



Spatial and vertical distribution of per- and polyfluoroalkyl substances (PFASs) in the water columns of the regional seas of South Korea

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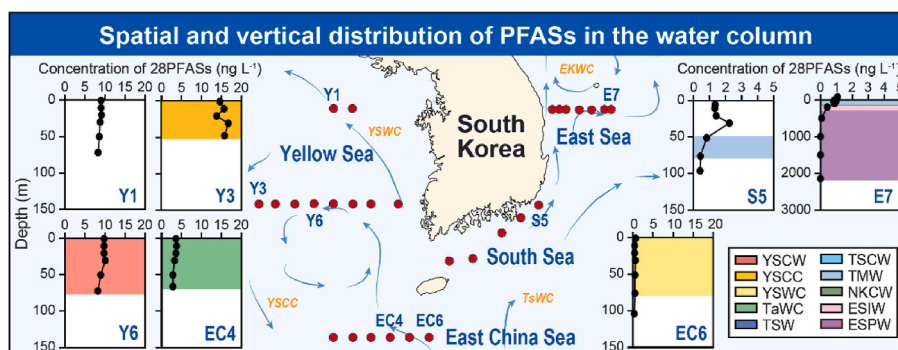
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HIGHLIGHTS

- PFAS concentrations were highest in the Yellow Sea, followed by the East China Sea.
- PFOA was the most predominant compound in water columns of the regional seas.
- Short-chain and alternatives, L-PFBS, Gen-X, and PFBA, were detected in seawater.
- PFASs in the Yellow Sea are likely transported to the East Sea via ocean currents.
- The East China Sea appears to be a mixing zone for PFASs from various sources.

GRAPHICAL ABSTRACT



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ABSTRACT

This study focused on analyzing the spatial and vertical distributions of 28 per- and polyfluoroalkyl substances (PFASs), which comprised five precursors and three alternatives, in the water columns of the regional seas surrounding South Korea, such as the Yellow Sea (YS, Y1–Y10), East China Sea (ECS, EC1–EC6), South Sea (SS, S1–S5), and East Sea (ES, E1–E7). The concentrations of these PFASs detected in 204 seawater samples varied from below the limit of detection (<LOD) to 17 ng L⁻¹ in the YS, 0.26–17 ng L⁻¹ in the ECS, 0.08–3.4 ng L⁻¹ in the SS, and <LOD to 1.4 ng L⁻¹ in the ES, with perfluorooctanoic acid being identified as the most abundant compound. Principal component analysis grouped water masses and regions based on PFASs concentrations and compositions, enabling the identification of PFASs sources and their fate. PFASs are mainly derived from land and are transported via ocean currents, where their compositions tend to remain conservative. PFASs entering the YS are likely conveyed to the ES through ECS and SS, following the northward movement of the Taiwan Warm Current and Kuroshio Current. The ECS serves as a mixing zone for PFASs from various sources. This study provides valuable baseline data for understanding PFASs transport and the characteristics of water masses in the regional seas around South Korea.

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1. Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are synthetic compounds characterized by a fluorinated carbon backbone and unique physicochemical properties, including high surface activity, exceptional chemical stability, and effective water and oil repellency (Cousins et al., 2020; Lindstrom et al., 2011; Park et al., 2020). C₈ perfluoroalkyl sulfonic acids (PFASs), C₈ perfluoroalkyl carboxylic acids (PFCAs), and their salts are regulated under the Stockholm Convention owing to their persistence and potential toxicity (UNEP, 2010, 2019). As a result, the use of newer fluorinated alternatives has increased, including short-chain PFASs (e.g., PFASs with chains shorter than C₆ and PFCAs with chains shorter than C₈), as well as novel compounds like 2,3,3,3-tetrafluoro-2-(1,1,2, 2,3,3,3-heptafluoropropoxy)-propanoic acid (Gen-X), dodecafluoro-3H-4,8-dioxanonoate (ADONA), and 9-chlorohexadecafluoro-3-oxanonane sulfonate (F53B) (Lu et al., 2019). Although these substances are regarded as alternatives, they have exhibited harmful impacts similar to perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), along with comparable environmental persistence (Munoz et al., 2019; Wang et al., 2015). This underscores the necessity of exploring the distribution and fate of emerging PFASs in the environment.

PFASs are known to be transported through water owing to their high polarity and solubility (Prevedouros et al., 2006; Zhao et al., 2015). Short-chain PFASs are especially known for their higher hydrophilicity and greater stability in aquatic environments (Li et al., 2018). For example, perfluorobutanoic acid (PFBA), which belongs to short-chain PFCA (C₄–C₇), has been shown to enter marine environments more readily than long-chain compounds and can travel significant distances without substantial degradation (Hong et al., 2013; Shi et al., 2015; Wang et al., 2016). PFASs have also been suggested as useful chemical tracers for investigating oceanic circulation patterns and water mass characteristics, as they primarily undergo horizontal seawater transport rather than vertical sinking (Yamashita et al., 2008).

Previous research on PFASs in Korean waters has primarily focused on rivers and wastewater treatment plants (WWTPs) (Lam et al., 2014; Son et al., 2013). PFASs enter the regional seas through rivers and estuaries, stemming from sources such as waste disposal sites, landfill leachates, effluents from WWTPs, and industrial discharges (Ahrens, 2011; Helmer et al., 2022; Prevedouros et al., 2006; Zhou et al., 2018b). Thus, the influence of the Yellow Sea (YS) via the South Sea (SS) is expected to serve as a significant source of PFASs in the surface waters of the East Sea (ES). Nevertheless, there has been limited research on PFASs in the regional seas surrounding South Korea (Lee et al., 2020; Shen et al., 2018), even though these regional seas are vital for the global transport of PFASs, enabling their extensive movement through ocean currents (Kannan et al., 2001; Boisvert et al., 2019; MacInnis et al., 2017).

This research intends to explore the spatial and vertical distribution characteristics of PFASs, including legacy and emerging compounds and their precursors, within the water columns of the seas adjacent to Korea. Specific objectives are: (i) to determine the distributions of PFASs in the water column, (ii) to evaluate the spatial variability of PFASs through regional comparisons, and (iii) to enhance the understanding of seawater circulation patterns in the region. This study aims to provide baseline data on the distribution of existing and emerging PFASs in the water column of Korean waters. It also seeks to offer fundamental insights for assessing the risks to the coastal ecosystem. In addition, the present study is expected to contribute to a better understanding of water mass characteristics and ocean circulation patterns in the regional seas of Korea.

2. Materials and methods

2.1. Study area, seawater sampling and in situ measurements

This study was conducted in three regional seas of Korea: the YS, SS, and ES. The YS is geographically adjacent to the northern East China Sea (ECS) continental shelf, bordered by Korea, China, and Japan, and is influenced by the Tsushima Current and diluted coastal waters from river discharges in China (Beardsley et al., 1985). The main current in the SS is the Tsushima Current, with contributions from discharges of the Yangtze River reaching this region (Lim et al., 2007). The ES, a semi-enclosed sea, has surface waters influenced by currents from the ECS and the North Pacific Ocean via the Tsushima Strait (Gamo and Horibe, 1983).

A total of 204 seawater samples were collected from the YS (Y1–Y10, maximum depth: 87 m) and the ECS (EC1–EC6, maximum depth: 103 m) using the R/V *ISABU* in January 2024, from the SS (S1–S5, maximum depth: 110 m) using the R/V *NARA* in May 2023, and from the ES (E1–E7, maximum depth: 2165 m) using the R/V *ISABU* in January 2023 (Fig. 1). The full list of samples collected is provided in the Supplementary Materials (Table S1). Seawater samples were collected from various depths with Niskin bottles connected to a rosette sampler, and preserved in 1-L HDPE bottle. To ensure sample integrity, the bottles were pre-treated with methanol (MeOH) and rinsed with seawater three times before sampling. Glass fiber filters (GF/F) were not utilized to minimize the PFASs adsorption. Additionally, after collecting samples at each sampling site and draining all seawater from the Niskin bottles, field blanks (1 L of Milli-Q water, n = 12) were collected by passing Milli-Q water through the Niskin bottles to monitor potential contamination. Seawater temperature (T), salinity (S), and depth were measured on-site with CTD sensors. The T and S data from these sensors were used to examine thermohaline characteristics and classify the water masses. Detailed CTD data are provided in Table S2.

2.2. Chemicals and reagents

The 11 PFCAs analyzed in this study include PFBA, perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTriDA), and perfluorotetradecanoic acid (PFTeDA). The 7 linear and 2 branched PFASs were analyzed including perfluorobutane sulfonate (L-PFBS), perfluoropentane sulfonate (L-PFPeS), perfluorohexane sulfonate (L-PFHxS), branched (br)-PFHxS, perfluoroheptane sulfonate (L-PFHpS), L-PFOS, br-PFOS, perfluorononane sulfonate (L-PFNs), and perfluorodecane sulfonate (L-PFDS). In addition, five precursors were analyzed as N-methylperfluoro-1-octane sulfonamidoacetic acid (N-MeFOSAA), N-ethylperfluoro-1-octanesulfonamidoacetic acid (N-EtFOSAA), 1H,1H,2H,2H-perfluorohexanesulfonate (4:2 FTS), 1H,1H,2H,2H-perfluorooctanesulfonate (6:2 FTS), and 1H,1H,2H,2H-perfluorodecane sulfonate (8:2 FTS). Finally, three emerging PFASs were investigated such as ADONA, Gen-X, and F53B. The standard materials utilized included PFAC-MXH and the single compounds L-PFHxS, L-PFOS, ADONA, Gen-X, and F53B. Mass-labeled standard mixtures (MPFAC-MXA) were used as internal standards (IS). All standards were purchased from Wellington Laboratories (>98% purity, Guelph, Canada). Detailed information on the target compounds and standards, and the materials and solvents used in this study are presented in Tables S3 and S4, respectively.

