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Fate of PFAS in the Aquatic Environment: From Source to Sink

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Layman Summary

Per- and polyfluoroalkyl substances (PFAS) are a group of chemicals widely used in everyday products and industrial processes since the 1940s. These substances have drawn attention due to their persistence in the environment and potential health risks. PFAS can accumulate in aquatic environments, posing threats to ecosystems and potentially human health.

In this review, we explore pathways of PFAS in water systems, from their sources to where they end up. There are thousands of different PFAS compounds, but most research has focused on only a handful of them. This leaves a significant gap in our understanding of how these different chemicals behave in the environment.

PFAS can enter water bodies from various sources like industrial sites and everyday household objects, and they tend to persist once they are released. They can travel through rivers and streams, eventually reaching coastal and marine environments. They can be found almost everywhere in the world's waters, from rivers in industrialised cities to rural coastal waters.

Within water, PFAS can undergo changes. Some compounds break down into other forms, while others persist and accumulate. The review highlights that many studies concentrate on specific PFAS types, like perfluoroalkyl acids, while overlooking the broader diversity of PFAS and their fate.

The review emphasises the need to better understand this diversity. PFAS vary greatly in their chemical composition and behaviour, which affects how they move and accumulate in water and sediments. Current studies often focus on measuring a limited set of well-known PFAS, neglecting thousands of other variants that could be equally important.

To manage PFAS contamination effectively, more attention to detecting and characterising lesser-known PFAS and understanding how they interact with the environment is necessary. This means developing more precise methods to measure PFAS concentrations across different environmental compartments, and within an aquatic ecosystem itself.

In summary, PFAS present a complex challenge in aquatic environments due to their diversity and persistence. While much research has been done on specific PFAS types, there is a need to expand our understanding to include a broader range of these chemicals and their interactions in the aquatic environment. This knowledge will support developing strategies to mitigate the environmental impact of PFAS contamination and protect aquatic ecosystems in the long term.

Abstract

Per- and polyfluoroalkyl substances (PFAS) represent a diverse group of chemical compounds extensively used in consumer and industrial applications, contributing to persistent environmental contamination and associated health risks. This review explores the fate of PFAS in aquatic environments, synthesising existing knowledge and highlighting key research gaps. PFAS exhibit complex behaviours within aquatic ecosystems, influenced by diverse molecular structures and environmental conditions. Their accumulation in aquatic environments poses risks to organisms and ecosystems, with potential far-reaching consequences in highly industrial areas, as well as regions with little to no human interaction. This review underscores the need to comprehend the step-by-step pathways of PFAS fate in aquatic environments, encompassing different types of PFAS, sources, transport mechanisms, and final destinations. Existing studies predominantly focus on a limited subset of well-known PFAS compounds, leaving numerous other PFAS variants understudied. Key findings reveal that diverse PFAS, from well-known perfluoroalkyl acids to emerging alternatives like fluorotelomers and fluoropolymers, undergo transformations leading to the accumulation of terminal perfluoroalkyl acids in aquatic environments. However, the majority of studies overlook this diversity, concentrating on specific PFAS subsets and overlooking crucial transportation pathways. To advance our understanding, future research should prioritise detecting and characterising understudied PFAS and their interactions in various aquatic compartments. More specialised analytical approaches are needed to accurately assess PFAS concentrations and behaviours across interfaces and between aquatic compartments, enhancing our ability to mitigate PFAS-related environmental risks effectively. This review consolidates current knowledge on PFAS dynamics in aquatic environments, emphasising the need for comprehensive assessments of PFAS diversity and behaviours to guide effective environmental management strategies and future research.

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LIST OF ABBREVIATIONS

Abbreviation	Definition
<i>Chemical class</i>	
PFAS	Per- and polyfluoroalkyl substances
PFAA	Perfluoroalkyl acids
PFCAs	Perfluorocarboxylic acids
PFSAs	Perfluorosulfonic acids
FASAs	Perfluoroalkane sulfonamides
FTOH	Fluorotelomer alcohol
FTCA	Fluorotelomer carboxylic acid
FTSA	Fluorotelomer sulfonic acid
FTAB	Fluorotelomer sulfonamide alkylbetaine
FASEs	Fluoroalkane sulfonamidoethanols
FASAAAs	Fluoroalkane sulfonamidoacetic acids
PAPs	Polyfluoroalkyl phosphate esters
PFPEs	Perfluoropolyethers
<i>Chemical substance</i>	
PFBA	Perfluorobutanoic acid
PFPeA	Perfluoropentanoic acid
PFH _x A	Perfluorohexanoic acid
PFHpA	Perfluoroheptanoic acid
PFOA	Perfluorooctanoic acid
PFDA	Perfluorodecanoic acid
PFBS	Perfluorobutanoic acid
PFH _x S	Perfluorohexane sulfonic acid
PFOS	Perfluorooctane sulfonic acid
PFOSA	Perfluorooctanesulfonamide
PTFE	Polytetrafluoroethylene
HFPO-DA	Hexafluoropropylene oxide dimer acid
<i>General terminology</i>	
AFFF	Aqueous film-forming foam
ECF	Electrochemical fluorination
FTF	Firefighting training facility
NAPL	Nonaqueous phase liquids
K _{oc}	Organic carbon-water partition coefficient
OECD	Organisation for Economic Co-operation and Development
WWTP	Wastewater treatment plants

1. INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) constitute a group of chemical compounds that have been used since the 1940s for a broad range of consumer products and industrial applications (Buck et al., 2011). Additionally, they have gained significant attention due to their environmental persistence and toxicological effects on both ecosystems and human health (DeWitt et al., 2019; East et al., 2023; Fenton et al., 2021; Flynn et al., 2022; Liu et al., 2019; Ma et al., 2022; Panieri et al., 2022). The complexity of the PFAS class is underscored by the vast array of chemical compounds it encompasses. While approximately 4700 substances are commonly classified as PFAS by the Organisation for Economic Co-operation and Development (OECD; (OECD, 2018)), alternative definitions extend this count to as high as 7 million (Schymanski et al., 2023; Z. Wang et al., 2021). Despite this apparent diversity, much of the discourse within the scientific literature converges upon a select few PFAS compounds and their precursors.

