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Per- and polyfluoroalkyl substances (PFAS) in the marine environment: An overview and prospects☆

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are synthetic chemicals known for their excellent persistence and surface activity. PFAS can be readily transported through waterways into the ocean, which serves as their primary global sink. This review summarizes recent studies on PFAS in the marine environment and outlines some key directions for future research. Certain PFAS may pose significant risks to human health through seafood consumption. Furthermore, marine predators such as cetaceans may be at high risk of PFAS exposure due to the biomagnification of PFAS in marine food webs. The ban on legacy PFAS has led to a dramatic increase in detecting various emerging PFAS. However, potential risks associated with these emerging PFAS remain largely underexplored. This review highlights critical research needs: tracing ultra-short-chain PFAS in the marine environment, establishing environmental quality standards to protect marine ecosystems and human health, and investigating toxicological effects and mechanisms of emerging PFAS.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a class of synthetic chemicals. Owing to their high C−F bonding strength (485 kJ/mol), PFAS are incredibly stable (Buck et al., 2011). With hydrophobic and lipophobic function groups, PFAS have excellent surfactant properties. The robust stability and high surface activity give PFAS unique characteristics for application in a wide array of products and processes, including electroplating, textile, and paper sectors for 70 years (Evich et al., 2022). Nowadays, the persistence and toxicity of PFAS have become a primary concern for society. PFAS have attracted much attention since the early 2000s, when the ubiquity and potential risk of two most famous PFAS (i.e., legacy PFAS), perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS), were reported (Giesy and Kannan, 2001). Early communications used multiple terms such as "per- and polyfluorinated chemicals," "perfluorinated organics," "perfluorochemical surfactants," and "highly fluorinated compounds." In 2011, Buck et al. published a milestone paper, providing the first clear structural definition of PFAS: "*PFAS are aliphatic substances containing one or more C atoms on which all the H substituents present in the non-fluorinated analogues from which they are notionally derived have been replaced by F atoms, in such a manner that PFAS contain the perfluoroalkyl moiety* C_nF_{2n+1}" (Buck et al., 2011).

In 2018, the Organisation for Economic Co-operation and Development (OECD) and the United Nations Environment Programme (UNEP) published a list of over 4700 PFAS that were known or likely to have been on the global market (OECD, 2018). The list includes substances that contain fully fluorinated carbon moieties but do not meet the PFAS definition in Buck et al. (2011) due to a large portion lacking a perfluorinated methyl group ($-CF_3$) in the molecule. In recent years, the advancement of nontarget analytical techniques has enabled the identification of many unknown PFAS in environmental and product samples. These developments motivated the reconciliation of the terminology of the PFAS universe, including a renewed look at the definition of PFAS. In 2021, the OECD presented a revised, broadly inclusive definition: "*PFAS are defined as fluorinated substances that contain*

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at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it), i.e., with a few noted exceptions, any chemical with at least a $-CF_3$ or a perfluorinated methylene group ($-CF_2-$) is a PFAS." This is the latest definition for PFAS, which is easily implementable to distinguish between PFAS and non-PFAS and understandable to experts and nonexperts alike (Wang et al., 2021b).

A series of control measures have been taken around the world to reduce the ecological and human exposure risks posed by PFAS. In 2000, 3M voluntarily took a proactive step to phase out the production of PFOS and its related compounds. In 2006, under the 2010/2015 PFOA Stewardship Program, eight major fluoropolymer and telomer manufacturers, including Arkema, Asahi, Ciba, Clariant, Daikin, DuPont, 3M/Dyneon, and Solvay Solexis, eliminated the production and use of PFOA and its related compounds (USEPA, 2006). PFOS and its related compounds have been listed under Annex B of the Stockholm Convention for global restriction since 2009 (UNEP, 2009). In 2012, some long-chain PFCAs (C11–C14), including perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA), and perfluorotetradecanoic acid (PFTeDA), which were identified as very persistent and very bioaccumulative chemicals, were included in the Candidate List of Substances of Very High Concern (SVHC) under the European Research, Education, and Action for Community Health (REACH) regulations (ECHA, 2013). Subsequently, in 2013, PFOA and ammonium perfluorooctanoate (APFO) were included in the Candidate List of SVHC under the REACH 2013 regulation (ECHA, 2013). In 2017, perfluorohexanoic acid (PFHxA) and perfluorodecanoic acid (PFDA), together with their related compounds, were identified as potentially hazardous chemicals and were added to the list of SVHC under the REACH regulations (ECHA, 2017). In 2019, PFOA and its related compounds were listed under Annex A of the Stockholm Convention for global elimination (UNEP, 2019). In 2020, perfluorobutane sulfonic acid (PFBS) and its salts as well as hexafluoropropylene oxide dimer acid (HFPO-DA) and its salts and acyl halides were added to the Candidate List of SVHCs in REACH because their levels of persistent, mobile, and toxic properties are equivalent to those of legacy PFAS (ECHA, 2019a, 2019b). In 2021, the European Union's restriction on C9–C14 PFCAs, their salts, and related substances was published in the Official Journal as Regulation (EU) No 2021/12971, which replaces Annex XVII, entry 68 in the REACH Regulation (EU, 2021). In 2022, perfluorohexane sulfonic acid (PFHxS) and its related compounds were listed under Annex A of the Stockholm Convention for global elimination (UNEP, 2022a). In the same year, it was also proposed that long-chain perfluorocarboxylic acids (PFCAs) with perfluorinated carbon chain lengths from 8 to 20 should be listed in the Stockholm Convention (UNEP, 2022b).

Because of the increasingly stringent regulations and initiative actions, the production and emission of long-chain PFAS (such as PFOA and PFOS) decreased, and the production was gradually replaced by short-chain homologues and functionalized perfluoropolyethers (Scheringer et al., 2014; Wang et al., 2013). The increasing levels and equivalent effects of alternatives to legacy PFAS warrant more attention. To date, several novel chemicals (e.g., HFPO-DA) have been used as alternatives. In addition, many PFAS alternatives such as chlorinated polyfluoroalkyl ether sulfonic acids (Cl-PFESAs) and perfluoroethylcyclohexane sulfonic acid (PFECHS), which have been designed and manufactured over decades, have just been identified and studied in environmental research in recent years and are regarded as "emerging PFAS" due to the increasing worldwide demands for their application in commercial products (Munoz et al., 2019; Nakayama et al., 2019). They have similar chemical structures and physicochemical properties as those of PFOS and PFOA and, therefore, likely have similar environmental fates and toxicities (Gomis et al., 2015; Wang et al., 2024). Fig. 1 shows the structures of representative legacy, emerging, and OECD-defined PFAS.