2.3. Analysis of PFASs

Target PFASs in seawater were extracted using Oasis® Wax cartridges (6 mL, 150 mg, 30 µm, Waters, Milford, MA) that were conditioned with 0.1% NH₄OH in MeOH, followed by MeOH, and water. A volume of 1 L of unfiltered seawater, spiked with 5 ng of IS, was processed through the cartridge at a flow rate ranging from 8 to 10 mL min⁻¹. Field blanks and seawater samples were collected on these pre-conditioned cartridges, stored in the dark at -20 °C, and later delivered to the laboratory for analysis. After loading 1 L of a seawater sample, the cartridge was rinsed with 4 mL of 25 mM NH₄CH₃CO₂ solution, then vacuum filtered using an aspirator for 20 min to remove any water remaining in the cartridge. Elution was carried out using 4 mL of 0.1% NH₄OH in MeOH, followed by an additional 4 mL of MeOH. The eluant was concentrated using nitrogen gas at 40 °C until the volume was reduced to below 0.2 mL. Finally, the volume was brought to 1 mL by adding MeOH, and the solution was passed through a 0.2 µm polypropylene filter before instrumental analysis (Fig. S1).

The analysis of the 28 PFASs was conducted using an Agilent 1290 Infinity II HPLC in combination with an Agilent 6470 triple quadrupole mass spectrometer (Agilent Technologies, Santa Clara, CA). For the separation of these PFASs, a BEH Shield RP18 column (2.1 × 100 mm, 1.7 µm, Waters) was utilized. The mobile phase was an 80:20 mixture comprising (A) water and (B) MeOH and acetonitrile, both containing 10 mM ammonium acetate. The PFASs were analyzed using the multiple reaction monitoring (MRM) mode of the mass spectrometer. Data analysis was conducted using MassHunter software. Further details regarding instrument settings and MRM transitions were provided in Tables S5 and S6.

2.4. Quality control

A nine-point standard calibration curve was created for each analyte, with concentrations of 0.1, 0.2, 0.5, 1, 2, 5, 10, 20, and 50 ng mL⁻¹. All curves showed satisfied linearity, with correlation coefficients (R²) > 0.99. Field and procedural blanks were analyzed to check for any contamination during the sample preparation process. The limit of detection (LOD) and the limit of quantification (LOQ) for each analyte were determined based on signal-to-noise ratios of 3:1 and 10:1, respectively (Table S7). In all blank samples, no PFASs were detected (<LOD). For seawater samples, the LOQs ranged from 0.03 to 0.76 ng L⁻¹. The recoveries of ISs in seawater samples, procedural blanks, and field blanks were 101 ± 11%, 109 ± 10%, and 109 ± 8.6%, respectively (Tables S8–10).

2.5. Statistical analysis

Principal component analysis (PCA) was performed using IBM SPSS Statistics 26 (Armonk, NY) to identify PFASs distribution patterns in the water masses of the study area and evaluate their movement through ocean currents. Before performing the analysis, the concentration data were log-transformed. For any PFASs concentrations below the LOQ, values of LOQ/2 were used in place. The adequacy of the dataset for PCA was validated through Kaiser-Meyer-Olkin (KMO) and Bartlett's test, with the KMO value exceeding the 0.6, confirming the data suitability for PCA.

3. Results and discussion

3.1. Characteristics of water masses in the study area

To investigate the distribution of PFASs across various water masses,

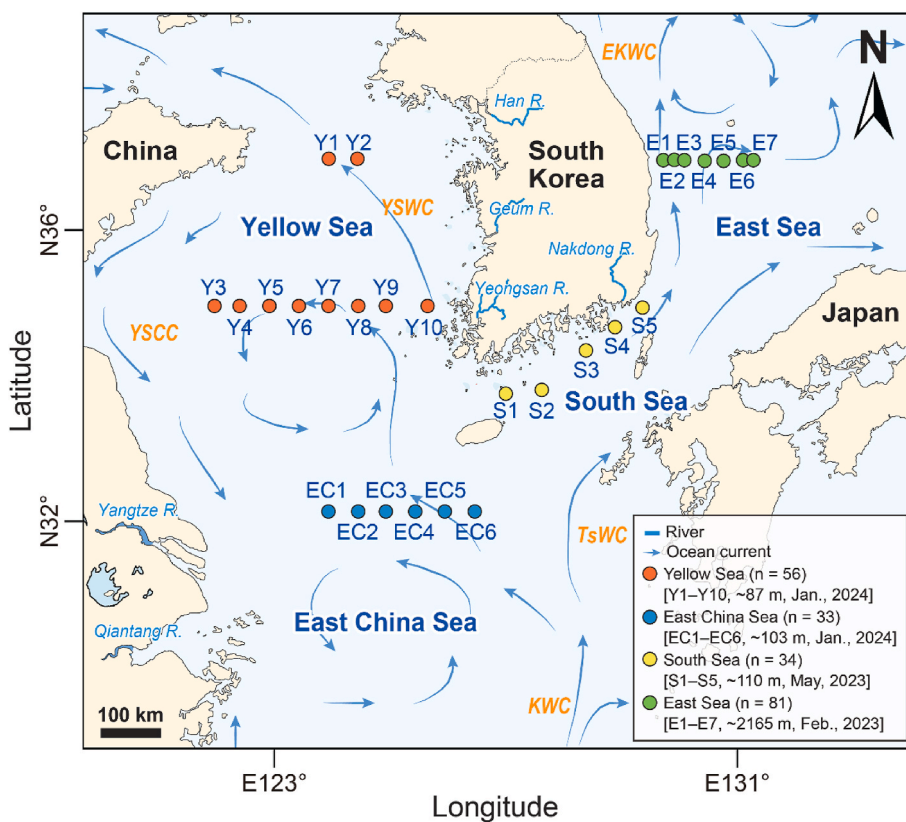


Fig. 1. Map of seawater sampling stations and ocean currents in the Yellow Sea, East China Sea, South Sea, and East Sea [YSCC: Yellow Sea Coastal Current; YSWC: Yellow Sea Warm Current; KWC: Kuroshio Warm Current; TsWC: Tsushima Warm Current; EKWC: East Korean Warm Current (Guo et al., 2006; Hu et al., 2011; Lee et al., 2009)].

the water column was divided into 10 water masses according to the T and S (Fig. S2) (Guo et al., 2020; Quan et al., 2013; Yoon et al., 2015). In the YS, two main water masses were identified: the Yellow Sea Cold Water (YSCW) and the Yellow Sea Coastal Current (YSCC). In the ECS, three water masses were observed: YSCW, Taiwan Warm Current Water (TaWC), and Yellow Sea Warm Current (YSWC). The YS and ECS are shallow, and due to vertical mixing during the winter, no significant water masses could be distinguished based on water depth. In the SS, the dominant water masses were Tsushima Surface Water (TSW), Tsushima Middle Water (TMW), and Tatar Surface Cold Water (TSCW). Specifically, TSW was prevalent at 0–10 m depth, TSCW around 20 m, and TMW between 30 and 100 m. In the ES, five distinct water masses were observed: TSCW, TMW, North Korean Cold Water (NKCW), East Sea Intermediate Water (ESIW), and East Sea Proper Water (ESPW). In this region, TSCW occurred at depths of 0–10 m, TMW at 10–100 m, NKCW at 150–200 m, ESIW at 200–350 m, and ESPW below 500 m. This distribution highlights the complex vertical and horizontal structure of water masses in the area.

3.2. Spatial distributions of PFASs in surface seawater

Among the 28 target compounds, 10 PFASs, including 6 PFCAs, 3 PFSAs, and 1 alternative, were detected in seawater samples from coastal waters around South Korea (Table S11). The PFASs concentrations varied widely from <LOD to 17 ng L⁻¹, with a mean concentration of 4.1 ng L⁻¹ (Fig. 2). A figure illustrating the overall characteristics of water masses in the study area is provided in Fig. S3. In all seawater samples, the levels of PFCAs were found to be consistently higher than PFSAs. Despite global regulatory efforts, PFOA (mean: 2.9 ng L⁻¹) dominated among the compounds in seawater, making up 93% of the total PFASs found. These findings align with previous results indicating that PFOA was the most dominant compound among PFASs in the East China Sea (Zhou et al., 2018a). PFOA is recognized as the dominant PFASs discharged from the Yangtze River, likely dispersing to other regions via ocean currents (Du et al., 2022). In addition to PFOA, PFHxA (mean: 0.34 ng L⁻¹) and L-PFBS (0.22 ng L⁻¹) were detected in 84% and 68% of the total seawater samples, respectively. PFBS has been reported as a more commonly used alternative to PFOS compared to other substitutes (Cai et al., 2012; Zhou et al., 2013). PFBA (mean: 0.09 ng L⁻¹), PFPeA (0.18 ng L⁻¹), PFHpA (0.16 ng L⁻¹), PFNA (0.03 ng L⁻¹), L-PFOS (0.13 ng L⁻¹), L-PFNS (0.009 ng L⁻¹), and GenX (0.05 ng L⁻¹) had detection frequencies of 28%, 47%, 50%, 30%, 40%, 1%, and 21%, respectively.