PFAS compounds exhibit a notable trend of accumulation in the aquatic environment, posing a significant risk to aquatic organisms and, consequently, the broader ecosystem (Cara et al., 2022; Khan et al., 2023; Miranda et al., 2021; Sun et al., 2022; Tansel, 2024; W. Wang et al., 2020). The movement of PFAS in the aquatic environment from their sources to sinks follows a trajectory through surface water systems, wherein rivers lead to their transport towards coastal and marine environments. The implications of this migration extend beyond local ecosystems, with the potential for far-reaching ecological repercussions, even to places with little to no industrial activity (Kowalcsyk et al., 2020). Both physiochemical properties of the various PFAS and environmental conditions in different aquatic compartments differ greatly and change the influence of exposure to human and environmental exposure. Therefore, understanding the abundance and behaviour of PFAS within and between the different compartments in the aquatic environment is crucial to understanding the fate of PFAS.

Given the aforementioned complexities and environmental concerns associated with PFAS, the objective of this literature review is to elucidate the fate of these substances within aquatic environments. By synthesising existing knowledge and identifying key research gaps, such an overview can inform future studies and guide strategies for mitigating the adverse impacts of PFAS contamination in aquatic ecosystems. Therefore, this review aims to provide a step-by-step approach to PFAS dynamics in aquatic environments, encompassing their sources, transformation, transport mechanisms, and eventual fate. A comprehensive review of recent literature on the mechanistic behaviour of PFAS compounds was conducted utilising databases such as Scopus, PubMed, and PubChem. Relevant sources and reviews were identified and selected based on criteria encompassing mechanistic insight and relevance to the research objectives.

2. WHAT ARE PFAS?

2.1. DEFINITION OF PFAS

PFAS are a broad group of chemicals for which an exact consensus of the chemical classification is lacking. A general description on the classification of PFAS is given by Buck et al. (Buck et al., 2011) which is used in most research on PFAS. It states that a PFAS comprises a fully fluorinated carbon chain with the $-C_nF_{2n+1}$ moiety, and typically a functional group. More recent literature also adds the $-C_nF_{2n-}$ moiety as PFAS, adding non-terminal fluorinated carbon chains (Buck et al., 2021; OECD, 2018). Other studies regard every molecule with a perfluorinated methyl group ($-CF_3$) or a perfluorinated methylene group ($-CF_2-$; thus, without any H/Br/Cl/I atom attached to the fluorinated carbon atom) as a PFAS, which would also include substances as medicines with only a single fluorinated carbon atom as PFAS, molecules with completely different environmental and human health-related characteristics (Schymanski et al., 2023). The functional group(s) typically exhibit polarity, while the fully fluorinated carbon chains are nonpolar, rendering PFAS molecules amphipathic with surfactant properties. Therefore, most PFAS exhibit notable solubility, unlike typical persistent organic pollutants like lipophilic polycyclic aromatic hydrocarbons. The carbon-fluorine (C-F) bond within PFAS is renowned for its strength in organic chemistry, owing to the high electronegativity of fluorine atoms, thereby conferring exceptional chemical and thermodynamic stability (O'Hagan, 2008). Under ambient conditions, these bonds are recognised for their resistance to degradation in the environment. Because of these properties, PFAS are used as surfactants and surface protection coatings, among other uses further discussed in Chapter 3.1. PFAS are classed into several groups depending on their structure. The main groups of PFAS are perfluorinated compounds, polyfluorinated compounds, and fluoropolymers (Figure 1). These groups will be further discussed separately below.

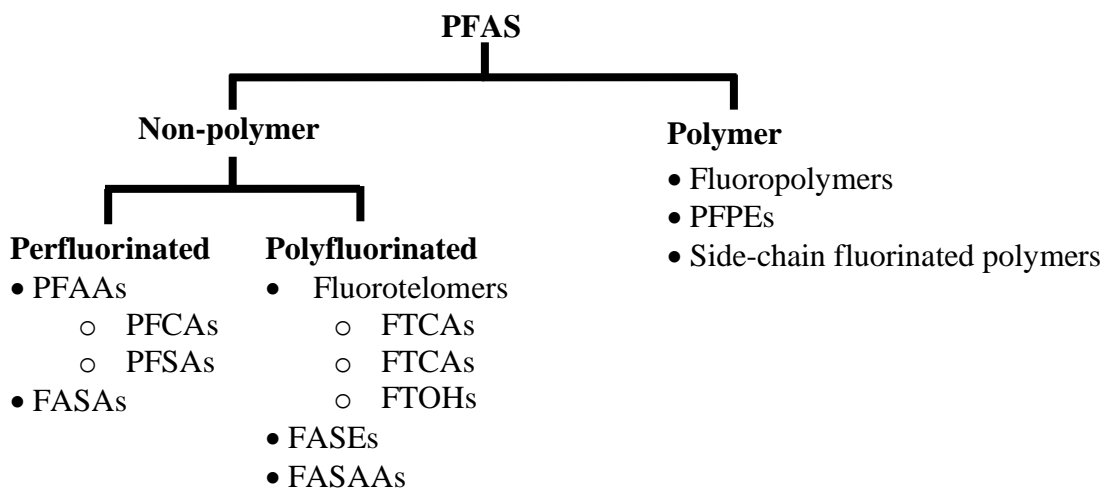


Fig. 1: Comprehensive overview of the classification of several environmentally relevant PFAS

2.2. PERFLUORINATED COMPOUNDS

Perfluorinated compounds are PFAS that feature a polar head group coupled with an alkyl group in which all hydrogen atoms linked to the carbon atoms in the alkyl chain are substituted for fluor atoms (Liu et al., 2019). Notable perfluorinated compounds are the perfluoroalkyl acids (PFAAs), such as the perfluorocarboxylic acids (PFCAs) and perfluorosulfonic acids (PFSAs), categorised by their acidic head groups and containing perfluorinated carbon chain of varying lengths. Examples are perfluorooctanoic acid (PFOA; Table 1) and perfluorooctane sulfonic acid (PFOS; Table 1), containing a carboxylic and sulfonic acid head group and a perfluorinated carbon chain of 8 molecules. The varying carbon chain lengths are termed by their whole carbon chain length, including the carboxylic carbon atom in the nomenclature. Therefore, the PFSAs contain one more -CF₂- moiety than the PFCAs with the same alkyl chain length in their name, adding to their higher molecular weight (Table 1). These chain lengths are further divided into short-chain and long-chain PFAS. PFCAs with a chain length of 4-6 carbon atoms and PFSAs with a chain length of 4-5 carbon atoms are defined as short-chain PFAS, such as perfluorohexanoic acid (PFHxA; Table 1) and perfluorohexane sulfonic acid (PFHxS; Table 1). Long-chain PFAS are n<6 chains as long-chain PFAS (OECD, 2018). The long-chain PFAAs, such as PFOA and PFOS, are generally solid at room temperature, while the long-chain PFAAs are liquid in ambient conditions.