Based on their functional groups and physicochemical properties, PFAS can be divided into ionic PFAS (i-PFAS) and neutral PFAS (n-PFAS), and these two categories exhibit different environmental fates (Lindstrom et al., 2011). Perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkyl sulfonic acids (PFSAs) are the most common i-PFAS. Owing to their hydrophilic head groups (such as carboxylate and sulfonate groups), i-PFAS have relatively high air–water partition coefficients (K_{aw}) and *n*-octanol–water partition coefficients (K_{ow}) and are more likely to be found in the aquatic environment rather than in the atmosphere (Wang et al., 2011). Log K_{ow} (3–7) of i-PFAS increase with the carbon chain length (Higgins and Luthy, 2006). Therefore, i-PFAS, especially long-chain i-PFAS (C > 7), are likely to be absorbed onto the solid phase, such as suspended particulate matter, airborne particulate matter (APM), soil, and sediment in the environment (Ahrens, 2011). For n-PFAS, fluorotelomer alcohols (FTOHs), perfluoroalkyl sulfonamides (FASAs), and perfluoroalkyl sulfonamide ethanols (FASEs) are the most studied in the environment (Wang et al., 2020; Wang et al., 2011; Xie and Ebinghaus, 2011). Compared to i-PFAS, n-PFAS are more volatile and mainly distributed in the gaseous phase (Shoeib et al., 2008; Wang et al., 2020). Meanwhile, n-PFAS are not as stable as i-PFAS in the environment, and some n-PFAS (e.g., 8:2 FTOH) can be transformed into certain i-PFAS [e.g., perfluorononanoic acid (PFNA) and PFOA] via photochemical and biological processes in the atmospheric and terrestrial environments (Butt et al., 2014; Liu et al., 2007). As a result, most studies about PFAS in the marine environment have focused on i-PFAS. Table 1 shows the abbreviation, full name, formula, and estimated log K_{ow} of common i-PFAS.

The marine environment covers 71% of the Earth's surface and exerts substantial influence on critical biogeochemical cycles, climate regulation, and biodiversity conservation. Today, PFAS are widely detected in the abiotic and biotic matrices (Evich et al., 2022). PFAS can enter the environment via many pathways, such as air emission, wastewater, and leachate. In particular, PFAS can be readily transported in aquatic systems due to their relatively high solubility and polarity, and thus the ocean is considered as one of their final global sinks (Evich et al., 2022; Wang et al., 2023b). Long-range air transport can be

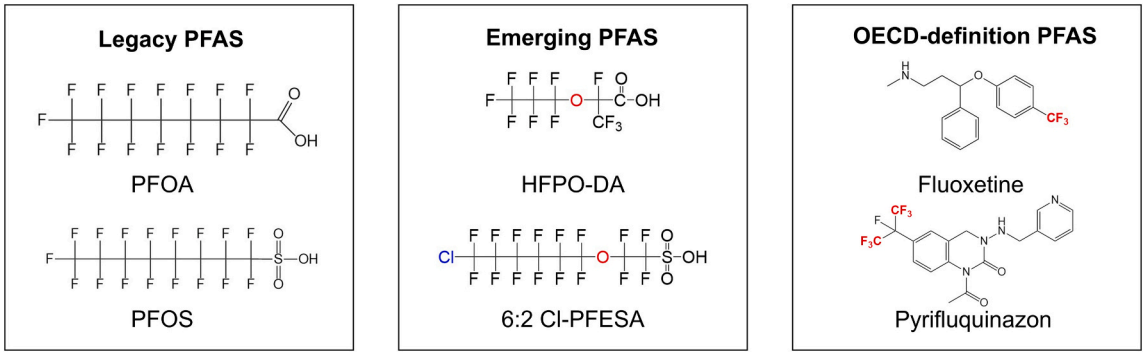


Fig. 1. Structure of typical legacy, emerging, and OECD-defined PFAS.

Table 1
Formula, and estimated log K_{ow} of common i-PFAS.

Abbreviation	Full name	Formula	Log K_{ow} [a]	Log K_{ow} [b]
PFBA	Perfluorobutanoic acid	$C_3F_7CO_2H$	2.14	2.82
PFPeA	Perfluoropentanoic acid	$C_4F_9CO_2H$	2.81	3.43
PFHxA	Perfluorohexanoic acid	$C_5F_{11}CO_2H$	3.48	4.06
PFHpA	Perfluoroheptanoic acid	$C_6F_{13}CO_2H$	4.14	4.67
PFOA	Perfluorooctanoic acid	$C_7F_{15}CO_2H$	4.81	5.30
PFNA	Perfluorononanoic acid	$C_8F_{17}CO_2H$	5.48	5.92
PFDA	Perfluorodecanoic acid	$C_9F_{19}CO_2H$	6.15	6.50
PFUnDA	Perfluoroundecanoic acid	$C_{10}F_{21}CO_2H$	6.82	7.15
PFDoDA	Perfluorododecanoic acid	$C_{11}F_{23}CO_2H$	7.49	7.77
PFTeDA	Perfluorotetradecanoic acid	$C_{13}F_{27}CO_2H$	8.16	8.16
PFHxDA	Perfluorohexadecanoic acid	$C_{15}F_{31}CO_2H$	8.83	8.83
PFOcDA	Perfluorooctadecanoic acid	$C_{17}F_{35}CO_2H$	9.50	9.50
PFBS	Perfluorobutane sulfonic acid	$C_4F_9SO_3H$	1.82	3.90
PFPeS	Perfluoropentane sulfonic acid	$C_5F_{11}SO_3H$	2.49	/
PFHxS	Perfluorohexane sulfonic acid	$C_6F_{13}SO_3H$	3.16	5.17
PFHpS	Perfluoroheptane sulfonic acid	$C_7F_{15}SO_3H$	3.82	/
PFOS	Perfluorooctane sulfonic acid	$C_8F_{17}SO_3H$	4.49	6.43
PFNS	Perfluorononane sulfonic acid	$C_9F_{19}SO_3H$	5.16	/
PFDS	Perfluorodecane sulfonic acid	$C_{10}F_{21}SO_3H$	5.83	/
HFPO-DA	Hexafluoropropylene oxide dimer acid	$C_6HF_{11}O_3$	3.36	6.60
6:2 Cl-PFESA	6:2 Chlorinated polyfluorinated ether sulfonic acid	$C_8HClF_{16}O_4S$	5.24	7.03
8:2 Cl-PFESA	8:2 Chlorinated polyfluorinated ether sulfonic acid	$C_{10}HClF_{20}O_4S$	6.58	/
ADONA	4,8-Dioxa-3H-perfluorononanoic acid	$C_7H_2F_{12}O_4$	3.57	4.85
PFECHS	Perfluoroethylcyclohexane sulfonic acid	$C_8HF_{15}O_3S$	4.42	/
Cl-PFOS	Chlorinated perfluorooctane sulfonic acid	$C_8HClF_{16}O_3S$	4.81	/
6:2H-PFESA	6:2 Hydrogen-substituted polyfluorinated ether sulfonic acid	$C_8H_2F_{16}O_4S$	4.32	/
8:2H-PFESA	8:2 Hydrogen-substituted polyfluorinated ether sulfonic acid	$C_{10}H_2ClF_{20}O_4S$	5.66	/
4:2 FTS	4:2 Fluorotelomer sulfonic acid	$C_6H_5F_9O_3S$	1.99	3.21
6:2 FTS	6:2 Fluorotelomer sulfonic acid	$C_8H_5F_{11}O_3S$	2.66	4.44
8:2 FTS	8:2 Fluorotelomer sulfonic acid	$C_{10}H_5F_{13}O_3S$	3.33	5.66
10:2 FTS	10:2 Fluorotelomer sulfonic acid	$C_{12}H_5F_{15}O_3S$	4.00	6.91
FPrPA	3:3 Fluorotelomer carboxylic acid	$C_6H_5F_7O_2$	3.12	/
FPePA	5:3 Fluorotelomer carboxylic acid	$C_8H_5F_9O_2$	4.46	/
FHpPA	7:3 Fluorotelomer carboxylic acid	$C_{10}H_5F_{11}O_2$	5.80	/
6:2-diPAP	6:2 Fluorotelomer phosphate diesters	$C_{16}H_9F_{26}O_4P$	9.42	8.41
6:2/8:2-di PAP	6:2/8:2 Fluorotelomer phosphate diesters	$C_{18}H_9F_{30}O_4P$	10.76	10.15
8:2 di-PAP	8:2 Fluorotelomer phosphate diesters	$C_{20}H_9F_{34}O_4P$	12.10	10.93
FOSAA	Perfluorooctane sulfonamido acetic acid	$C_{10}H_4F_{17}NO_4S$	5.52	/
N-EtFOSAA	*N*-ethyl perfluorooctane sulfonamido acetic acid	$C_{12}H_8F_{17}O_4NS$	6.22	/
N-MeFOSAA	*N*-methyl perfluorooctane sulfonamido acetic acid	$C_{11}H_6F_{17}O_4NS$	5.73	/
6:6 PFPi	6:6 Perfluorophosphinic acid	$C_{12}HF_{26}O_2P$	6.96	/
6:8 PFPi	6:8 Perfluorophosphinic acid	$C_{14}HF_{30}O_2P$	8.30	/
8:8 PFPi	8:8 Perfluorophosphinic acid	$C_{16}HF_{34}O_2P$	9.64	/