Concentrations of PFASs in surface water (0–10 m) were highest in the YS (mean: 9.9 ng L⁻¹), followed by the ECS (mean: 8.1 ng L⁻¹), SS (mean: 1.4 ng L⁻¹), and ES (mean: 0.97 ng L⁻¹) (Fig. 2). PFASs concentrations were generally higher in coastal areas of China within the YS and ECS, gradually decreasing towards the open sea with observations reported in previous studies. The concentrations have also shown a decreasing trend over time (Zhou et al., 2018a). A similar pattern was observed in the concentrations of PFCAs, PFSAs, short-chain PFASs, and

emerging PFASs in surface seawater (Fig. S4). This trend is likely influenced by dilution effects caused by ocean currents (Wang et al., 2019). Furthermore, the decreasing trend may result from biogeochemical processes, including the adsorption of PFASs onto sediment particles, followed by removal through sedimentation, in addition to dilution effects (Cai et al., 2012; Wang et al., 2019; Yamazaki et al., 2019; Zheng et al., 2017). In the SS, PFASs concentrations were highest at station S3, near Tongyeong, and exhibited a dilution trend moving northeastward with the Tsushima Current. This pattern may be attributed to treated emissions from industrial complexes and nearby sewage treatment plants in Tongyeong City (Bae et al., 2017). The East Sea coast is significantly influenced by the monsoon. During summer, the southeast wind drives surface currents northward, while in winter, the northwest wind induces southward flows that hinder the northward currents. As a result, the East Sea coast exhibits a unique water mass structure where cold and warm currents interact, leading to complex spatial and temporal distribution patterns of highly conservative factors such as water temperature and salinity. In the ES, PFASs concentrations showed no significant variation between sites, potentially due to the more variable temperature profiles and complex ocean currents present above the thermocline in this region compared to other sea areas (Kim et al., 2004).

3.3. Vertical distributions of PFASs in water columns

In the vertical profiles, PFASs concentrations were as follows: in the YS, they ranged from 0.19 to 17 ng L⁻¹ (mean: 10 ng L⁻¹); in the ECS, from 0.26 to 17 ng L⁻¹ (6.1 ng L⁻¹); in the SS, from 0.08 to 3.4 ng L⁻¹ (1.1 ng L⁻¹); and in the ES, concentrations were below the detection limit to 1.4 ng L⁻¹ (0.66 ng L⁻¹) (Fig. 3). In the YS and ECS, PFASs levels did not exhibit a consistent decrease with depth, likely due to the shallow nature of these waters and the vertical mixing that occurs during the winter (Steinke et al., 2011). In contrast, the SS and ES showed a downward trend in PFASs concentrations as depth increased, consistent with findings from earlier studies (Han et al., 2022). The same pattern was observed in the vertical profiles of PFCAs, PFSAs, short-chain PFASs, and emerging PFASs in seawater (Fig. S5). In the ES, decreased PFASs concentrations with depth may also result from perfluorochemicals being adsorbed onto suspended particulate matter and settling into bottom sediments, leading to their removal from the water column (Yamazaki et al., 2019). Future studies should examine PFAS concentrations in sediments in relation to total organic carbon for a deeper understanding of this phenomenon. Additionally, PFASs may be diluted from the water column through vertical eddy diffusion driven by concentration gradients across ocean depths or through the subduction of water masses originating from different sources at varying depths (Lohmann et al., 2013). The study area is affected by the East Asian monsoon, characterized by strong winter monsoons with cold, dry northwest or northeast winds. Vertical mixing is known to occur more deeply in winter and less in summer due to the effects of the monsoon (Steinke et al., 2011). Overall, the vertical distribution of PFASs in

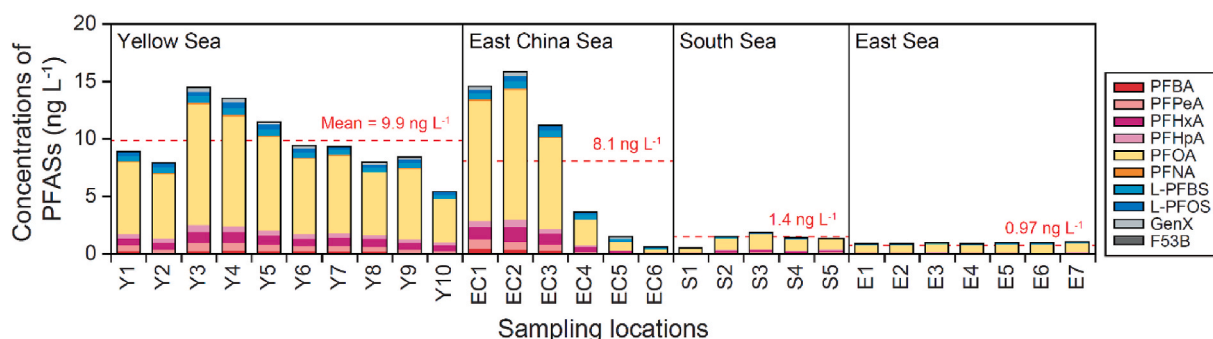


Fig. 2. Concentrations of PFASs in surface seawater in the Yellow Sea, East China Sea, South Sea, and East Sea.

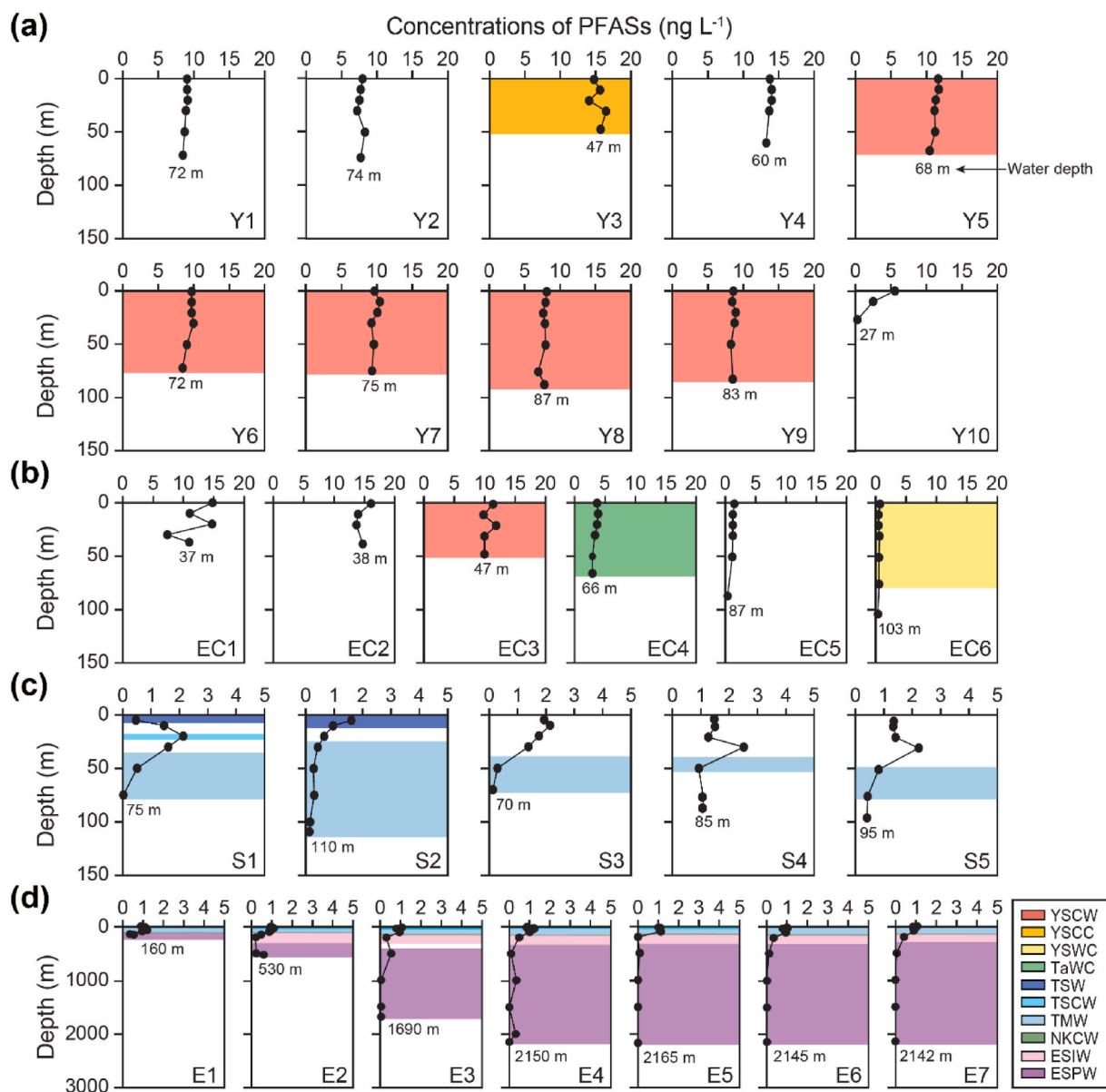


Fig. 3. Vertical profiles of PFASs in the water column of the (a) Yellow Sea, (b) East China Sea, (c) South Sea, and (d) East Sea [YSCW: Yellow Sea Cold Water; YSCC: Yellow Sea Coastal Current; YSWC: Yellow Sea Warm Current; TaWC: Taiwan Warm Current; TSW: Tsushima Surface Water; TSCW: Tatar Surface Cold Water; NKWC: North Korean Cold Water; TMW: Tsushima Middle Water; ESIW: East Sea Intermediate Water; ESPW: East Sea Proper Water (Guo et al., 2020; Quan et al., 2013; Yoon et al., 2015)].

seawater was significantly influenced by seasonal vertical mixing, except in deeper waters where wind does not affect water movement.