Perfluoroalkane sulfonamides (FASAs) are a group of perfluorinated compounds that are similar to PFCAs and PFSAs but with a sulfonamide head group. They are susceptible to degradation into the other PFAAs. For instance, perfluorooctane sulfonamide (PFOSA) is known to degrade to the highly stable PFOS (Buck et al., 2011; Zhao et al., 2018). Although PFOS and PFOA, as well as PFHxS, their salts, and related compounds have been added to the list of the Stockholm Convention on Persistent Organic Pollutants or are under review to be phased out of production processes, their presence in the environment persists (UNEP, 2019a, 2019b, 2021, 2022).

2.3. POLYFLUORINATED COMPOUNDS

Unlike perfluorinated compounds, polyfluorinated compounds consist of molecules that do not only have a fully fluorinated side chain and functional head group. The polyfluorinated compounds encompass multiple functional groups besides the fluorinated side chain, often incorporating hydrogen or oxygen atoms attached to the carbon. These functional groups make the molecule exhibit anionic, cationic, or zwitterionic properties, resulting in diverse environmental behaviours (Brusseau & Van Glubt, 2019).

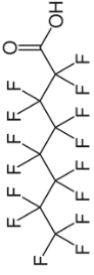
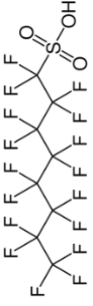
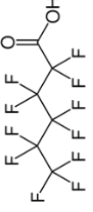
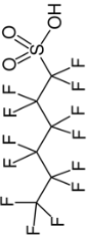
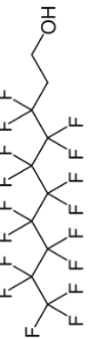

Name	Group	Average Molecular weight (g/mol)	Melting point (°C)	Boiling point (°C)	Molecular structure
PFOA	PFCA	414.1	54-57.9	188-204	
PFOS	PFSA	500.1	53	249	
PFHxA	PFCA	314.1	7.8-14.8	157-236	
PFHxS	PFSA	400.1	41	238.5	
8:2 FTOH	FTOH	464.1	44.8-50	201.3-202	
6:2 FTOH	FTOH	364.1	-16.4-35	90-174	

Table 1: Physiochemical properties of various PFAS and related compounds at ambient conditions. Note; values can differ between methods and experiments, and are therefore indicative. Structural images are in the public domain. All information is obtained from the PFAS Technical and Regulatory Guidance Document (ITRC, 2023).

One of the major groups of polyfluorinated compounds are the fluorotelomers. Fluorotelomers, such as fluorotelomer sulfonic acids (FTSAs; Fig. 2a), fluorotelomer carboxylic acids (FTCAs; Fig. 2b), and fluorotelomer alcohols (FTOHs; Fig. 2c), differ from PFAAs by containing hydrogenated carbon atoms (-CH₂-) between the fluorinated side chain and head group. They are classified by their fluorinated carbon chain, hydrogenated carbon atoms, and functional head group respectively, e.g. 8:2 FTOH (Table 1 & Fig. 2c) for the fluorotelomer alcohol with eight fully fluorinated carbon atoms, two hydrogenated carbon atoms, and an alcohol head group (Buck et al., 2011).

Other polyfluorinated compounds such as fluoroalkane sulfonamidoethanols (FASEs) and fluoroalkane sulfonamidoacetic acids (FASAAs) possess an extra head group compared to perfluoroalkyl compounds (Buck et al., 2011). Both groups are known to degrade to PFAAs in the atmosphere (D'Eon et al., 2006; Mejia Avendaño & Liu, 2015). These substances display greater volatility than PFAAs, facilitating dispersion before degradation into more stable PFAAs within the environment, thereby contributing to the overall environmental burden of PFOS, PFOA, and related compounds as precursor molecules. The non-fluorinated portions of these molecules serve as susceptible points for environmental degradation.

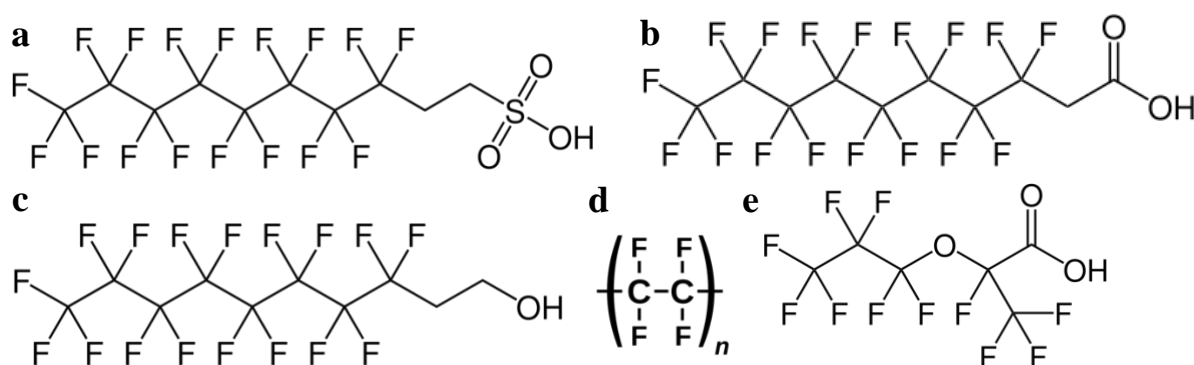


Fig. 2: Chemical structures of 8:2 FTOH (a), 8:2 FTCA (b), 8:2 FCSA (c), PTFE (d), and HFPO-DA (e)