[a] Estimated using the EPISuite (USEPA, 2012).
[b] Estimated using the COSMOtherm model (Gomis et al., 2015; Wang et al., 2011).

another possible source for PFAS in the marine environment (Wang et al., 2020). Due to bioaccumulation potential, PFAS can affect marine organisms when their tissue concentrations reach effect thresholds. Understanding PFAS dynamics within marine ecosystems and their effect thresholds to marine life are fundamental for developing strategies to minimize their risks and preserve essential ecosystem functions across varied spatial and temporal scales. Additionally, seafood, a staple food consumed by billions of people worldwide, represents one of the last remaining wild food sources, with marine aquaculture anticipated to experience significant growth in the coming decade (Nations, 2020). This review provides a timely overview of PFAS in the marine environment and outlines future research directions for investigation. Our prime objective is to contribute to the development of effective strategies for improving marine ecosystem management practices in light of the rapidly increasing pollution risks associated with PFAS and their emerging alternatives.

2. PFAS in marine matrices

PFAS can be readily transported via aquatic systems, and oceans are believed to be one of their final global sinks (Nakayama et al., 2019; Prevedouros et al., 2006; Wang et al., 2023b). Industrial wastewater, landfill leachate, and effluents discharged from wastewater treatment plants are essential sources of coastal water pollutants. Therefore, PFAS can be emitted directly into runoffs and transported to the marine environment (Ahrens, 2011; Zhao et al., 2017). Meanwhile, the precursors of PFAS can be degraded during long-range atmospheric transport, which is considered an indirect source of PFAS in the coastal environment (Yamashita et al., 2008). Urbanized coastal environments are susceptible to and highly affected by various pollutants, including PFAS (Habibullah-Al-Mamun et al., 2016; Zhao et al., 2013). Ahrens et al. (2010) analyzed PFAS in northern Europe and the Atlantic Ocean, and found C8 PFAS (i.e., PFOA and PFOS) were the primary pollutants, with average concentrations of 223 pg/L and 232 pg/L, respectively . Kwok et al. (2015) analyzed 23 PFAS in the South China Sea (SCS) and found PFOA was the predominant pollutant, with the total PFAS (ΣPFAS) concentrations in seawater from the SCS ranging from 95 pg/L to 4925 pg/L. They also found a significant positive correlation between PFAS concentrations and gross domestic product, indicating that human activities had a driving effect on the occurrence of PFAS in the surrounding environment. Zheng et al. (2017) analyzed 17 PFAS in the SCS and East China Sea (ECS) and found that the range of PFAS concentrations in the ECS and the SCS was 181−2658 pg/L and 62−494 pg/L, respectively, while noting that river flow and ocean current played a significant role in the distribution of PFAS in the ECS. Wang et al. (2019) analyzed 20 PFAS, including five novel PFAS alternatives, in the Pearl River Estuary (PRE) and SC, and found that the range of PFAS concentrations in the surface and bottom seawaters was 125−1015 pg/L and 38−779 pg/L, respectively, and 6:2 and 8:2 Cl-PFESAs (two emerging PFAS) were widely detected. Hu et al. (2021) investigated PFAS, including *p*-perfluorous nonenoxybenzenesulfonate (OBS) and legacy perfluoroalkyl acid (PFAA) isomers in the seawater from the ECS, and found PFOA and PFOS were the predominant PFAS; they detected OBS in 46% of seawater samples from the ECS with concentrations up to 5.2 pg/L, and the spatial distribution trend suggested the riverine input was a vital source of PFAAs and 6:2 Cl-PFESA in this sampling area. Joerss et al. (2019) investigated 29 PFAS in the surface water from the coastal Baltic Sea and German Bight in the North Sea, reporting total PFAS concentrations ranging from 1600 to 5200 pg/L and 4700 to 7400 pg/L, respectively. Notably, the level of HFPO-DA in the German Bight was higher than PFOA, potentially influenced by the point source and the