PFASs concentrations peaked in water masses influenced by YSCW in the YS and ECS, TSW at 0–10 m in the SS, and TMW at depths of 0–100 m in the ES. High PFASs levels in the ECS are largely attributed to pollution from the Yangtze River runoff (Yanagi, 2002). This river is a key factor in increasing PFASs levels in the southern parts of the YS and ECS. The Tsushima Warm Current (TSWC), a branch of the KWC, moves water from the continental shelf of the ECS to the southwestern ES through the Tsushima Strait (Hase et al., 1999; Ito et al., 2014; Senjyu et al., 2006). PFASs were likely transported by the TSWC, impacting surface waters in the SS. Similarly, the surface water of the East Sea may have been affected by perfluorochemicals carried along by the Tsushima Warm Current. Further explanation of this phenomenon follows in section 3.5.

3.4. PFASs compositions in water columns

PFOA constituted the largest proportion of all PFASs detected in the water columns across all study areas, with PFCAs being more prevalent than PFASs (Fig. S6). The composition of PFASs in seawater showed a higher proportion of long-chain compounds at greater depths. Long-chain compounds, which have lower water solubility and higher Log K_{ow} values than short-chain compounds, are expected to settle and accumulate in deeper waters (Yeung et al., 2017; Zhou et al., 2018a). The PFASs composition in bottom waters across the four sea areas followed similar trends to surface waters (Fig. S7). The YS, ECS, and SS have relatively shallow depths; thus, the composition of PFASs in the surface water may also influence the bottom water due to wind-driven mixing. On the other hand, the ES is deep and exhibits strong stratification, resulting in very slow vertical mixing between surface and bottom waters (Kim et al., 2004; Yanagi et al., 2002). In the bottom layer of the ES (below ~300 m), there is cold and dense water known as ESPW,

characterized by a temperature range of 0.0–0.1 °C and a salinity range of 34.06–34.08; this water mass accounts for 84% of the total volume of the ES (Nihashi et al., 2017; Yanagi et al., 2002; Yoon et al., 2017) (Fig. S3). Consequently, the vertical mixing of PFASs is considered to be very limited.

The distribution of PFASs varied distinctly by sea area (Figs. S3 and S4). In water samples from the YS, the detected PFCAs included PFBA, PFPeA, PFHxA, PFHpA, PFOA, and PFNA, while L-PFBS, br-PFHxS, and L-PFOS were the dominant PFASs. PFASs congeners are commonly produced through the electrochemical fluorination (ECF) method, which typically generates a mixture with a higher proportion of linear congeners compared to branched congeners (Benskin et al., 2010; Ma et al., 2018). In this study, L-PFOS and br-PFHxS were detected, while L-PFHxS and br-PFOS were not. These findings highlight the differing behaviors of linear and branched congeners in the environment and within organisms due to structural differences (Schulz et al., 2020). While prior studies have predominantly examined PFASs isomers in lakes, sediments, and rivers, limited research has focused on the distribution of PFOS and PFHxS isomers in the marine environment (Schulz et al., 2020). Further research on PFASs isomer profiles is essential to better understand their distribution patterns in seawater and to elucidate the physicochemical processes influencing isomer composition. The alternative compound Gen-X was detected exclusively in the YS and the ECS among the four sea areas. PFHxA and Gen-X have been widely

reported as common alternatives to PFOA in China (Feng et al., 2020; Zhong et al., 2021). In water samples from the SS, PFPeA, PFHxA, PFHpA, and PFOA were the detected PFCAs, while L-PFBS, L-PFOS, and L-PFNS were identified among the PFASs. In the ES, PFHxA, PFHpA, and PFOA were the primary PFCAs, with L-PFBS detected among the PFASs. PFOA was the prevalent substance in the seawater of the YS, whereas PFBS was the dominant compound in the ECS and ES. Despite being regulated as persistent organic pollutants, PFOA remained the most prevalent PFASs in seawater, suggesting it continues to be the primary PFASs released into the Korean coastal environment (Lee et al., 2020). This persistence may be attributed to the annual bulk load of PFOA discharged from the Yangtze River, estimated at 26.8 tons, accounting for over 80% of the total discharge in the coastal region (Du et al., 2022). These PFASs persist in the environment due to their recalcitrant nature and continue to be transported along ocean currents, remaining detectable in seawater.

3.5. Principal component analysis of PFASs compositions across water masses

PCA was conducted to examine the distribution of PFASs across water masses within the water column, based on the composition of individual compounds. The sampling sites and depths were grouped into four categories, each characterized by regionally dominant compounds

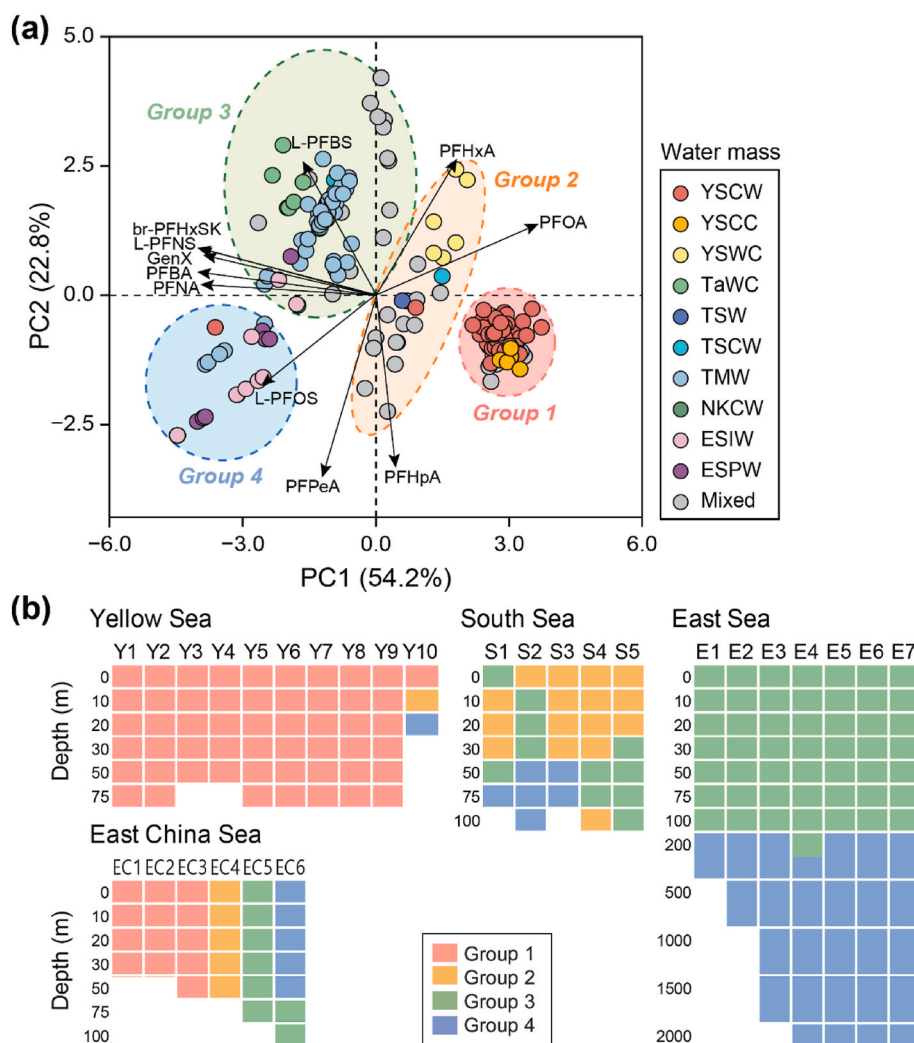


Fig. 4. (a) Principal component analysis (PCA) of PFAS compositions and water masses by water depth in the study area. (b) Distribution of PFAS groups by station and water depth within the study area based on PCA.