2.4. FLUOROPOLYMERS

Fluoropolymers are polymer structures comprising at least one fully fluorinated carbon chain, exemplified by polytetrafluoroethylene (PTFE; Fig. 2d), commonly known by its trademark name Teflon. Initially produced with PFOA, PTFE production has shifted towards using alternatives such as hexafluoropropylene oxide dimer acid (HFPO-DA; Fig. 2e) with the so-called GenX technology by using its ammonium salt (Beekman et al., 2016). Perfluoropolyethers (PFPEs) consist of fluorinated monomer polymer chains connected via ether linkages, existing as stable liquids employed as lubricants, which can degrade into their constituent perfluorinated monomers under specific conditions (Kwiatkowski et al., 2020). Side-chain fluorinated polymers feature polymer chains with fluorinated side chains attached to functional groups. The weak link

in these molecules lies within the side chain coupling the polymer to the perfluorinated compound, allowing degradation into short-chain perfluorinated compounds, including PFAAs (Schwartz-Narbonne et al., 2023).

2.5. NOMENCLATURE OF IONIC FORMS OF PFAS

PFAS predominantly exist and are measured in their ionic form in the environment e.g. dissociating their hydrogen ion for strong acids such as PFAAs. Yet, they are commonly referenced in literature in their neutral form (Butt et al., 2010; Dixit et al., 2022). Furthermore, they are regularly produced and/or traded as a salt, dissociating their salt group (i.e. sodium or ammonium) when in environmental conditions. As a result, these molecules end up as the same conjugated base in the environment. The abbreviations for PFOS, PFOA etcetera refer to their protonated, non-ionic form. However, when discussing it in an environmental or analytical context at a neutral pH, they are regularly deprotonated (i.e. perfluorooctanoate and perfluorooctane sulfanoate instead of PFOA and PFOS resp.). Therefore, Buck et al. proposed to use this form for both the acid and its conjugated base (Buck et al., 2011). The same is mostly true for the other PFAS, as they are referenced in their neutral form, while regularly being anionic, cationic, or zwitterionic, depending on their pKa, pH, and other factors (Vierke et al., 2013). Buck et al. proposed that when the distinction is explicitly relevant, use the abbreviations for PFAS for both the protonated and anionic form, i.e. PFOA for the acid, PFO for the anion and PFO(A) for both the acid and the anionic form of the PFCA with an eight carbon atom chain and H-PFOS, PFOS and (H-) PFOS for the protonated, anionic and combined forms of the PFSA with eight carbon atom chain for the lack of A from acid in its abbreviation (Buck et al., 2011; Vierke et al., 2013). As this review discusses the PFAS molecules in environmental conditions, the main abbreviation will be used for the environmental isomer of the molecule in this literature review, i.e. PFOS and PFOA for their anionic conjugate.

3. SOURCES OF PFAS

Polyfluoroalkyl and perfluoroalkyl substances are released into the aquatic environment throughout their life cycle, e.g. during the production phase, product use, and after disposal of the product. To understand the presence of PFAS in the aquatic environment, it is important to elucidate the sources of PFAS, and what the differences within the chemical compositions are at those different sources.

3.1. MANUFACTURING METHODS

In general, two different processes have been used to produce PFAS, namely electrochemical fluorination (ECF) and telomerisation. These two methods result in different isomeric compositions. Synthesis through ECF yields even- and odd-numbered, branched, and linear perfluoroalkyl chains, whereas telomerisation produces even-numbered, linear chains resulting

from their different pathways. Nowadays, telomerisation is mainly utilised for the production of PFAS (Buck et al., 2011).

3.1.1. ELECTROCHEMICAL FLUORINATION

ECF is a process used to produce perfluorooctane sulfonyl fluoride, a key compound in the manufacturing of PFOS-related chemicals. This process involves passing an electric current through a solution of anhydrous hydrogen fluoride and octane sulfonyl fluoride. The ECF process replaces hydrogen atoms in the carbon-hydrogen bonds of octane nyl fluoride with fluorine atoms, resulting in a mixture of isomers and homologues, including linear, branched, and cyclic perfluoroalkanes and ethers, as well as other by-products (Wiebe et al., 2018). ECF normally obtains an isomer composition of linear (n-) PFOS at 70-80%, PFOA at 80-85% and PFHxS at ~95%, while the remaining product is mostly branched isomers (Schulz et al., 2020). While ECF is mostly phased out in favour of fluorotelomerisation, it is still used in some factories in China to produce PFOS, PFOA, and their precursors (Chen et al., 2015).

3.1.2. FLUOROTELOMERISATION

While ECF can be used to produce both PFOA, PFOS, and their related precursors, fluorotelomerisation can only be used to produce PFOA and related precursors (Smith et al., 2016). Fluorotelomerisation is a process used to produce oligomers of PFAS molecules. Generally, it involves a reaction of perfluoroethyl iodide with tetrafluorethylene to yield a mixture of even-numbered linear carbon perfluoroalkyl iodides with the structural formula of $C_nF_{2n+1}-I$, where $n=4, 6, 8, 10$, etc. These products react with ethylene to make the new fluorotelomer iodide with the structural formula of $C_nF_{2n+1}CH_2CH_2-I$ where $n=4, 6, 8, 10$, etc. Lastly, these products react through a substitution step of the iodide head group to make fluorotelomer alcohols with the structural formulas of $C_nF_{2n+1}CH_2CH_2-OH$, $n=4, 6, 8, 10$, etc. These three products can be used as the basis for fluorotelomer-based PFAS and polymeric products, such as the fluorotelomer carboxylates and sulfonates, and side chain polymeric fluorotelomers respectively (Buck et al., 2011; Kissa, 2001). The long-chain fluorotelomers are recognised to degrade into PFOA and other long-chain PFCAs, and the production of fluorotelomers is recently switched on short-chain based fluorotelomers (Peshoria et al., 2020).

