2211	Electric Utilities		221112	Fossil Fuel Electric Power Generation
2211	Electric Utilities		221113	Nuclear Electric Power Generation
2211	Electric Utilities		221114	Solar Electric Power Generation
2211	Electric Utilities		221115	Wind Electric Power Generation

Index Codes				
2211	Electric Utilities		221116	Geothermal Electric Power Generation
2211	Electric Utilities		221117	Biomass Electric Power Generation
2211	Electric Utilities		221118	Other Electric Power Generation
2211	Electric Utilities		221119	Other Electric Power Generation
2211	Electric Utilities		221122	Electric Power Distribution Transmission and Control
4246	Chemical Wholesalers		424690	Other Chemical and Allied Products Merchant Wholesalers
4247	Petroleum Bulk Terminals		424710	Petroleum Bulk Stations and Terminals

POTENTIAL PFAS CONTAMINATION CHECKLIST

If you identify any of the following off-site conditions, there is potential for PFAS contamination. If any of the following are identified, then PFAS may need to be discussed in the Phase I Environmental Site Assessment (if a discussion of PFAS is approved by the client as a non-scope item).

On-Site Sources

1 mile	
DOD, FUDS, or Military Base (that would have utilized AFFF) within 1 mile of subject property (SP)? *	☐ No ☐ Yes
0.5 mile	
Airport (large enough to have a fire training area) within 0.5 mile of SP?	☐ No ☐ Yes
1,000 feet	
Large petroleum refinery (that would use AFFF) within 1,000 feet of SP?	☐ No ☐ Yes
Landfill or WWTP within 500 feet of SP?	☐ No ☐ Yes

500 feet	
Elevated PFAS detected in well (groundwater or drinking water**) within 500 feet of SP?	☐ No ☐ Yes
Chromium plating facility within 500 feet of SP that has identified or potential contamination (i.e., release or VCP listings in the database report)?	☐ No ☐ Yes
Large-scale industrial facility with large-scale wastewater discharges within 500 feet of SP that has identified or potential contamination (i.e., release or VCP listings in the database report)?	☐ No ☐ Yes
Landfill or WWTP within 500 feet of SP?	☐ No ☐ Yes
Adjoining	
High risk industrial operations adjoining SP that includes wastewater discharges (NPDES listings) or significant aerial dispersion (air permit listings)?	☐ No ☐ Yes

* Check the below link to determine if PFAS are under investigation:

Installations Where DOD is Performing Assessment of PFAS Use/Potential Release

If the site is listed, search online for any publicly available information (i.e., in states like CA, information will be on GeoTracker; in other states, information may not yet be available to the public.

**A drinking water sample may not be representative of groundwater conditions. If elevated drinking water is identified, you may need to confirm its source at the subject property (i.e., public drinking water supply or private drinking water well).

- If public drinking water is the source and it is verified to be from an off-site source, then it does not represent localized groundwater conditions.
- If a private drinking water well is the source, then it will represent groundwater conditions.

NEXT STEPS

AEI's team of geologists and engineers are experienced in the investigation and remediation of PFAS contaminated properties. Every site will have its unique challenges.

Please visit **www.aeiconsultants.com** or contact your AEI representative for assistance. **1.800.801.3224** | **info@aeiconsultants.com**

ADDITIONAL RESOURCES

Interactive Map of PFAS sites

www.ewg.org/interactive-maps/pfas_contamination/map/

Military Installations were the DOD is assessing PFAS

media.defense.gov/2021/Mar/23/2002606229/-1/-1/0/INSTALLATIONS-BEING-ASSESSED-FOR-PFAS-USE-OR-POTENTIAL-RELEASE-AS-OF-DEC-31-2020.PDF/INSTALLATIONS-BEING-ASSESSED-FOR-PFAS-USE-OR-POTENTIAL-RELEASE-AS-OF-DEC-31-2020.PDF

https://partner-mco-archive.s3.amazonaws.com/client_files/1524589484.pdf

AFF

www.michigan.gov/pfasresponse/investigations/firefighting-foam

SDWIS Federal Reports Search

ordspub.epa.gov/ords/sfdw/sfdw/r/sdwis_fed_reports_public/200

ITRC

pfas-1.itrcweb.org/
pfas-1.itrcweb.org/fact-sheets/

ADDITIONAL RESOURCES

Superfund/NPL

www.epw.senate.gov/public/index.cfm?p=Superfund-Sites-Identified-by-EPA-to-have-PFAS-Contamination

EPA

www.epa.gov/system/files/documents/2021-10/pfas-roadmap_final-508.pdf
www.epa.gov/chemical-research/status-epa-research-and-development-pfas
www.epa.gov/sites/default/files/2016-05/documents/pfoa_health_advisory_final_508.pdf
www.federalregister.gov/documents/2020/03/10/2020-04145/announcement-of-preliminary-regulatory-determinations-for-contaminants-on-the-fourth-drinking-water
www.federalregister.gov/documents/2021/03/03/2021-04184/announcement-of-final-regulatory-determinations-for-contaminants-on-the-fourth-drinking-water
www.epa.gov/dwucmr/fifth-unregulated-contaminant-monitoring-rule
ordspub.epa.gov/ords/guideme_ext/f?p=104%3A41
enviro.epa.gov/
www.epa.gov/enviro/tri-search
www.epa.gov/research-states/pfas-science-webinars-epa-region-1-and-state-tribal-partners