(Fig. 4a). Group 1 primarily included the YSCW, YSCC, and YSWC water masses and was distinguished by the presence of PFOA. PFOA is known to be a predominant compound in the Yangtze River during winter, suggesting that this group was significantly influenced by inflows from the Yangtze River (Zheng et al., 2017). Group 2 was characterized by the YSWC and mixed water masses. This group is characterized by short-chain compounds, such as PFHxA, PFHpA, and PFPeA, commonly used as PFOA replacements. YSWC is recognized as a tributary of the TsWC (Xu et al., 2009). Thus, this group likely reflects the influence of the TsWC, which transports water from the ECS continental shelf (Hase et al., 1999; Ito et al., 2014; Senjyu et al., 2006). Group 3 included the TMW, TSCW, and TaWC water masses and was characterized by short-chain and alternative compounds, such as L-PFBS, br-PFHxS, L-PFNS, Gen-X, and PFBA, which are used as replacements for PFOS and PFOA. Similar to Group 2, these water masses may have been influenced by the TsWC. Additionally, compounds from industrial complexes in the SS could have been transported northeastward along ocean currents. These findings support the trend that consumption patterns are shifting towards short-chain PFASs and alternatives (Cai et al., 2012; Zhou et al., 2013). Group 4 was characterized by the presence of L-PFOS and included the TMW, ESIW, and ESPW water masses. L-PFOS was the dominant compound in TMW, indicating the potential for its transport from Group 4 sites to deeper waters.

The groups exhibited distinct regional characteristics based on water depth within the study area (Fig. 4b). Group 1 was predominantly

associated with sites in the YS and ECS, spanning from the surface to the bottom waters. Group 2 was primarily linked to sites in the ECS and SS, extending from the surface to the middle waters, suggesting the potential movement of water from the YS and ECS into the SS.

Group 3 was mainly associated with sites in the middle waters of the SS and the surface waters of the ES, positioned below the Group 2 water. Group 4 was primarily linked to sites in the middle to deep waters of the ES. These findings suggest that PFASs are transported along the Tsushima Current, with water masses from Group 1 in the YS moving towards Groups 3 and 4 in the SS and ES. The ECS appears to be a mixing zone for PFASs from various sources. PCA is a powerful method for extracting principal components and reducing data dimensionality. In this study, the similarity in the composition of PFAAs and their relationship with water masses were successfully identified through the interpretation of each component. Future research could consider incorporating an ocean current transport model. This model could enhance the interpretation of principal components derived from PCA by providing insights into the transport pathway, distribution, mixing, and diffusion processes of PFASs, which is expected to improve the reliability of the study.

3.6. Comparison of PFASs in the water column in the regional seas of East Asian countries

PFASs are produced globally in fluoridation plants and are reported to be used in significant quantities (Du et al., 2022). Consequently,

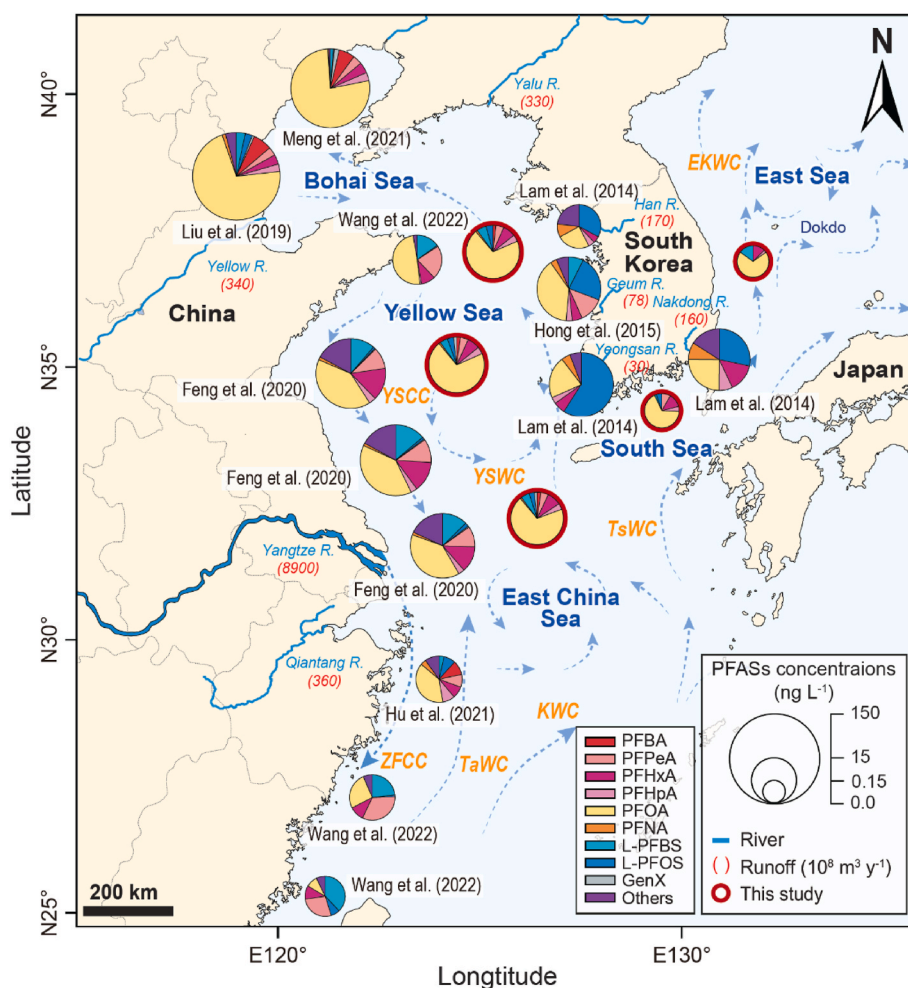


Fig. 5. Comparison of the concentrations, compositions, and major river runoff of PFASs in water samples collected from freshwater, coastal, and regional waters in China and South Korea [YSCC: Yellow Sea Coastal Current; YSWC: Yellow Sea Warm Current; KWC: Kuroshio Warm Current; ZFCC: Zhejiang-Fujian Coastal Current; TaWC: Taiwan Warm Current; TsWC: Tsushima Warm Current; EKWC: East Korean Warm Current (Du et al., 2022; Guo et al., 2006; Hu et al., 2011; Lee et al., 2009; MLTMA, 2011)].

numerous studies have been conducted worldwide to assess PFASs concentrations in seawater and sediments (Cheng et al., 2023; Zhong et al., 2021). In this study, we compared PFASs concentrations identified in the YS, ECS, SS, and ES with those reported in seawater and freshwater from rivers in the surrounding regions (Fig. 5 and Tables S12 and S13). The PFASs concentrations detected in surface seawater in the YS (mean: 9.9 ng L⁻¹), ECS (8.1 ng L⁻¹), SS (1.4 ng L⁻¹), and ES (0.97 ng L⁻¹) were below those recorded in the Geum River (19 ng L⁻¹), Nakdong River (26 ng L⁻¹), and Yeongsan River (19 ng L⁻¹). The elevated levels in these rivers can likely be attributed to the proximity of large coastal cities and residential areas (Lu et al., 2015). Coastal regions of the study area showed that PFASs discharged from rivers were diluted after entering the ocean, resulting in lower concentrations in seawater (Cai et al., 2012; Wang et al., 2019; Zheng et al., 2017).

Compared to findings from other regions, PFASs concentrations were reported as follows: Bohai Sea (mean: 150 ng L⁻¹), ECS (4.5 ng L⁻¹), South China Sea (2.6 ng L⁻¹), and Nantong Coast (26 ng L⁻¹) (Feng et al., 2020; Liu et al., 2019; Wang et al., 2022). These reported values surpass the concentrations observed in the current study. It is essential to emphasize that this research concentrated solely on PFASs concentrations, without considering the average annual discharge of nearby rivers. According to previous research, the Yangtze River is known to have the highest PFASs discharge flux among 91 estuarine rivers in China. With a mean annual discharge of $8900 \times 10^8 \text{ m}^3 \text{ y}^{-1}$, it is the largest river in the study area (Du et al., 2022). Although the PFASs concentration in the Yangtze River (82 ng L⁻¹) is moderate compared to most rivers when both concentration and discharge are considered, it may have significant impacts on the YS and ECS, which are focal points of this study (Du et al., 2022). In major rivers in Korea, L-PFOS and PFOA were the main compounds. PFASs in major rivers in Korea were studied in 2014 and 2015, before the regulation of PFOA was implemented, and may not have been reflected soon after the regulation of PFOS was implemented. In areas along the coast of China, such as the Bohai Sea, ECS, South China Sea, and Nantong Coast, PFOA and L-PFBS were the main compounds. PFASs in these areas were studied after 2019, and the results suggest that the production and use patterns of PFASs are shifting to short-chain PFASs due to the regulation of PFOS and PFOA, and PFOA is still the main pollutant.

As mentioned above, PFASs in seawater from the SS and ES may be influenced by water transported via the TsWC, a branch of the KWC. To further investigate PFASs movement about ocean currents, we conducted a PCA based on PFASs compositions identified in both previous studies and the current research (Fig. S8). The PCA results indicated that as water flows northward through the SS, ECS, and YS, it converges at sites in the YS and ECS, consistent with earlier observations. This analysis confirmed the northward flow of the TaWC and KWC, demonstrating that these currents transport water toward areas near the ECS, including Yancheng, Nantong, and Lianyungang. Previous study has reported that Sn, among metals, was transported by northeast-flowing ocean currents, showing a pattern similar to that of PFASs observed in this study (Um et al., 2024).