3.2. APPLICATIONS OF PFAS

3.2.1. FIREFIGHTING FOAMS

Aqueous film-forming foam (AFFF) containing PFAS has been used extensively since its first development (Place & Field, 2012). PFAS-containing AFFF is used in flammable fuel fires in combination with water as fire extinguishing agent. The mixture has surface-tension-lowering properties and spreads rapidly across the surface of hydrocarbon-based fuels, cooling the liquid fuel by forming a water film beneath the foam, resulting in superior firefighting capabilities (McGuire et al., 2014). The use of AFFF has resulted in PFAS contamination of soil,

groundwater, surface waters, and biota worldwide. In particular, PFOS and related PFSAs were used before PFOS was phased out and replaced with other alternatives (Backe et al., 2013; Barzen-Hanson et al., 2017). In addition to the use of PFSAs and primarily after its phasing out, fluorotelomers, specifically FTSAs, are used as alternatives to PFSAs after phasing out PFOS and its precursors (Backe et al., 2013; Place & Field, 2012).

3.2.2. CONSUMER RELATED-PRODUCTS

PFAS are ubiquitously used in consumer products that require non-reactive and stable surfactants or oil and water repellency, i.e. non-stick cookware, stain and water repellent fabrics, food packaging, paper-related products, and personal care products (Glüge et al., 2020). Recently, textile and paper surfaces have been coated with side-chain fluoropolymers, fluorotelomers, and Polyfluoroalkyl phosphoric acid esters as alternatives for long-chain PFAAs. These PFAS have the potential as indirect sources for PFCAs and PFSAs due to transformation (Benskin et al., 2010; Langberg et al., 2021; Thompson et al., 2023).

A study in Norway indicated a paper production facility as the source of contamination to a lake with $\Sigma 29$ PFAS (including PFAAs, perfluoroalkyl sulfonamide derivatives, and FT-based precursors) concentrations in the river sediments near the facility at 2450 mg/kg and the downstream lake sediment with the total PFAS concentrations at 6.1 mg/kg (Langberg et al., 2021). Decreasing concentrations were observed with increasing distance from the site. Another study performed in the United States on PFAS in food-packaging paper found that 46% of the sampled food contact paper contained PFAS and 20% of the paperboard contained PFAS as well, in concentrations ranging from 16 up to 300 ng/cm² (Schaidler et al., 2017). Furthermore, they found that the presence of PFAS was higher in paper food wraps used for their grease-repellent properties than in food containers for liquids or non-food contact papers, showing a clear link between the particular use of the food-packaging paper and the presence of PFAS. The most detected types of PFASs were PFCAs, PFSAs, and fluorotelomer sulfonates. In a different study on toilet paper, 6:2 polyfluoroalkyl phosphoric acid diester was found to be the most abundant PFAS, showing the variety of PFAS used even within a group of products (Thompson et al., 2023). Furthermore, this study also found that the abundance of PFAS, such as polyfluoroalkyl phosphoric acid diesters, was not only found in non-recycled paper but in comparable levels in the more environmentally-friendly perceived recycled paper. This underscores the persistence of various PFAS in the consumer chain.

3.2.3. INDUSTRIAL AND TRANSPORTATION-RELATED PRODUCTS

PFAS are utilised in a broad range of industrial processes. Their non-reactive and stable properties make them suitable e.g. protection against oxidation, high temperatures, and friction resistance. These properties are essential for multiple appliances within the chemical and mechanical industries (Glüge et al., 2020). In a study on the industrial sources of PFAS in Shanghai, China by Chai et al. the researchers found high concentrations of $\Sigma 17$ PFAS at rivers near three industrial sites (Chai et al., 2017). The airport site showed concentrations ranging from 142 to 264 ng/L with the long-chain PFAAs PFOA and two short-chain PFAAs perfluoropentanoic acid (PFPeA) and perfluorobutanoic acid (PFBS) as most prevalent. The concentrations at a site near the fluorochemical plant and metal plating facility ranged from 200 to 2143 ng/L and 211 ng/L to 705 ng/L with PFOA as the most prevalent PFAS. These results showed the link between certain

industries and the ubiquitous levels of PFAS in the surrounding areas. A study in the United States was performed to assess the presence and levels of PFAS in automotive lubricants and hydraulic fluids (Zhu & Kannan, 2020). The researchers measured levels of 13 PFAAs before and after oxidation to account for the potential effects transformation of PFAA precursors could have on the total PFAA composition. They discovered that the Σ PFAA was up to two orders of magnitude higher (from 196 to 8300 $\mu\text{g}/\text{kg}$) after oxidation than before (from 5.96 to 344 $\mu\text{g}/\text{kg}$), suggesting the presence of PFAA precursors in the liquids. Long-chain PFCAs, such as PFOA, accounted for ~70% of the PFAS content before oxidation. Contrastingly, the highest mean molar increase (in pM/g) upon oxidation of the lubricants varied among the PFCAs, with the short-chain PFBA increasing two-fold higher than the second-highest long-chain perfluorodecanoic acid (PFDA; 1350 to 670 for PFBA and PFDA resp.). These results showed the ongoing use of long-chain PFAS in industry, as well as the presence of PFAA precursors leading to short-chain as well as long-chain PFAS.

PFAS usage in products such as AFFF and lubricants causes it to leak directly into the soil and/or surface water, or indirectly through wear of PFAS-coated products such as paper, textiles, and metal plating. With these processes, all different manufactured molecules within the PFAS group in all different compositions leak into the environment. These molecules go through various processes caused by e.g. sorption and transformation.

4. DISTRIBUTION OF PFAS IN THE AQUATIC ENVIRONMENT

4.1. FROM SOIL AND GROUNDWATER TO SURFACE WATER

4.1.1 AFFF EMISSION INTO THE AQUATIC ENVIRONMENT

When AFFF containing PFAS is used, the leachate could be contained on the site itself, but most of it will make its way into the surrounding soil before reaching surface water and groundwater. Multiple studies were performed to assess the levels of different PFAS in the soil and subsequently surface runoff and groundwater. A study by Høisæter et al. at an airport firefighting training facility (FTF) showed that on the site, which was designed with membranes and collection systems for the fuel, water, and AFFF to protect the groundwater, PFAS were still able to leach into the soil due to wind and the increasing spraying range of modern firefighting engines (Høisæter et al., 2019). Furthermore, the extensive use of water during firefighting training facilities resulted in a higher infiltration of the PFAS into the soil. The study showed that PFOS accounted for 96% of the total Σ 12 PFAS found, with a total concentration of PFAS ranging from <0.3 to 6500 $\mu\text{g}/\text{kg}$. The highest concentrations were found in the first two meters of soil, (1000-6500 $\mu\text{g}/\text{kg}$ and 1000-3500 $\mu\text{g}/\text{kg}$, for 1-2 and 2-3 m resp.) and PFAS was found up to 4 m deep. Other studies showed similar results (Baduel et al., 2017; Filipovic et al., 2015; McGuire et al., 2014).