NAICS/SIC

www.census.gov/naics/?48967
www.osha.gov/data/sic-search
pubs.rsc.org/en/content/articlelanding/2020/em/d0em00291g
www.nj.gov/dep/srp/guidance/srra/pfas_handling_industry_sectors.pdf
www.pca.state.mn.us/sites/default/files/p-gen1-22b.pdf

State regulations

www.saferstates.com/toxic-chemicals/pfas/

Mass

www.mass.gov/doc/final-pfas-related-changes-to-the-mcp-2019-12-13/download
www.mass.gov/doc/pfas-mcl-revisions-to-310-cmr-2200-clean-version-9-16-2020/download

Portland Water District Joins Wastewater Multidistrict Law Suit (MDL) Against PFAS Manufacturers

06.05.2024

The Portland Water District (PWD) filed a complaint in the U.S. District Court, against manufacturers of per- and polyfluoroalkyl substances (PFAS) chemicals, 3M, DuPont, and others. The suit seeks to recover costs associated with PFAS mitigation in wastewater treatment, including treatment byproducts, effluent, and biosolids.

Protecting public health, safety, and the environment is PWD's top priority. By taking legal action against manufacturers of PFAS, PWD is holding accountable those responsible for pollution," stated PWD's General Manager Seth Garrison.

PFAS are persistent, toxic, and bioaccumulative compounds that are found in numerous products. They have been found virtually everywhere in the environment. PFAS contamination impacts both surface water (lakes, rivers, oceans, etc.) and groundwater, as well as soils and living organisms. The widespread presence of PFAS in consumer and commercial products means PFAS is present in landfills and wastewater treatment systems.

Even though there is no measurable PFAS in Greater Portland's drinking water, it has been found in wastewater. PWD's four treatment facilities show 15-32 parts per trillion (ppt) of PFAS in the effluent.

In response to PFAS contamination, Maine became the first and only state in the nation to end the beneficial use (land application) of wastewater biosolids, which created a series of challenges for Maine's wastewater systems and produced landfill capacity problems. As a result, PWD has taken a proactive approach to finding solutions for biosolids management, including commissioning a Biosolids Master Plan to examine steps to manage PFAS-containing biosolids.

In collaboration with a team of law firms, SL Environmental Law Group is handling the Portland Water District's case along with several other wastewater systems across the country.

PWD is also a claimant in a federal multi-district case related to water utilities and PFAS contamination. Because PWD has no measurable PFAS in the drinking water, it is designated as a Class 2 claimant and reimbursement will be limited to covering the costs of the extra testing that has been required.

PWD is committed to protecting public health and supports keeping PFAS out of the environment. It supports efforts to reduce PFAS at its sources, including keeping them out of consumer products. PWD is proactively engaged at the local, state, and national levels, exploring solutions to combat the widespread PFAS problem.

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PFAS Removal from Wastewater: Traditional vs Modern Treatment Methods

4.1.24

In the ongoing quest to combat water contamination, water and wastewater utilities face an evolving battlefield marked by pervasive pollutants like per- and poly-

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public health, the limitations of conventional wastewater treatment technologies become apparent. The 20th century's wastewater treatment processes, sufficient for the pollutants they target, fall short against a new generation of **contaminants like PFAS**. Proactively addressing modern water and wastewater pollution with more advanced technologies can help community leaders protect their water sources and be prepared for upcoming regulations.

In this article, we'll illuminate the differences between conventional and advanced treatment technologies and their abilities to remove PFAS from water to serve as a guide for the type of technologies wastewater treatment plants may need in the 21st century.

Traditional Methods for Wastewater Treatment

Traditional treatment techniques have managed **wastewater pollution** in cities across the world for over a century. They've proven effective at removing organic matter, bacteria, and other household contaminants from sewage to maintain the health of water bodies that provide communities with drinking water and recreation. However, these established methods show significant treatment deficiencies when confronted with PFAS. Sometimes referred to as "forever chemicals," PFAS resist degradation and pass through the physical, chemical, and biological treatment processes that were designed for more organic pollutants. These treatment stages, effective

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Sedimentation and Filtration:

Physical treatments like sedimentation and filtration separate solids from wastewater. They act as filters that facilitate the efficacy of downstream treatment stages to ultimately comply with Clean Water Act standards. While physical treatment removes organic solids and nutrients, PFAS move onto subsequent treatment stages unabated due to their solubility in water, bypassing most physical barriers found at wastewater treatment plants. PFAS can be removed from wastewater via biosolids or filtered out by activated carbon and certain membrane filters common in water treatment. However, these processes do not destroy PFAS, leaving open the possibility for them to wind up elsewhere in the system.