The YS was positioned negatively along PC1, indicating limited distribution compared to other sea areas. Its PFASs composition resembled that of the ECS at certain sites. The ECS, in turn, exhibited a PFASs composition similar to the Bohai Sea, likely due to the northward movement of water via the TaWC and KWC, which flow through the YSWC, pass through the Bohai Sea, and circulate back via the YSCC. As a result, the PFASs composition in the ECS aligns with some Bohai Sea sites. Additionally, the PFASs composition in the ECS extends into the SS and ES, with the distribution in the SS reaching the ES. This supports previous findings that the TsWC transports water from the continental shelf of the ECS southwestward to the ES via the Tsushima Strait (Hase et al., 1999; Ito et al., 2014; Senjyu et al., 2006). This study confirms that PFASs are transported from the ECS via the TsWC, through the SS, to the ES. Overall, the PCA of PFASs compositions from previous and current studies supports the role of ocean currents in PFASs distributions.

4. Conclusion

This study is the first to comprehensively assess PFASs, their precursors, and alternatives in the regional seas surrounding Korea. PFASs concentrations in seawater varied significantly across regions, with PFOA remaining the dominant contaminant despite global regulations. The detection of precursors and alternatives suggests a shift in PFASs consumption patterns. The areas with high concentrations of PFASs are likely associated with fluoropolymer production and industrial activities in coastal cities of China. PFASs discharged into the sea via rivers were gradually diluted as they dispersed into the open ocean, leading to lower concentrations in seawater. PCA of PFASs compositions across various water masses showed that ocean currents play a significant role in shaping the distribution patterns of PFASs in the waters surrounding Korea. This suggests that oceanic transport mechanisms may contribute to the long-range movement of these compounds, indicating the potential for these persistent contaminants to spread across the global marine environment rather than being confined to local sources. Further research is needed to establish baseline data, monitor temporal trends, and inform future research and management efforts.

CRediT authorship contribution statement

Sunmi Yang: Writing – original draft, Visualization, Investigation, Formal analysis, Data curation, Conceptualization. **Jiyun Gwak:** Visualization, Investigation, Formal analysis. **Mungi Kim:** Methodology, Formal analysis. **Jihyun Cha:** Formal analysis. **Younghnam Kim:** Formal analysis, Data curation. **Yeonjung Lee:** Writing – review & editing, Formal analysis. **Hyo-Bang Moon:** Writing – review & editing, Formal analysis, Data curation. **Seongjin Hong:** Writing – review & editing, Visualization, Supervision, Project administration, Investigation, Funding acquisition, Formal analysis, 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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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2024.144042>.

Data availability

Data will be made available on request.

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<Supplementary Materials>

**Spatial and vertical distribution of per- and polyfluoroalkyl substances
(PFASs) in the water columns of the regional seas of South Korea**

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Supplementary Tables

Table S1. Information on sampling sites, including time, location, seawater collection depth, and bottom depth.

Regional sea	Station	Year	Month	Latitude	Longitude	Sample depth (m)	Bottom depth (m)
Yellow Sea	Y1	2024	1	37° 00.07 N	124° 00.37 E	0, 10, 20, 30, 50, 72	72
	Y2	2024	1	36° 59.57 N	124° 29.52 E	0, 10, 20, 30, 50, 74	74
	Y3	2024	1	34° 59.54 N	122° 04.17 E	0, 10, 20, 30, 47	47
	Y4	2024	1	34° 59.58 N	122° 30.03 E	0, 10, 20, 30, 60	60
	Y5	2024	1	34° 59.58 N	122° 59.57 E	0, 10, 20, 30, 50, 68	68
	Y6	2024	1	34° 59.53 N	123° 29.54 E	0, 10, 20, 30, 50, 72	72
	Y7	2024	1	34° 59.56 N	123° 59.55 E	0, 10, 20, 30, 50, 75	75
	Y8	2024	1	34° 59.59 N	124° 29.59 E	0, 10, 20, 30, 50, 75, 87	87
	Y9	2024	1	34° 59.52 N	124° 59.59 E	0, 10, 20, 30, 50, 83	83
	Y10	2024	1	35° 00.02 N	125° 40.45 E	0, 10, 27	27
East China Sea	EC1	2024	1	32° 10.48 N	123° 59.59 E	0, 10, 20, 30, 37	37
	EC2	2024	1	32° 12.28 N	124° 30.02 E	0, 10, 20, 38	38
	EC3	2024	1	32° 10.50 N	124° 59.54 E	0, 10, 20, 30, 47	47
	EC4	2024	1	32° 11.45 N	125° 30.02 E	0, 10, 20, 30, 50, 66	66
	EC5	2024	1	32° 11.46 N	125° 59.22 E	0, 10, 20, 30, 50, 87	87
	EC6	2024	1	32° 11.01 N	126° 30.14 E	0, 10, 20, 30, 50, 75, 103	103
South Sea	S1	2023	5	33° 51.05 N	127° 38.30 E	5, 10, 20, 30, 50, 75	75
	S2	2023	5	33° 47.57 N	127° 02.30 E	5, 10, 20, 30, 50, 75, 100, 110	110
	S3	2023	5	34° 23.47 N	128° 24.20 E	5, 10, 20, 30, 50, 70	70
	S4	2023	5	34° 41.53 N	128° 52.08 E	5, 10, 20, 30, 50, 75, 85	85
	S5	2023	5	34° 57.41 N	129° 20.29 E	5, 10, 20, 30, 50, 75, 95	95
East Sea	E1	2023	2	37° 00.38 N	129° 40.39 E	0, 10, 20, 30, 50, 75, 100, 150, 160	160
	E2	2023	2	37° 00.37 N	129° 49.36 E	0, 10, 20, 30, 50, 75, 100, 150, 200, 500, 530	530
	E3	2023	2	37° 00.38 N	130° 00.00 E	0, 10, 20, 30, 50, 75, 100, 200, 500, 1000, 1500, 1690	1690
	E4	2023	2	36° 59.58 N	130° 20.38 E	0, 10, 20, 30, 50, 75, 100, 200, 500, 1000, 1500, 2000, 2150	2150
	E5	2023	2	37° 00.02 N	130° 40.00 E	0, 10, 20, 30, 50, 75, 100, 200, 500, 1000, 1500, 2165	2165
	E6	2023	2	37° 00.38 N	131° 00.01 E	0, 10, 20, 30, 50, 75, 100, 200, 500, 1000, 1500, 2145	2145
	E7	2023	2	35° 23.03 N	131° 10.04 E	0, 10, 20, 30, 50, 75, 100, 200, 500, 1000, 1500, 2142	2142

Table S2. Information on sampling sites, including location, bottom depth, sampling depth, seawater temperature, and salinity.

Sampling sites	Longitude (°E)	Latitude (°N)	Bot. depth [m]	Sampling depth [m]	Temperature [°C]	Salinity
Y1	124.0105	37.00204	72	0	8.5598	32.4601
Y1	124.01052	37.00204	72	10	8.564	32.4657
Y1	124.01052	37.00204	72	20	8.5661	32.4662
Y1	124.01052	37.00204	72	30	8.5639	32.4658
Y1	124.01052	37.00204	72	50	9.1808	32.6411
Y1	124.0105	37.00204	72	72	9.1898	32.6501
Y2	124.4978	36.99928	74	0	8.8361	32.626
Y2	124.4978	36.99928	74	10	8.8377	32.6258
Y2	124.4978	36.99928	74	20	8.838	32.6251
Y2	124.4978	36.9993	74	30	8.8371	32.6254
Y2	124.4978	36.99928	74	50	8.846	32.6258
Y2	124.4978	36.9993	74	73	8.8523	32.6264
Y3	122.0716	34.99838	47	0	9.5904	31.5682
Y3	122.0716	34.99838	47	10	9.5921	31.5729
Y3	122.0716	34.99838	47	20	9.596	31.5763
Y3	122.0716	34.99836	47	30	9.5979	31.5844
Y3	122.0716	34.99838	47	46	9.5846	31.5847
Y4	122.5011	34.99954	60	0	10.2907	31.7882
Y4	122.5011	34.99954	60	10	10.2941	31.7884
Y4	122.5011	34.99954	60	20	10.2903	31.7868
Y4	122.5011	34.99954	60	30	10.2978	31.7873
Y4	122.5011	34.99954	60	59	10.3212	31.794
Y5	122.9993	34.99956	68	0	10.9846	32.208
Y5	122.9993	34.99956	68	10	11.0157	32.2172
Y5	122.9993	34.99956	68	20	11.0613	32.2316
Y5	122.9993	34.99956	68	30	11.2019	32.2796
Y5	122.9993	34.99954	68	50	11.266	32.3051
Y5	122.9993	34.99956	68	68	11.6156	32.4603
Y6	123.4986	34.99806	72	0	11.6091	32.4859
Y6	123.4986	34.99806	72	10	11.6126	32.4863
Y6	123.4986	34.99806	72	20	11.6137	32.4858
Y6	123.4986	34.99808	72	30	11.6283	32.4907
Y6	123.4986	34.99806	72	50	11.775	32.5419
Y6	123.4986	34.99805	72	72	12.5046	32.8289
Y7	123.9986	34.99907	75	0	11.1131	32.4387
Y7	123.9986	34.99908	75	10	11.1124	32.438
Y7	123.9986	34.99908	75	20	11.1147	32.4392
Y7	123.9986	34.99906	75	30	11.1325	32.4454
Y7	123.9986	34.99906	75	50	11.1988	32.4699
Y7	123.9986	34.99904	75	75	11.486	32.579
Y8	124.4998	34.99982	87	0	11.8256	32.7134
Y8	124.4998	34.99984	87	10	11.8269	32.714
Y8	124.4998	34.99984	87	20	11.8586	32.7187
Y8	124.4998	34.99984	87	30	11.8507	32.7164
Y8	124.4998	34.99986	87	50	11.8537	32.7191
Y8	124.4998	34.99986	87	75	11.9639	32.7528
Y8	124.4998	34.99986	87	87	11.9423	32.7497
Y9	124.9997	34.99778	83	0	10.8822	32.6457
Y9	124.9997	34.99779	83	10	10.884	32.6462
Y9	124.9997	34.9978	83	20	10.8821	32.6461