Another study by Dauchy et al. showed concentrations of up to 12112 $\mu\text{g}/\text{kg}$ soil for Σ 34 PFAS (Dauchy et al., 2019). Opposingly to Høisæter et al., 50% to >99% of the PFAS content was contributed to fluorotelomers, mostly 6:2 fluorotelomer sulfonamide alkylbetaine (FTAB) and 6:2 FTSA, and PFSAs contributing to <1 to 46% of the total PFAS content, mostly PFOS. PFAS were found up to 15 m deep into the soil, reaching the groundwater levels. In the groundwater around

airports, levels of PFAS were found up to 45 µg/L (Filipovic et al., 2015; Høisæter et al., 2019). In monitoring wells near an FTF, it was found by Dauchy et al. that close to the expected source site, 6:2 FTAB was quantified in concentrations of 45 to 635 ng/L 20 m below the surface, showing mobility despite being zwitterionic (Dauchy et al., 2019). Further away from the source, PFSAs and PFCAs were more dominant in the groundwater, showing differences between the groundwater close to the source and in the surface soil. Furthermore, for both the PFCAs and PFSAs, the short-chain PFHxS, PFBS, PFPeA, and PFHxA fractions were predominant over PFOS. This indicates that PFOS is less mobile than the short-chain PFAAs and underscores the difference in mobility related to chain length and head groups in soil to groundwater. Furthermore, the PFAS-contaminated groundwater interacts with surface water, exchanging water and subsequent pollutants within these different compartments (Winter et al., 1998).

4.1.2. PFAS TRANSPORT THROUGH WWTP

Wastewater treatment plants (WWTP) are sites that generally filter and purify the water flow from municipal and industrial waste before discharging it into surface water (Barisci & Suri, 2021). Its influent originates from consumers and industry and therefore the PFAS related to these sources flow through the WWTP before entering the aquatic ecosystem. A study on the PFAS levels in 19 Australian WWTPs showed concentrations ranging from 9.3 to 520 ng/L of $\Sigma 21$ PFAS in aqueous samples (Coggan et al., 2019). Furthermore, the levels of both long-chain and short-chain PFCAs, PFPeA, PFHxA, perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), and PFDA, were generally higher in the effluent as compared to the influent, a trend that is seen for most WWTPs worldwide (Hamid & Li, 2016). Additionally, 6:2 FTSA, 8:2 FTSA, and 6:2 chlorinated polyfluoroalkyl ether sulfonate, commercially known as F-53B, were detected at the Australian WWTP influents. F-53B was used as an alternative for PFOS in the metal industry (He et al., 2022).

Furthermore, A study performed in the United States found that the levels of short-chain PFAAs, such as PFBA and PFHxA, increased greatly in 2014 as opposed to 2009 after the reduction of the use of long-chain to short-chain PFAAs (Houtz et al., 2016). WWTP generally purify the water from pollution by e.g. using settling tanks to separate the solids and biodegradation through microbial cultures, but the high aqueous solubility and resistance to (bio)degradation of PFAS prevent this from happening to PFAS (Hamid & Li, 2016). Therefore, it is expected that the water that has been purified from other pollutants would still contain equal concentrations of PFAS in the effluent as in the influent. Furthermore, precursors PFAS are likely degraded and transformed into persistent and stable PFAAs, with a recent higher trend of short-chain PFAAs.

Despite their generally high aqueous solubility, a certain fraction of the PFAS entering WWTPs will sorb to the solid or semi-solid products of WWTP, known as sludge, or referred to as biosolids when used outside of the WWTP e.g. fertilisers (Eggen et al., 2019; Hamid & Li, 2016). Long-chain PFAS are more likely to sorb to the sludge than short-chain due to their hydrophobicity (Eggen et al., 2019). Furthermore, PFSAs are more likely to sorb to the sludge than PFCAs due to their higher hydrophobicity. A study in the Czech Republic measured the levels of $\Sigma 32$ PFAS in

sludge samples from 43 WWTPs. Sewage sludge levels varied from 6 to 960 $\mu\text{g}/\text{kg}$ total PFAS between locations, with PFOS accounting for approximately 60% of the total mass. Furthermore, 20% of the total mass of the samples contained more short-chain than long-chain PFAS, showing the shift to using short-chain PFAS in products. Additionally, HFPO-DA (GenX), the replacement in industry that is used for the manufacture of Teflon, was detected in 7 samples at concentrations ranging from 0.3 to 1.2 $\mu\text{g}/\text{kg}$ (Semerád et al., 2020). These levels were minor as opposed to the total but yet showed an increasing concentration of alternative PFAS. Furthermore, Dauchy et al. found that only long-chain PFCAs, namely PFOA, PFNA, and PFDA, were decreased in the aqueous fraction after floatation treatment which resulted from settling the solids (Dauchy et al., 2017). Therefore, it is likely these long-chain PFCAs deposited into the sludge are linked to the higher hydrophobicity due to more CF_2 -moieties.

A study in France on the presence of PFAS in organic waste products found PFOS, N-ethyl perfluorooctane sulfonamidoacetic acid, and cationic and zwitterionic electrochemical fluorination precursors to PFOS in historic biosolids and composts in the period 1976-1998 (Munoz et al., 2022). Contrastingly, they found that organic waste products in the period 2009-2017 were dominated by zwitterionic fluorotelomers, representing on average 55% of the $\Sigma 160$ PFAS, up to 97%. Specifically, the fluorotelomers sulfonamidopropyl betaines were found with the highest occurrence and prevalence, while it was already found in low concentrations as early as 1985. This study emphasised the recent emergence of zwitterionic and cationic PFAS, as opposed to the anionic conjugated bases of PFAAs. The biosolids and compost that contain PFAS were used as fertilisers in agriculture in the period 1976-1998, returning the PFAS into the environment. Through runoff of the agricultural land to surface water and leaching into the soil and groundwater, these PFAS can re-enter the aquatic environment.