Coagulation and Flocculation:

Chemistry plays an essential role in wastewater treatment. Chemical treatment processes like coagulation and flocculation facilitate physical and biological treatment methods, bringing certainty to a sensitive system. Coagulant is added to wastewater to bind naturally repelling organic materials to one another and stirred by "mixers" to form what are called "floc." These heavier, agglomerated particles weigh themselves down to precipitate in the sedimentation stage.

Coagulation and flocculation are proven methods in treating typical sewage. However, the unique properties of the many PFAS chemicals make them immune to the binding properties of coagulants and polymers. Unaffected by the chemical treatment process, many PFAS remain dissolved in wastewater, though organic solids often harbor PFAS and are found in the biosolids handling stage of wastewater treatment.

naturally occurring nitrogen cycle. These bacteria are fostered by aerobic and anaerobic zones that transform toxic substances like ammonia into nitrogen gas, which off-gasses into the atmosphere. While this process is routinely effective at removing ammonia, nitrogen compounds, and phosphorus from wastewater, it does not biodegrade PFAS compounds. PFAS' robustness means they can pass through the biological treatment phase untouched as other pollutants diminish.

These traditional treatment methods, while effective against their original target contaminants, now obligate wastewater plants to shift towards more innovative technologies in order to meet the modern challenges posed by PFAS contamination and other contaminants of emerging concern. The adoption of modern treatment solutions may be unavoidable for these facilities to comply with regulations, protect public health, and stop the pollution cycle.

Modern Methods for PFAS Removal

In response to the treatment barriers posed by PFAS and other contaminants, the wastewater sector is increasingly turning towards solutions that complement and enhance traditional treatment methods. These modern approaches address the limitations of conventional techniques, promising effective PFAS removal from wastewater.

Membrane Bioreactors (MBRs)

Initially incorporated within the aeration basin to replace bulky clarifiers at space-deficient treatment plants, MBRs are a membrane filtration technology that effectively

membranes used in these systems feature micro-lesions that are so small that bacteria, viruses, and even PFAS cannot physically pass through them. This method enhances treatment performance and consistently ensures the physical separation of PFAS from wastewater. While MBRs deliver PFAS-free effluent that meets upcoming regulations, they do not destroy PFAS, leaving them and other contaminants to absorb into biosolids.

Granular Activated Carbon (GAC) Adsorption

Activated carbon reacts with almost everything. Its ability to absorb dissolved contaminants in water has made it a standout treatment for a wide array of pollutants, including PFAS. Commonly used in water treatment plants, wastewater plants wanting to remove PFAS from their effluent can employ these same carbon filter beds to facilitate a chemical reaction between activated carbon and the pollutants within wastewater. GAC effectively removes long-chain PFAS, is easy to procure, and relatively **cost-effective**. However, GAC systems often require the most space compared to other PFAS treatments, struggle to remove short-chain PFAS, and require longer retention times than ion exchange resins.

Further innovations, such as combining GAC with proprietary media like Fluoro-sorb™, show promise in enhancing PFAS removal rates, removing of a broader span of PFAS chemicals, as well as the lifespan of GAC media.

Advanced Oxidation Processes (AOPs)

AOPs are a cutting-edge approach against PFAS and other hard-to-treat chemicals, combining hydrogen peroxide with ozone or UV light to generate potent hydroxyl radicals. Hydroxyl radicals react with and break

creates a pathway to significantly mitigate their environmental impact. AOPs offer a chance for treatment plants to destroy PFAS on site in addition to complying with environmental regulations. However, the consumption of hydrogen peroxide and ozone generation can be costly. Its practicality is case-by-case.

These modern treatment methods underscore a pivotal shift towards more effective strategies in combating PFAS contamination. However, they come with a financial burden not all communities are ready to take on.

Cost Effectiveness and Treatment Efficiency

Navigating the financial complexities of modernizing wastewater treatment facilities to address contamination requires innovative thinking and strategic planning. The shift towards advanced treatment methods, while necessary for environmental and public health, presents a significant financial undertaking for wastewater utilities nationwide. It's critical to consult a professional engineer to analyze your contamination situation and determine the best method for your system. This involves a thorough analysis of treatment options, potential for scalability, and the long-term applicability of equipment investments in the face of increasingly stringent regulations.

A proactive approach coupled with an understanding of available cost recovery mechanisms can mitigate the financial burden. By exploring grants, state funding, and legal avenues for cost recovery, wastewater utilities can

For communities that want to take on a proactive approach to removing PFAS from their wastewater, SL environmental law group offers a **Cost Recovery Guide for Wastewater Utilities**. If your community is interested in exploring how litigation against the manufacturers responsible for the PFAS contamination can fund the next generation of wastewater treatment equipment at your facility, **contact us** today for a free consultation.

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