exchange with ocean current as indicated by the positive correlation between salinity and HFPO-DA concentrations. They also reported wide detection of PFECHS in the Baltic Sea. Lee et al. (2020) analyzed 25 PFAS along the Korean coast and found PFOA and PFOS were the predominant contaminants, with average concentrations of 2170 pg/L and 289 pg/L, respectively. Shan et al. (2021) analyzed 23 PFAS in the surface seawaters collected in the northwestern Pacific Ocean (NW-PO), eastern Indian Ocean (E-IO) and Southern Ocean (SO), with NW-PO exhibiting the highest ΣPFAS concentration ranging from 413 pg/L to 1380 pg/L. The declining trends of 6:2 Cl-PFESA from nearshore to offshore suggested inland sources from China and Australia, highlighting the risk of its global transportation. Han et al. (2022) analyzed 23 PFAS in the seawater of the Indian Ocean (IO), Northwest Pacific Ocean (NWPO), and a joining area of Asia and the Indian-Pacific Ocean (JAIPO). They found that PFOA and PFHpA were two predominant PFAS, with average concentrations ranging from 9.18 to 108 pg/L and 10.3 to 27.4 pg/L; they suggested that the Indonesian throughflow through JAIPO contributed to the PFOA level in the IO. Zhu et al. (2024) analyzed 38 PFAS including perfluorobutane sulfonamide (PFBSA) in the East China Sea and Antarctic Ocean and discovered that PFBSA levels with the average level of 101 pg/L were comparable to perfluorobutanoic acid (PFBA) and PFOA levels with average levels of 147 pg/L and 105 pg/L, respectively, in the Antarctic Ocean; possible sources to the high concentrations included the wastewater discharge from the research stations, the comparatively high hydrophilicity of PFBSA, and the degradation from precursors. A summary of reported concentrations for typical PFAS, specifically PFBA, PFOA, HFPO-DA, PFBS, PFOS, and 6:2 Cl-PFESA, is presented in Fig. 2A, with a comparison of their concentrations shown in Fig. 2B.

PFAS can be deposited in water through hydrologic and adsorption processes and then migrate to the sediment, which can be the source of PFAS in the sediment (Wang et al., 2025 and Eschauzier et al., 2010). Yin et al. (2022) reported that the pseudo-second-order kinetic model can well describe the sorption of PFAS onto sediment, where compound-specific and solution-specific parameters influence PFAS sorption; the sediment−water distribution coefficient (K_d) of PFAS increased with the increase in perfluorocarbon chain length and salinity, suggesting that hydrophobic and electrostatic interactions were involved in the adsorption process. Moreover, the increase in perfluorocarbon chain length and salinity decreased the desorption rate of PFAS from sediment, and PFCAs were desorbed more readily than PFSAs with the same perfluorocarbon chain length in all salinity groups. Ahrens et al. (2009)

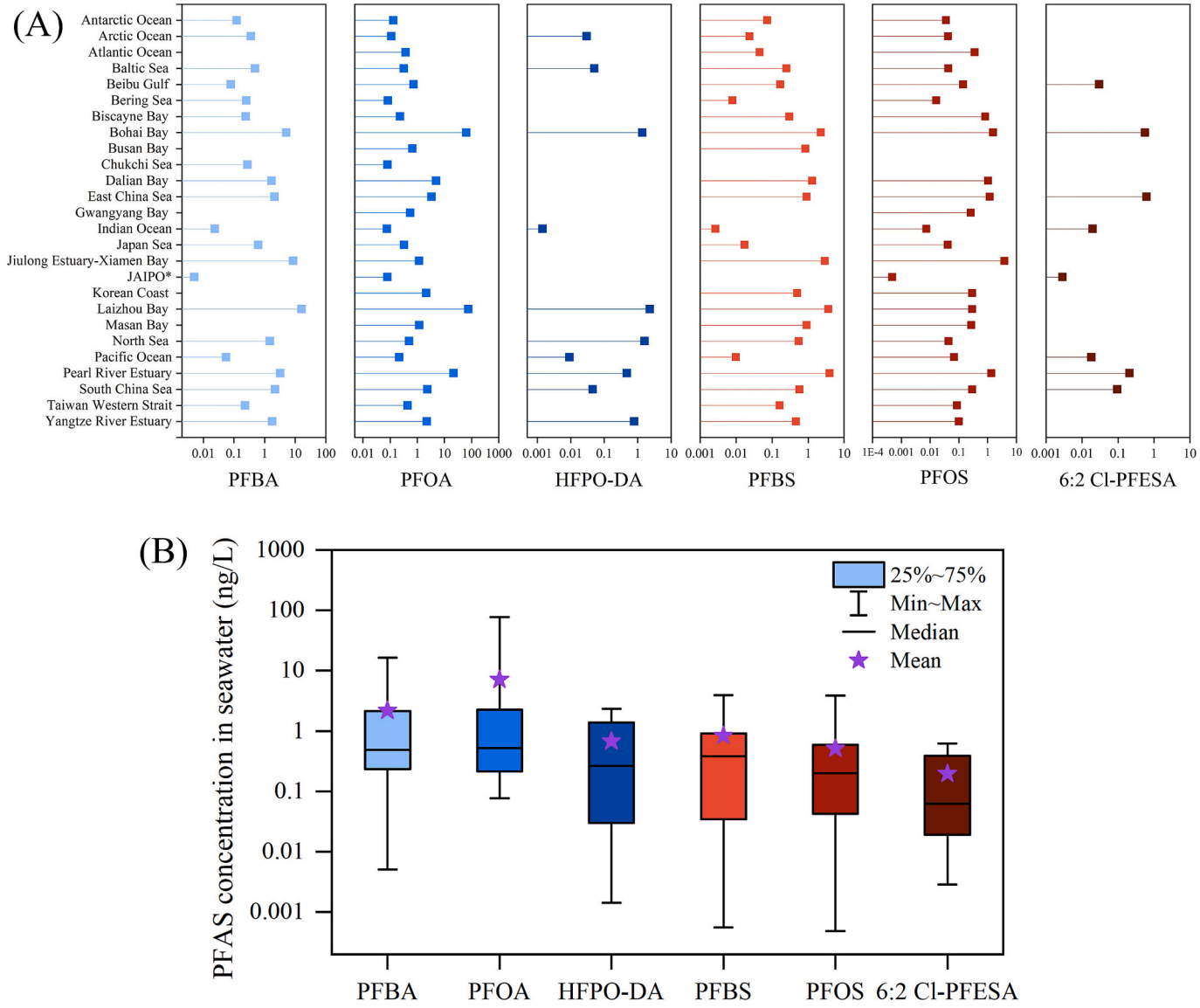


Fig. 2. (A) Reported concentrations of PFAS in seawater from and (B) comparison of different PFAS concentrations in seawater. *JAIPO represent joining area of Asia and the Indian-Pacific Ocean.