4.2. FROM SURFACE WATER TO COASTAL WATER

4.2.1. SPATIAL AND TEMPORAL TRENDS IN SURFACE WATERS

As described above, PFAS levels in the environment are strongly linked to their source, as levels are higher at industrial sites and WWTPs. Within a region, the differences in levels of PFAS can differ greatly. In Finland, researchers found the levels of $\Sigma 23$ PFAS in riverine waters to be higher in the downstream, urbanised areas (15-75 ng/L) than in the relatively rural north upstream (0.53-0.8 ng/L), with a high burden of PFOS (Junttila et al., 2019). Another study performed in the Ganges River in India showed the highest levels of both PFCAs and PFSAs of $\Sigma 15$ PFAS to be PFHxA (0.4-4.7 ng/L) and PFBS (<Method Quantitation Limit-10.2 ng/L) respectively (Sharma et al., 2016). As opposed to a European country like Finland with a longer history of industrialisation, the more recent industrialised sector of India showed that the transition from long-chain PFAS to short-chain PFAAs affects spatial differences in riverine waters worldwide. Within the Ganges River, the PFOS and PFOA emissions varied along the transect (0.20-190 and

0.03-150 g/d, respectively), and PFOS emissions were linked to urban residents of sub-catchments, further showing spatial differences within a specific region and the link to urban point-sources.

Temporal differences in PFAS concentrations can also be measured in the aquatic environment. A study in Sweden measured the PFAS concentrations at two river catchments in the vicinity of FTFs and AFFFs (Nguyen et al., 2022). They showed that levels of PFAS were 61 times higher for Stockholm Arlanda airport, and 4 times higher at a WWTP and a military airport as opposed to a reference site. Contrastingly, the two catchments showed different temporal PFAS trends. The Stockholm Arlanda airport catchment showed an increase in high water flow seasons, likely contributed to leaching from the airport FTF site, while the WWTP and military airport catchment showed a decrease in high water flow seasons, likely contributed by the dilution of the catchment. This underscores the differences in levels of PFAS linked to the different properties of the source and aquatic environment.

4.2.2. PFAS DISTRIBUTION BETWEEN WATER AND SEDIMENT

Sediment in surface water bodies contains organic carbon, making it a hydrophobic environment relative to the water itself. This causes more hydrophobic PFAS to partition into sediment (Ahmadireskety et al., 2021; Ahrens et al., 2011; Joerss et al., 2019; Osté, 2022; Sharp et al., 2021). The study by Ahrens et al. in laboratory conditions with three sediment types differing in organic content showed that the organic carbon normalised partition coefficient (K_{oc}) decreased in the order of PFOSA ($\log K_{oc} = 4.1 \pm 0.35 \text{ L/kg}$) > PFOS ($3.7 \pm 0.56 \text{ L/kg}$) > PFOA ($2.4 \pm 0.12 \text{ L/kg}$) for all cases (Ahrens et al., 2011). The level of organic content had a significant influence on the partitioning, as well as the difference in head group and length of the three PFAS. With sediment with negligible organic content, the density of the sediment became the most important factor influencing the partitioning, indicating that a higher density (more particles in the same volume) is linked with a higher $\log K_{oc}$, and therefore a higher PFAS content.

Additionally, the study by Li et al. investigated the effect of organic matter, minerals, water saturation, and solution chemistry on the transport and retention of long-chain and short-chain PFAS through modelling (Li et al., 2023). High content of organic matter and minerals, a low saturation, low pH, and divalent cations (such as Ca^{2+} and Mg^{2+}) had a great retardation effect on long-chain PFAS. This effect was mainly due to their hydrophobic interactions, while the effect of electrostatic interactions was more relevant for short-chain PFAS. Overall, the sorption can cause retardation of PFAS in surface waters, with a stronger effect on long-chain PFAS and relatively hydrophilic head groups.

4.2.3. PFAS DISTRIBUTION BETWEEN WATER, AIR AND NAPL

PFAS have a polar, hydrophilic head group and an apolar, hydrophobic tail, causing them to have surfactant properties. Consequently, they behave in a similar manner as other surfactants, such as soaps, and are manufactured for this purpose. Therefore, PFAS will accumulate at

interfaces such as the air-water and non-aqueous phase liquid-water interface. Nonaqueous phase liquids (NAPL) are organic liquids characterised by their immiscibility with water, such as (non-fluorinated) hydrocarbons. These interfaces provide an extra retention layer for PFAS as they move at a lower velocity than the bulk aqueous mass (Brusseau, 2018). Little is known in the literature about the concentration difference between the surface layer of a waterbody and the other depths of the same location, but information on the air-water and air-NAPL interfaces is available on the vadose zone, between surface and groundwater. Previous studies showed that accumulation at the air-water interface could significantly enhance the retention of PFAS in the unsaturated zone (Schaefer et al., 2023; Silva et al., 2022).

Further research suggested the link between retention in the air-water and air-NAPL interfacial adsorption to the chain length of PFCAs, suggesting the greater surface activity of long-chain PFAS (Silva et al., 2019). Furthermore, a study on the removal of PFCAs from aquatic environment reported that the aeration of PFAS-contaminated water increased sorption to activated carbon as a result of air-water interface adsorption, particularly on long-chain PFAS (Meng et al., 2019). Contrastingly, the accumulation of PFAS at air-liquid, air-NAPL, and air-sediment interfaces suggests that short-chain PFAS would partition more into the aqueous phase and, therefore are more mobile and get distributed more easily in the aquatic environment.

4.3. OCEANIC ENVIRONMENT

Most, if not all surface water bodies drain into the sea, where they flow through coastal areas into the vast seas and oceans. Once in the oceans and seas, PFAS will accumulate because these waters do not run off into other water bodies and therefore act as the final sink. Junttila et al. reported higher concentrations of $\Sigma 23$ PFAS in the surface waters of Finland than in the outflow areas in the Baltic Sea, but these levels were within the same order of magnitude (Junttila et al., 2019). Variations exist within levels of PFAS in the different seas and oceans, but the general levels are in the same order of magnitude as well (Muir & Miaz, 2021). Additionally, Miaz and Muir found that the number of measurements of PFAS in coastal waters and seas that are present does not translate into broad geographic coverage worldwide.