Y9	124.9997	34.99782	83	30	10.887	32.6466
Y9	124.9997	34.99782	83	50	10.887	32.6467
Y9	124.9997	34.9978	83	83	10.9407	32.6744
Y10	125.6793	35.00072	27	0	6.6575	31.8751
Y10	125.6793	35.00072	27	10	6.6632	31.8771
Y10	125.6793	35.00072	27	27	7.4649	32.0677
EC1	123.9999	32.1801	37	0	11.5739	31.446
EC1	123.9999	32.1801	37	10	11.5752	31.4477
EC1	123.9999	32.1801	37	20	11.5841	31.4493
EC1	123.9999	32.18008	37	30	11.5893	31.4503
EC1	123.9999	32.1801	37	37	11.5926	31.45
EC2	124.5007	32.20782	38	0	11.651	31.5631
EC2	124.5007	32.20782	38	10	11.6589	31.5628
EC2	124.5007	32.2078	38	20	11.6656	31.5631
EC2	124.5007	32.20784	38	38	11.6727	31.5672
EC3	124.9986	32.18066	47	0	12.4274	32.1844
EC3	124.9986	32.18066	47	10	12.4451	32.1859
EC3	124.9986	32.18066	47	20	12.4429	32.1869
EC3	124.9986	32.18066	47	30	12.521	32.2272
EC3	124.9986	32.18066	47	47	12.8622	32.4211
EC4	125.5007	32.19608	66	0	15.163	33.5543
EC4	125.5007	32.19608	66	10	15.1643	33.5551
EC4	125.5007	32.19608	66	20	15.1647	33.5559
EC4	125.5007	32.19608	66	30	15.1885	33.5663
EC4	125.5007	32.19608	66	50	15.2263	33.5841
EC4	125.5007	32.19608	66	66	15.2176	33.5828
EC5	125.9895	32.19614	87	0	16.782	33.934
EC5	125.9895	32.19614	87	10	16.7805	33.9337
EC5	125.9895	32.19612	87	20	16.7718	33.9362
EC5	125.9895	32.19614	87	30	16.7717	33.9381
EC5	125.9895	32.19614	87	50	16.7888	33.9519
EC5	125.9895	32.19614	87	87	16.8892	34.2036
EC6	126.504	32.18362	103	0	17.9342	34.267
EC6	126.504	32.18362	103	10	17.9459	34.2657
EC6	126.504	32.18362	103	20	17.9504	34.267
EC6	126.504	32.18362	103	30	17.9481	34.2658
EC6	126.504	32.18362	103	50	17.9518	34.2657
EC6	126.504	32.18362	103	75	17.9592	34.267
EC6	126.504	32.18362	103	103	16.5213	34.435
S1	127.6417	33.85	75	5	18.6512	33.9273
S1	127.6417	33.85	75	10	17.565	33.6498
S1	127.6417	33.85	75	20	15.4094	33.4769
S1	127.6417	33.85	75	30	15.2196	33.6984
S1	127.6417	33.85	75	50	15.5815	34.1694
S1	127.6417	33.85	75	75	15.6305	34.4549
S2	127.0417	33.79944	110	5	18.9186	33.6577
S2	127.0417	33.79944	110	10	17.7781	33.8279
S2	127.0417	33.79944	110	20	17.0582	34.114
S2	127.0417	33.79944	110	30	16.8627	34.2145
S2	127.0417	33.79944	110	50	16.9431	34.3411
S2	127.0417	33.79944	110	75	16.4311	34.4285
S2	127.0417	33.79944	110	100	15.8778	34.4294
S2	127.0417	33.79944	110	110	15.8501	33.5796
S3	128.4056	34.39639	70	5	16.9663	33.595
S3	128.4056	34.39639	70	10	16.5265	33.5989

S3	128.4056	34.39639	70	20	14.717	33.8167
S3	128.4056	34.39639	70	30	14.443	34.2634
S3	128.4056	34.39639	70	50	15.254	34.3961
S3	128.4056	34.39639	70	70	15.4519	33.5991
S4	128.8689	34.69833	85	5	15.783	33.7531
S4	128.8689	34.69833	85	10	15.6465	33.7623
S4	128.8689	34.69833	85	20	15.1199	33.903
S4	128.8689	34.69833	85	30	14.5797	33.8183
S4	128.8689	34.69833	85	50	13.8903	33.9777
S4	128.8689	34.69833	85	75	13.5991	33.9657
S4	128.8689	34.69833	85	85	13.6002	33.9643
S5	129.3414	34.96139	95	5	17.2049	33.7211
S5	129.3414	34.96139	95	10	16.7481	33.7611
S5	129.3414	34.96139	95	20	15.4365	33.7401
S5	129.3414	34.96139	95	30	14.7742	33.7774
S5	129.3414	34.96139	95	50	14.4139	33.8548
S5	129.3414	34.96139	95	75	14.7977	34.2839
S5	129.3414	34.96139	95	95	14.7311	34.2879
E1	129.6775	37.01056	160	0	11.016	34.8004
E1	129.6775	37.01056	160	10	12.1535	33.782
E1	129.6775	37.01056	160	20	12.1733	34.1796
E1	129.6775	37.01056	160	30	12.1811	34.1787
E1	129.6775	37.01056	160	50	12.1852	34.1784
E1	129.6775	37.01056	160	75	12.1533	34.2212
E1	129.6775	37.01056	160	100	10.6317	34.1703
E1	129.6775	37.01056	160	150	4.1296	34.0638
E1	129.6775	37.01056	160	160	4.0586	34.0606
E2	129.8269	37.01028	530	0	12.1281	35.8833
E2	129.8269	37.01028	530	10	11.9384	33.431
E2	129.8269	37.01028	530	20	11.9404	34.2002
E2	129.8269	37.01028	530	30	11.9404	34.1999
E2	129.8269	37.01028	530	50	11.9354	34.2012
E2	129.8269	37.01028	530	75	11.8389	34.1903
E2	129.8269	37.01028	530	100	9.5707	34.1148
E2	129.8269	37.01028	530	150	3.6535	34.0553
E2	129.8269	37.01028	530	200	1.7869	34.0406
E2	129.8269	37.01028	530	500	0.694	34.0579
E2	129.8269	37.01028	530	530	0.6932	34.0579
E3	130	37.01056	1690	0	12.2558	33.8874
E3	130	37.01056	1690	10	12.0555	34.188
E3	130	37.01056	1690	20	12.0638	34.1975
E3	130	37.01056	1690	30	12.0675	34.197
E3	130	37.01056	1690	50	12.0371	34.2046
E3	130	37.01056	1690	75	11.6356	34.179
E3	130	37.01056	1690	100	10.7219	34.1495
E3	130	37.01056	1690	200	2.235	34.0472
E3	130	37.01056	1690	500	0.6417	34.0602
E3	130	37.01056	1690	1000	0.3197	34.0698
E3	130	37.01056	1690	1500	0.2362	34.0696
E3	130	37.01056	1690	1690	0.237	34.0697
E4	130.3439	36.99972	2150	0	12.3082	34.1182
E4	130.3439	36.99972	2150	10	12.1942	33.4209
E4	130.3439	36.99972	2150	20	12.2968	34.1806
E4	130.3439	36.99972	2150	30	12.299	34.1805
E4	130.3439	36.99972	2150	50	12.2466	34.1832