analyzed PFAS in sediment core and pore water from Tokyo Bay, and found PFAS concentrations increased from 1985 to 2001; however, they decreased significantly after 2001, possibly due to 3M's phasing out of PFOS in 2000; short-chain PFCAs (C < 8) were only detected in pore water, and long-chain (C ≥ 11) PFCAs were only detected in sediments. They reported that compared to functional groups, the perfluorocarbon chain's length had a larger impact on PFAS's environmental distribution. Long-chain PFAS were more likely to be distributed into the sediment, and short-chain PFAS were more likely to be present in the water column (Higgins and Luthy, 2006). Five novel PFAS in the sediment from the PRE and the SCS were analyzed in 2019, where ΣPFAS concentrations ranged from 7.5 to 84.2 pg/g dry weight (dw), and the occurrence of some novel PFAS was for the first time reported in the SCS; comparable concentrations were found for 6:2 Cl-PFESA and PFOS, indicating the wide use of novel PFAS in the SCS, but these novel PFAS posed a low ecological risk (Wang et al., 2019). Hu et al. (2021) reported the occurrence and partitioning behaviors of legacy PFAS isomers and emerging PFAS in the sediment from the ECS, where branched PFOA and PFOS isomers were consistently much less frequently detected in sediments than in seawaters; the mean log-transformed organic carbon-normalized sediment–water partition coefficients (log K_{oc}) of 6:2 Cl-PFESA (2.6 ± 0.36) and OBS (2.7 ± 0.33) were comparable to those of PFOA (2.5 ± 0.32) and PFOS (2.8 ± 0.49), indicating their long-range transport potential in global oceans with ocean currents. Fig. 3A presents a summary of reported concentrations for typical PFAS, including PFOA, PFOA, PFDoDA, PFTrDA, and PFOS, while Fig. 3B provides a comparison of these concentrations.

Compared to seawater and sediment, PFAS in marine air was less frequently reported. n-PFAS in the air of the Bohai and Yellow Seas were investigated in 2012, where Σn-PFAS concentrations ranged from 79 pg/m^3 to 168 pg/m^3, from 157 pg/m^3 to 218 pg/m^3, and from 186 pg/m^3 to 4531 pg/m^3 in the air of the Bohai Sea, northern Yellow Sea, and the southern Yellow Sea, respectively (Zhao et al., 2017); most n-PFAS (>90%) were present in the gaseous phase rather than in the APM, and FTOHs (especially 8:2 and 10:2 FTOHs) were the predominant n-PFAS and accounted for >90% of ΣPFAS. Compared to the air from urban areas, n-PFAS composition in the air from remote regions was similar (i.e., 8:2 FTOH predominated, followed by 10:2 FTOH), conceivably owing to these compounds' relatively greater long-range atmospheric transport potential and longer atmospheric residence time. Few studies have been conducted on i-PFAS in the air from remote areas in China. Fang et al. (2018) and Zhao et al. (2020) conducted studies on i- and n-PFAS in the air in the Bohai and Yellow Seas in 2016, and they found that Σi- and Σn-PFAS concentrations ranged from 36 pg/m^3 to 110 pg/m^3 and from 6.8 pg/m^3 to 1703 pg/m^3, respectively; i-PFAS mainly occurred in APM, and n-PFAS mainly were present in the gaseous phase; the contributions of C4 and C6 i-PFAS were higher than those of C8 i-PFAS; C2 PFCA (i.e., trifluoroacetic acid, TFA) was the predominant i-PFAS, and 8:2 and 10:2 FTOHs predominated in the n-PFAS. n-PFAS levels were measured in the air in the SCS in 2013 (Lai et al., 2016), showing similar results to those of Fang et al. (2018). Lai et al. (2016) found that 8:2 and 10:2 FTOHs were the predominant n-PFAS, and detected most n-PFAS (>95%) in the gaseous phase, with a concentration range of 1.8–109 pg/m^3, which was lower than the concentrations found above the Bohai and Yellow Seas, as Fang et al. (2018) reported. In short, the strong stability and relatively high water solubility of i-PFAS allow them to be widespread in

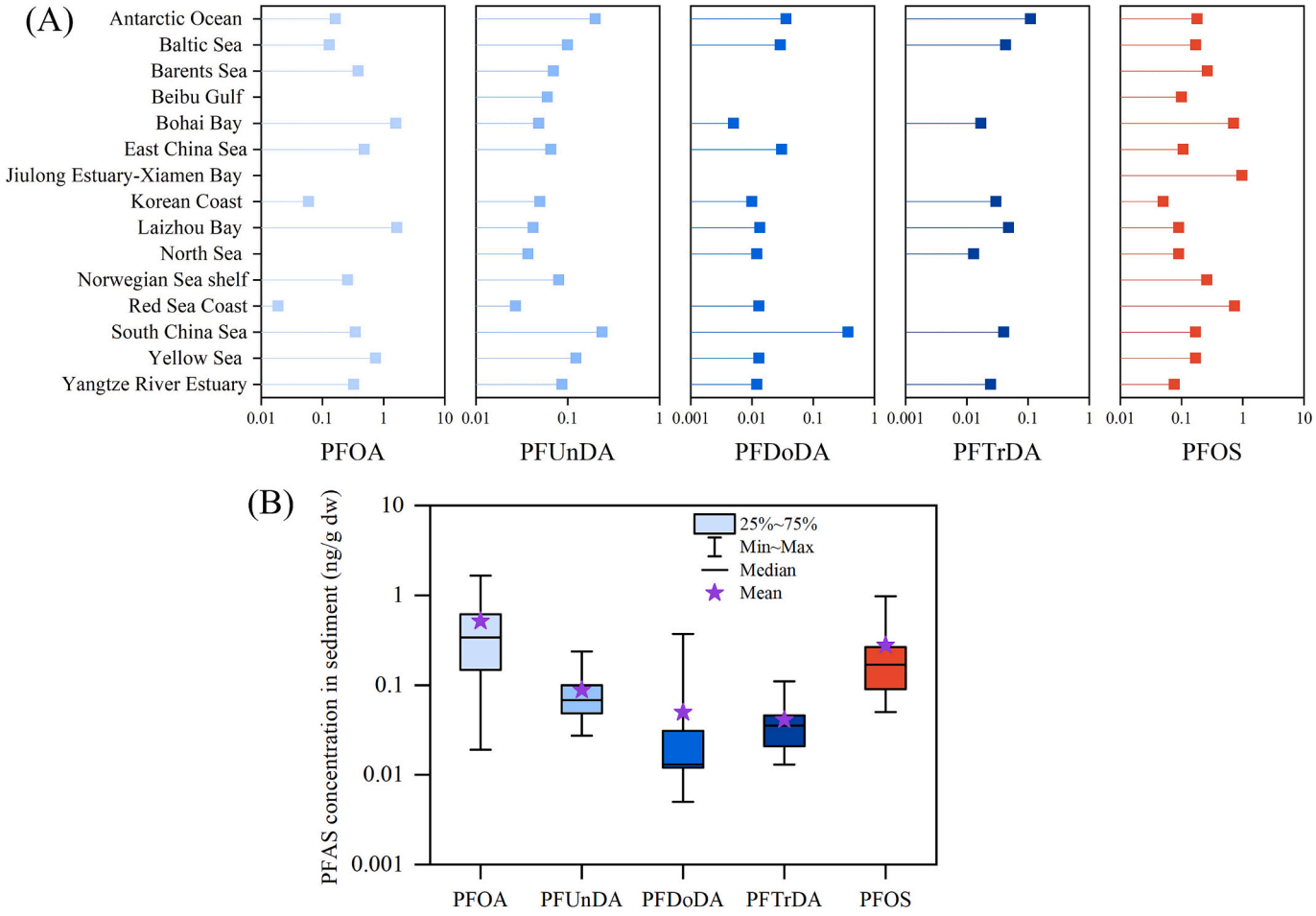


Fig. 3. (A) Reported concentrations of PFAS in sediment from and (B) comparison of different PFAS concentrations in sediment. *JAIPO represent joining area of Asia and the Indian-Pacific Ocean.