4.3.1. SPATIAL AND TEMPORAL TRENDS IN OCEANIC WATERS

PFAS concentrations in open seas and oceans are mostly available for Western Europe and East Asia (85% for PFSAs and 80% for PFCAs). Data on the PFAS concentrations for the coastal seas of North America, South Asia, and Australia are limited, and the data for the coastal seas of Africa and South America are lacking (Muir & Miaz, 2021). Furthermore, the data predominantly concern 14 PFCAs and PFSAs (including FOSA) to assess broad spatial and temporal trends, lacking adequate measurements for the replacement PFAS. Differences in spatial trend were estimated, but all median concentrations of $\Sigma C4-C6$ PFCAs, $\Sigma C7-C12$ PFCAs, and Σ PFCAs were within two orders of magnitude (0-14 ng/L, 0-12 ng/L and 0-4.9 ng/L resp.). showing the ubiquitous presence of PFAS in oceanic waters.

Temporal differences can be seen for different coastal seas in East Asia (Bohay and the Yellow Seas, East China Sea, and South China Sea) for the $\Sigma 14$ analysed PFAS (Muir & Miaz, 2021). The increases in PFAS in coastal East Asia from 2000-2009 to 2015-2019 were especially large for the short-chain PFCAs PFBA and PFPeA (56-1560x increase), as well as for PFHxA. Unfortunately, these differences could be due to changes in reporting limits rather than purely accounted for by changes in emission. Lower emissions were estimated in the North Sea in 2015-2019 than in 2000-2009 for most PFCAs as well as for PFOS, but within a tenfold decrease. Furthermore, the median concentrations for short-chain PFSAs PFBS and PFHxS were higher in the Baltic Sea in the same periods (31x and 25x increase). Overall, the levels of legacy PFAS as PFOA and PFOS seem to be within range of each other, while some short-chain PFAS seem to be more abundant in contemporary measurements.

4.3.2. EFFECT OF SALINITY

Behaviour of PFAS in oceanic waters is less studied. Zhang et al. found an inverse correlation between salinity and $\Sigma 21$ PFAS in the North-Eastern coastal waters of the United States (Zhang et al., 2019). Lower salinity in seawater is due to the higher content of surface water outflow. Therefore, this correlation is likely due to the dilution of PFAS-contaminated surface and coastal water, rather than the mechanistic link between salinity and PFAS levels. Generally, salinity can affect the sorption of PFAS in aquatic environments, but as oceanic salt levels do not differ greatly on a local scale, the effect in oceanic water is to be negligible for the fate of PFAS (You et al., 2010).

4.3.3. OCEAN-AIR PARTITIONING THROUGH SEA SPRAY AEROSOLS

Due to the volatility of certain PFAS and/or their behaviour to accumulate at the air-liquid interface, certain PFAS can re-enter the surface water environment through sea spray aerosols (SSA). A laboratory simulation of SSA reported that the water-to-air transport of C6-C14 PFCAs and C6, C8, and C10 PFSAs transferred to the atmosphere at less than 1%, but through sprays, the transfer rate would increase by up to 1360 times (Reth et al., 2011). Furthermore, the enhancement of volatilisation through the spray was correlated to the longer chain length of PFAAs for all except the longest measured C14 PFCA. Sha et al. reported a strong correlation of atmospheric PFAAs to the presence of SSA by measuring the Na^+ as the tracer ion at two locations at the Norwegian coast (Sha et al., 2022). Furthermore, the location closer to the coast showed a higher correlation between SSA and the PFAAs than more inland, emphasising the effect of PFAS-polluted seawater on PFAS levels in coastal areas and their potential return to surface water. No research is available on the presence of more volatile PFAS, e.g. fluorotelomer alcohols, which could potentially lead to higher levels of PFAS in SSA.

5. CONCLUSION AND RECOMMENDATIONS

This review emphasises the need to assess the step-by-step pathway of the fate of PFAS in aquatic environments. In doing so, several key points have been identified. The production of PFAS causes a wide array of structurally different PFAS to be emitted into the environment, ranging from well-known legacy PFAS such as the long-chain PFAAs to recent alternatives such as short-chain PFAAs, fluorotelomers, and fluoropolymers. The diversity of molecular compositions and subsequent interactions of PFAS with the environment causes them to accumulate differently in the aquatic environment. Throughout the chain, most PFAS such as fluorotelomers and fluoropolymers end up transforming into terminal PFAAs, adding up to the total of PFAAs in the environment. This study has produced an overview of the PFAS chain from source to sink using recent literature, providing a complement to previous reviews that primarily focused on a small group of PFAS, a single compartment in the aquatic environment, or specific interactions of PFAS and their environment.

Individual studies and governmental reports on PFAS predominantly focus on measuring a limited subset of 10-30 well-known PFAS compounds, leaving at least 4000 other PFAS understudied. Emphasis is often placed on PFAAs, with lesser attention given to fluorotelomers and specific PFAS variants (such as PAPs and fluoropolymers). Additionally, natural processes, such as oxidation, are sometimes simulated, although these simulations may not accurately represent real-world scenarios. Greater attention should be directed towards detecting other PFAS variants that may currently evade analysis.

PFAS compounds are often grouped together across all compartments, despite clear evidence indicating that the composition of the chain, functional groups, and other structural features significantly influence their behaviour in the environment. Measurements should focus more on interfaces rather than the total concentrations in compartments such as water and sediment.

With the increasing volume of information on PFAS, it has become challenging to consolidate all sources. The diversity of methods and environmental factors complicates the precise identification of discussed PFAS compounds. Often, generalised forms are referred to, whereas transformed, branched, or ionised isomers are more likely present. Lacking the specific structure and state of the molecule can lead to over- and underestimations of PFAS concentrations.

Overall, there is a growing interest in various aspects of PFAS behaviour in the environment within contemporary scientific discourse. However, insufficient attention has been paid to the diversity of PFAS compounds. In the future, greater consideration should be given to understudied PFAS variants and the partitioning processes associated with these differences.

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