seawater. Short-chain PFAS are more likely to migrate via ocean currents, and long-chain PFAS have the potential to be distributed to sediments and organisms. In addition to direct water influx, the degradation of atmospheric PFAA precursors may be an important source of PFAS in the marine environment. The predominant PFAS in the seawater, sediment, and air are usually PFOA, PFOS, and 8:2 FTOH, respectively. However, this trend has changed somewhat in recent years with the increasing production and use of short-chain PFAS (e.g., PFBA and 6:2 FTOH) and emerging PFAS (e.g., HFPO-DA and 6:2 Cl-PFESA).

3. PFAS in marine organisms

3.1. PFAS bioaccumulation and biomagnification

Laboratory experiments have demonstrated that the bioaccumulation of PFCAs and PFSAs in the rainbow trout (*Oncorhynchus mykiss*) was strongly related to the perfluorocarbon chain length and head functional groups (Martin et al., 2003). Unlike other POPs, PFAS are found predominantly in protein-rich compartments (e.g., blood and liver) rather than in lipid-rich tissues in biota, indicating that the bioaccumulation potential of PFAS cannot be inferred simply based on log K_{ow} (Conder et al., 2008; Wang et al., 2022). A schematic diagram of PFAS in the marine environment is shown in Fig. 4.

In the marine environment, most studies have focused on i-PFAS in aquatic organisms due to the relatively high water solubility of PFAS. Kwadijk et al. (2010) investigated 15 PFAS in the European eel (*Anguilla anguilla*) and their inhabiting water from the Netherlands, and reported log-transformed bioaccumulation factors (log BAFs) of C7–C9 PFCAs and C4–C8 PFSAs were 1.09–3.26 and 1.4–3.3, respectively; they found perfluorocarbon chain length was strongly correlated with BAFs (Kwadijk et al., 2010). Hong et al. (2015) analyzed 12 PFAS in water from the west coast of South Korea, and they found the bioaccumulation of PFAAs varied among species, where higher concentrations of PFAAs were found in the intestine of amblyopinae (eel gobies), indicating that PFAAs were mainly accumulated via dietary exposure. In comparison, they found higher PFAA concentrations in oysters' gills and intestines, implying waterborne and dietary exposure to PFAAs in these bivalves. Ahrens et al. (2015) collected the European perch (*Perca fluviatilis*) and water samples around a PFAS-contaminated region (airport) in Stockholm, and found the whole-body burden of PFAS in *P. fluviatilis* was 334 ± 80 μg/g dw, with the distribution of PFAS in the fish body as follows: gonad > liver ≈ muscle > blood > gill, and the BAFs increased by 0.6–1.7 log units for each additional $-CF_2-$ moiety for PFCAs and PFSAs. Zhang et al. (2015) studied the trophic transfers of structural PFCA isomers in 11 marine species collected from Liaodong Bay in China, and found the total concentrations of linear PFCAs were 0.35–1.10, 0.93–2.61, and 2.13–2.69 ng/g wet weight (ww), and the corresponding proportions of branched PFCAs in linear PFCAs were 6.6%–15.5%, 4.2%–9.9%, and 4.5%–6.0% in invertebrates, fishes, and birds, respectively. They also found significant positive relationships between tissue concentrations of most target linear and branched PFCAs and trophic levels, and the trophic magnification factors (TMFs) of these PFCAs ranged from 1.90 to 4.88; the higher biomagnification of linear PFCAs than that of branched PFCAs indicated that liner PFCAs were more bioaccumulative, leading to high proportions of liner PFCAs among ΣPFCAs in organisms at high trophic levels. Munoz et al. (2017) surveyed the trophodynamics of 19 PFAS in a temperate estuary from Gironde in France, and they found PFOS, perfluorooctane sulfonamide (FOSA), and C8–C14 PFCAs were the predominant PFAS in the biota. Table 2 shows the TMFs of PFOS and PFOA in marine food webs across various studies.

Apart from legacy PFAS, researchers have also found that many emerging PFAS are bioaccumulative in marine organisms. Liu et al. (2017) reported the frequent occurrence of Cl-PFESAs and their trophic transfer behavior in marine organisms in the Chinese Bohai Sea, where 6:2 Cl-PFESA (<0.016–0.575 ng/g ww) was the dominant congener, followed by 8:2 Cl-PFESA (<0.022–0.040 ng/g ww) compared to other PFAS, the Cl-PFESA levels were lower in marine organisms, but like

Table 2
Trophic magnification factors (TMFs) of PFOS and PFOA in marine food webs. ND denotes not determined.

TMF for PFOS	TMF for PFOA	Taxonomic group	Reference
4.88	ND	Invertebrates and fish	(Wang et al., 2023a)
1.75	0.35	Invertebrates and fish	(Pan et al., 2021)
1.62	ND	Phytoplankton, zooplankton, invertebrates, and fish	(Li et al., 2021)
−1.57	0.2	Invertebrates and fish	(Du et al., 2021)
1.53	ND	Invertebrates and fish	(Miranda et al., 2021)
3.94	−1.57	Invertebrates, fish, seabirds, and marine mammals	(Chen et al., 2018)
17.4	3.28	Phytoplankton, zooplankton, fish, and marine mammals	(Kelly et al., 2009)
6.3	2.1	Zooplankton, fish, and marine mammals	(Tomy et al., 2009)
1.8	6.3	Phytoplankton, fish, and marine mammals	(Houde et al., 2008)
3.1	/	Zooplankton, Invertebrates, fish, and marine mammals	(Tomy et al., 2004)

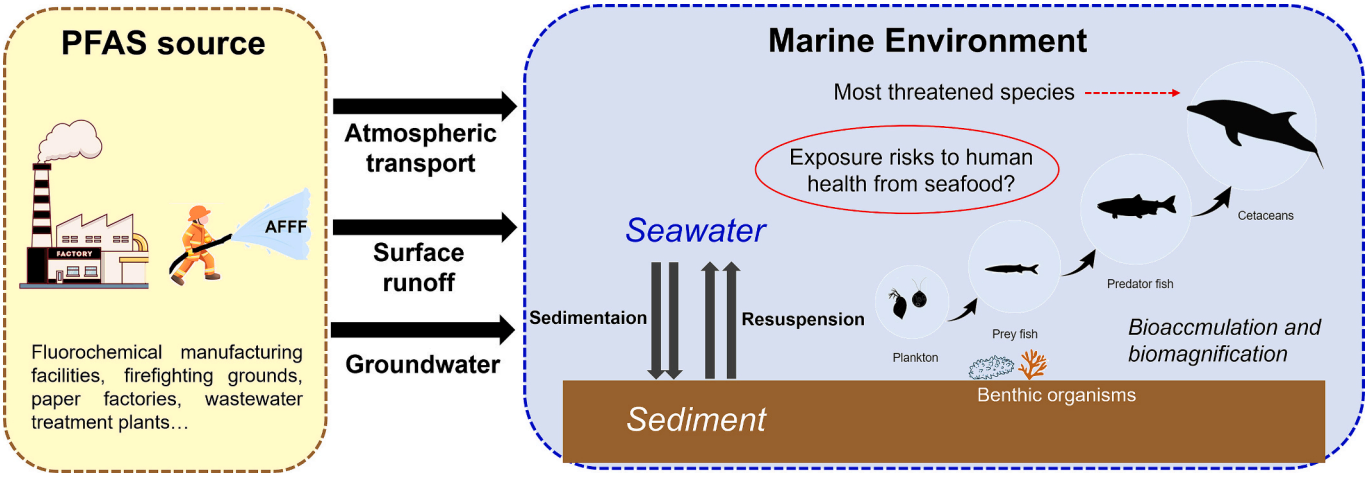


Fig. 4. A schematic diagram of PFAS in the marine environment.

PFOS and long-chain PFCAs, 6:2 Cl-PFESA could be biomagnified along the food chain. Similarly, 6:2 Cl-PFESA was also widely detected and found to have biomagnification potential in a subtropical marine food web in the Beibu Gulf (Pan et al., 2021). Li et al. (2021) detected elevated perfluoroalkyl ether carboxylic acid (PFECA) concentrations in organisms from the Xiaoqing River estuary in China. They found perfluoro-2-methoxyacetic acid (PFMOAA), hexafluoropropylene oxide trimer acid (HFPO-TrA), and PFOA were predominant in biotas; PFECAs with ≥6 perfluorinated carbons (HFPO-TrA, HFPO-TeA) and perfluoro (3, 5, 7, 9, 11-pentaoxadodecanoic) acid (PFO5DA) were biomagnified along the food chain (TMF > 1), and PFMOAA with the least perfluorinated carbons underwent trophic dilution (TMF < 1). However, reports about the bioaccumulation potential of emerging PFAS are still relatively limited compared to legacy PFAS (e.g., PFOS and PFOA). Considering the increasing use and detection of emerging PFAS, more research on the bioaccumulation potential of emerging PFAS should be conducted.

It should be noted that fish, shrimp, crabs, bivalves, and various other marine organisms also serve as a staple food source for billions of people worldwide. The significant bioaccumulation of PFAS in marine organisms may bring PFAS exposure risk to humans via daily seafood consumption. For example, the total PFAS concentrations in the shellfish samples collected from four coastal cities along the Yellow Sea and Bohai Sea of Shandong Province, China ranged from 0.061 ng/g ww to 178.259 ng/g ww, and a potential health risk of PFOA was found for local residents who frequently consume crabs and mollusks (Zhang et al., 2023). In the Zhangjiang River Estuary, China, the mangrove estuarine residents had a high PFAS exposure risk through the intake of estuarine fish (Xie et al., 2024). Eating marine shellfish and crustaceans from Yellow-Bohai Sea coasts in China may bring PFAS exposure risk (Guo et al., 2023). Nevertheless, although many emerging PFAS (e.g., 6:2 Cl-PFESA and PFECHS) were also reported to have bioaccumulation potential, the risk they may bring is rarely reported as the relevant toxicological data are still limited. In addition, the effects of the PFAS mixture are rarely investigated.

3.2. PFAS in marine apex predators

High concentrations of PFAS were observed in top predators (e.g., polar bears and marine mammals) due to biomagnification, and thus potential ecological risks of PFAS exposure have also increased exponentially (Chen et al., 2021). Lam et al. (2016) analyzed PFAS in the liver samples of Indo-Pacific humpback dolphins (*Sousa chinensis*) and finless porpoises (*Neophocaena phocaenoides*) from the SCS between 2002 and 2014, and found the ΣPFAS levels ranged from 136 to 15,300 and 30.5 to 2720 ng/g dw in dolphins and porpoises, respectively; significant increasing trends of some PFCAs and PFBS were found in marine mammal samples between 2002 and 2014, but no trend was found for ΣPFAS over the sampling period, which was attributed to the increasing use of PFCAs and C4-based PFSAs following the restriction on the production and use of PFOS-related products. In addition, they observed significantly increasing temporal shifting trends of PFOS to PFBS, indicating the substitution of PFOS with the shorter chain C4 alternative; risk assessment was also conducted in that study and based on the toxicity thresholds, PFOS might pose a risk to marine mammals in the SCS. Similarly, in another study on dolphins and porpoises in the SCS stranded between 2012 and 2018, PFOS and 6:2 Cl-PFESA were reported to pose adverse effects on most of the investigated cetaceans (Wang et al., 2021a).

Gebbink et al. (2016) investigated legacy PFAS and their precursors and alternatives in the livers of ringed seals, polar bears, and killer whales from East Greenland, and found the concentrations of PFBS and 6:2 Cl-PFESA were approximately four orders of magnitude lower than those of PFOS, and PFOS was positively correlated with 6:2 Cl-PFESA but not PFBS; the mean ΣPFAS concentration in killer whales (269 ng/g dw) was comparable to that in ringed seals (138 ng/g dw) but was an order of magnitude lower than that in polar bear livers (2336 ng/g dw); FOSA was the only detected PFAS precursor, and the ratios of FOSA/PFOS and isomer patterns indicated that killer whales might have a lower metabolic capacity for FOSA degradation than polar bears and ringed seals. Spaan et al. (2020) investigated PFAS in marine mammals from the Northern Hemisphere, including the east coast of the United States (US) and the west and east coasts of Greenland, Iceland, and Sweden, using the target and suspect screening methods; they also analyzed the extractable organofluorine (EOF). The results showed that PFOS was the predominant PFAS, followed by one precursor, 7:3 fluorotelomer carboxylic acid (7:3 FTCA); twenty routinely monitored PFAS accounted for only a tiny fraction of the EOF in the investigated marine mammals, indicating that PFAS exposure was likely underestimated in the studied species from the Northern Hemisphere.

Long-chain PFAS have been widely detected in marine organisms globally since 2001. The strong bio-persistence and high bioaccumulation potential of long-chain PFAS (e.g., PFOS) have attracted worldwide attention, whereas short-chain PFAS are less frequently detected in marine organisms due to their low bioaccumulation potential. Because of the long-term production and use, PFOS is usually the predominant PFAS in marine organisms although it has been phased out globally since 2009. The high PFOS concentrations in marine organisms can be attributed to the exemptions of use, precursor degradation, and strong environmental persistence of PFOS. The PFOS concentrations in half of the digestive content samples of striped dolphins (*Stenella coeruleoalba*) from the NW Mediterranean Sea exceeded Environmental Quality Standards (EQSs) concentrations set in 2014 by the European Union, implying that PFOS-contaminated prey species may pose potential health risks to striped dolphins. However, current EQSs for PFAS aiming at protecting marine organisms mainly focus on PFOS and PFOA. When sufficient toxicity data are available, more EQSs should be derived, and the application of quantitative structure–activity relationship (QSAR) tools may be helpful.

4. Conclusions and Perspectives

Despite the widespread use and release of PFAS, research on the occurrence and distribution of PFAS in marine ecosystems remains less than studies conducted in freshwater ones. Existing studies on PFAS in the marine environment primarily focus on PFAAs, although there has been a growing interest in emerging PFAS and PFAS precursors in recent years. In addition, studies on the environmental risk brought by emerging PFAS are limited, which can be attributed to a lack of comprehensive toxicokinetic and toxicodynamic studies about emerging PFAS, especially on marine species.

Given the potential for bioaccumulation and biomagnification, PFAS (mainly PFOS and PFOA) in marine organisms could lead to adverse effects on humans through consumption of contaminated seafood. In particular, PFAS (mainly PFOS and some long-chain PFCAs) could be highly accumulated in marine predators (e.g., marine cetaceans) along the food chain and bring adverse effects on them. For future research on PFAS in the marine environment, we recommend exploring the following research directions:

4.1. Ultrashort-chain PFAS in the marine environment requires further investigation

The ultrashort-chain PFAS are usually referred to as PFCAs and PFSAs with carbon chain lengths of no more than three. These PFAS, characterized by their high lipophobicity, can be easily transported into marine ecosystems. Nevertheless, knowledge regarding the environmental behavior and toxicity of these ultrashort-chain PFAS in the marine environment is still scanty. Some ultrashort-chain PFAS, for example, TFA, are usually considered as a “safe” and “stable” degradation product of long-chain PFAS. Nevertheless, many studies have reported that ultrashort-chain PFAS, like TFA, can also pose adverse

effects after reaching a certain concentration (Solomon et al., 2016). Thus, investigating ultrashort-chain PFAS is of certain significance in the context of gradually phasing out long-chain PFAS. It is recommended to optimize the existing general PFAS analysis method to meet the requirements of ultrashort-chain PFAS analysis. Ultrashort-chain PFAS, especially for TFA, should be included in the regular PFAS target list to systematically study their occurrence, distribution, and environmental fate in the marine environment. In addition, ultrashort-chain PFAS are generally not considered to have significant potential for bioconcentration and biomagnification. However, long-term releases of these highly water-soluble chemicals may bring adverse effects on marine organisms with relatively low trophic levels, such as algae and bivalves. Conducting more research on the toxicity of ultrashort-chain PFAS to different marine species and constructing corresponding species sensitivity distribution curves are important for protecting marine ecosystems from the threat of ultrashort-chain PFAS. In addition, the extreme stability of ultrashort-chain PFAS in the marine environment means they can serve as effective tracers of ocean currents. This characteristic can facilitate studies on oceanographic processes and pollutant transport. In addition, the release of PFAS during lithium-ion battery recycling has attracted increasing attention in recent years (Rensmo et al., 2023). Many of these PFAS, such as bis(trifluoromethylsulfonyl) imide, which contains fewer than four carbon atoms, exhibit properties similar to ultrashort-chain PFAAs, including high water solubility and strong environmental persistence. These characteristics enhance their potential to migrate into the marine environment. Consequently, future research should elucidate the environmental fate and evaluate the potential ecological risks of these lithium-ion battery recycling-related PFAS in marine ecosystems.

4.2. Establishing environmental quality standards (EQSs) in the marine environment

Environmental factors in the marine environment, such as salinity, tides, and ocean currents, play a critical role in the fate of pollutants. In addition, physiological differences between freshwater and marine organisms, particularly in osmoregulation, significantly influence the bioavailability and toxic mechanisms of pollutants. However, research on the ecological risks of PFAS in the marine environment remains limited compared to freshwater systems. This knowledge gap has further resulted in the lack of established EQSs for most PFAS in marine ecosystems. Given the biomagnification potential of PFAS, both aquatic predators and human populations are particularly susceptible to their toxic effects. Considering the significant role seafood plays in the diets of many populations, particularly in coastal communities, it is vital to assess human exposure to PFAS through consuming PFAS-contaminated marine organisms. Establishing related EQS will help ensure that seafood remains safe for human consumption and that regulatory measures are in place to protect public health. In addition, it is particularly important to establish EQSs focusing on protecting marine predators, as they are at a higher risk because of the biomagnification of PFAS. By setting EQSs, we can create a framework for monitoring, risk management, and mitigating the impacts of PFAS on marine biodiversity.

4.3. More research into the environmental fate and toxic effects of emerging PFAS and their mixtures

With the increasing detection of emerging PFAS in the environment, it is important to conduct further studies on their environmental behavior, toxicological effects, and mechanisms. As emerging PFAS continue to expand after the phase-out of legacy PFAS, understanding their toxicological profiles is critical for assessing potential risks to ecological systems and human health. Comprehensive toxicological studies will help identify the adverse effects associated with these emerging PFAS, ultimately guiding the development of safer and more environmentally friendly chemical alternatives. The application of QSAR in this process can help address the challenges posed by the growing numbers of newly synthesized and applied emerging PFAS. Furthermore, PFAS often exist in a cocktail and thus future risk assessment should consider their combined effects through establishment and application of appropriate models. The advancement of relevant knowledge and information will be highly essential to the sustainable use of chemicals, because we will be able to select and keep using those less harmful PFAS while phasing out the persistent, bioaccumulative and toxic ones. This will also support the United Nations' Decade of Ocean Science for Sustainable Development (2021−2030) to create cleaner and safer oceans for all.

CRediT authorship contribution statement

Qi Wang: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Yetong Shao:** Writing – review & editing. **Kenneth M.Y. Leung:** Writing – review & editing, Supervision. **Paul K.S. Lam:** Writing – review & editing, Supervision. **Yuefei Ruan:** Writing – review & editing, Supervision, Resources, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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