E4	130.3439	36.99972	2150	75	12.0214	34.1918
E4	130.3439	36.99972	2150	100	11.9675	34.1926
E4	130.3439	36.99972	2150	200	4.4083	34.091
E4	130.3439	36.99972	2150	500	0.7452	34.0558
E4	130.3439	36.99972	2150	1000	0.3707	34.0699
E4	130.3439	36.99972	2150	1500	0.2663	34.0698
E4	130.3439	36.99972	2150	2000	0.2406	34.0702
E4	130.3439	36.99972	2150	2150	0.2435	34.0703
E5	130.6669	37.00083	2165	0	12.6652	35.19
E5	130.6669	37.00083	2165	10	12.2428	32.764
E5	130.6669	37.00083	2165	20	12.4658	34.2315
E5	130.6669	37.00083	2165	30	12.4679	34.2318
E5	130.6669	37.00083	2165	50	12.4603	34.2378
E5	130.6669	37.00083	2165	75	12.4304	34.2435
E5	130.6669	37.00083	2165	100	12.17	34.2129
E5	130.6669	37.00083	2165	200	4.2789	34.0675
E5	130.6669	37.00083	2165	500	0.7412	34.0568
E5	130.6669	37.00083	2165	1000	0.3747	34.0701
E5	130.6669	37.00083	2165	1500	0.2692	34.0698
E5	130.6669	37.00083	2165	2165	0.2482	34.0703
E6	131.0003	37.01083	2145	0	12.1322	33.9228
E6	131.0003	37.01083	2145	10	12.0169	32.8976
E6	131.0003	37.01083	2145	20	12.0959	34.2188
E6	131.0003	37.01083	2145	30	12.0777	34.2193
E6	131.0003	37.01083	2145	50	12.0692	34.2185
E6	131.0003	37.01083	2145	75	11.9776	34.2076
E6	131.0003	37.01083	2145	100	11.7883	34.2007
E6	131.0003	37.01083	2145	200	3.3026	34.0353
E6	131.0003	37.01083	2145	500	0.7004	34.0593
E6	131.0003	37.01083	2145	1000	0.3342	34.0698
E6	131.0003	37.01083	2145	1500	0.2432	34.0696
E6	131.0003	37.01083	2145	2145	0.2318	34.0703
E7	131.1678	37.00056	2142	0	12.1144	33.9991
E7	131.1678	37.00056	2142	10	12.0385	34.1964
E7	131.1678	37.00056	2142	20	12.0426	34.1966
E7	131.1678	37.00056	2142	30	12.0314	34.1982
E7	131.1678	37.00056	2142	50	12.0139	34.1997
E7	131.1678	37.00056	2142	75	12.0067	34.1989
E7	131.1678	37.00056	2142	100	11.8489	34.1836
E7	131.1678	37.00056	2142	200	2.4147	34.0216
E7	131.1678	37.00056	2142	500	0.7184	34.059
E7	131.1678	37.00056	2142	1000	0.3089	34.0699
E7	131.1678	37.00056	2142	1500	0.2409	34.0701
E7	131.1678	37.00056	2142	2142	0.2341	34.0702

Table S3. Information on target compounds, CAS numbers, and analytical standards used in this study.

Compounds	Acronym	Analytical standard	CAS number	Supplier, purity, and concentration	
PFCAs	PFBA	Perfluoro-n-butanoic acid	375-22-4	PFAC-MXH (mixture)	
	PFPeA	Perfluoro-n-pentanoic acid	2706-90-3	Wellington Laboratories >98%	
	PFHxA	Perfluoro-n-hexanoic acid	307-24-4	1000-4000 ng/mL \pm 5% of the single compounds	
	PFHpA	Perfluoro-n-heptanoic acid	375-85-9		
	PFOA	Perfluoro-n-octanoic acid	335-67-1		
	PFNA	Perfluoro-n-nonanoic acid	375-95-1		
	PFDA	Perfluoro-n-decanoic acid	335-76-2		
	PFUnDA	Perfluoro-n-undecanoic acid	2058-94-8		
	PFDoDA	Perfluoro-n-dodecanoic acid	307-55-1		
	PFTriDA	Perfluoro-n-tridecanoic acid	72629-94-8		
	PFTeDA	Perfluoro-n-tetradecanoic acid	376-06-7		
	PFSAs	L-PFBS	Potassium perfluoro-1-butanefulfonate	29420-49-3	PFAC-MXH (mixture)
		L-PFPeS	Sodium perfluoro-1-pentanesulfonate	630402-22-1	Wellington Laboratories >98%
				1000-4000 ng/mL \pm 5% of the single compounds	
L-PFHxS		Sodium perfluoro-1-hexanesulfonate	82382-12-5	L-PFHxS	
				Wellington Laboratories >98 %	
				50 \pm 2.5 μ g/mL	
br-PFHxS		Potassium perfluorohexanesulfonate (linear and branched isomers)	3871-99-6	PFAC-MXH (mixture)	
				Wellington Laboratories >98%	
				1000-4000 ng/mL \pm 5% of the single compounds	
L-PFOS		Sodium perfluoro-1-octanesulfonate	4021-47-0	L-PFOS	
			Wellington Laboratories >98 %		
			50 \pm 2.5 μ g/mL		
br-PFOS	Potassium perfluorooctanesulfonate (linear and branched isomers)	2795-39-3	PFAC-MXH (mixture)		
			Wellington Laboratories >98%		
L-PFHpS	Sodium perfluoro-1-heptanesulfonate	21934-50-9	1000-4000 ng/mL \pm 5% of the single compounds		
L-PFNS	Sodium perfluoro-1-nonanesulfonate	98789-57-2			
L-PFDS	Sodium perfluoro-1-decanesulfonate	2806-15-7			
Precursors	N-MeFOSAA	N-methylperfluorooctanesulfonamidoacetic acid (linear and branched isomers)	2355-31-9	PFAC-MXH (mixture)	
				Wellington Laboratories >98%	
	N-EtFOSAA	N-ethylperfluorooctanesulfonamidoacetic acid (linear and branched isomers)	1336-61-4	1000-4000 ng/mL \pm 5% of the single compounds	
	4:2 FTS	Sodium 1H, 1H, 2H, 2H-perfluorohexanesulfonate	27619-93-8		
	6:2 FTS	Sodium 1H, 1H, 2H, 2H-perfluorooctanesulfonate	27619-94-9		

	8:2 FTS	Sodium 1H, 1H, 2H, 2H-perfluorodecanesulfonate	27619-96-1	
Alternatives	ADONA	Sodium dodecafluoro-3H-4, 8-dioxananoate	958445-44-8	NaDONA Wellington Laboratories >98 % 50 ± 2.5 µg/mL
	Gen-X	2,3,3,3-Tetrafluoro-2-(1,1,2,2,3,3,3-heptafluoropropoxy)-propanoic acid	13252-13-6	HFPO-DA (trade name: Gen-X Wellington Laboratories >98 % 50 ± 2.5 µg/mL
	F53B	Potassium 9-chlorohexadecafluoro-3-oxanonane-1-sulfonate	73606-19-6	9Cl-PF3ONS Wellington Laboratories >98 % 50 ± 2.5 µg/mL
Internal standards	MPFDA	Perfluoro-n-[1,2- ¹³ C ₂]decanoic acid	960315-50-8	MPFAC-MXA (mixture)
	MPFDoDA	Perfluoro-n-[1,2- ¹³ C ₂]dodecanoic acid	960315-52-0	Wellington Laboratories >98%
	MPFHxA	Perfluoro-n-[1,2- ¹³ C ₂]hexanoic acid	307-24-4	2000 ng/mL ± 5% of the single compounds
	MPFNA	Perfluoro-n-[1,2,3,4,5- ¹³ C ₅]nonanoic acid	960315-49-5	
	MPFOA	Perfluoro-n-[1,2,3,4- ¹³ C ₄]octanoic acid	960315-48-4	
	MPFUdA	Perfluoro-n-[1,2- ¹³ C ₂]undecanoic acid	960315-51-9	

Table S4. Chemicals used for sample preparation, cleaning, and analysis.

Chemical	CAS number	Specification	Supplier
Methanol	67-56-1	for HPLC, $\geq 99.9\%$	Honeywell Fluka, USA
Milli-Q water		Milli-Q Integral 5 system, purified by SPE	Merck, Germany
Acetic acid	64-19-7	glacial, $\geq 99.99\%$ trace metals basis	Merck, Germany
Ammonium acetate	631-61-8	For mass spectrometry, eluent additive for LC-MS	Merck, Germany
Ammonium hydroxide solution	1336-21-6	ACS reagent, 28.0-30.0% NH ₃ basis	Merck, Germany

Table S5. Instrumental conditions for the analysis of 28 per- and polyfluoroalkyl substances (PFASs) in seawater using HPLC-MS/MS.

Instrument	HPLC: Agilent Infinity 1290 II		
	MS/MS: Agilent 6470 triple quadrupole mass spectrometer		
Column	Waters Acquity UPLC BEH Shield RP ₁₈ , 2.1 x 100 mm, 1.7 μ m		
Column temperature	30 °C		
Mobile phase	(A): Water; (B): 80:20 MeOH:ACN (both with 10mM NH ₄ Ac buffer)		
Mobile phase gradient		Mobile phase	
	Time (min)	A (%)	B (%)
	0.0	70	30
	6.0	10	90
	7.0	10	90
	7.5	70	30
	9.0	70	30
Injection volume	5 μ L		
Flow rate	0.3 mL min ⁻¹		
Ion source	ESI (electrospray ionization)		
Polarity	Negative		
Ion spray voltage	2500 V (negative)		
Gas temperature	230 °C		
Sheath gas temperature	250 °C		
Nebulizer gas	N ₂ (15 psi)		
Sheath gas flow	12 L/min		

Table S6. Optimization of compound-specific parameters in a tandem mass spectrometer for the analysis of 28 PFASs and 6 internal standards.

Compounds	Molecular weight	RT (min)	Precursor ion (m/z)	Product ion (m/z)	Fragmentor (volts)	CE (volts)
<i>PFCA</i>s						
PFBA	212.97	2.129	213.0 [M-H] ⁻	168.9	80	5
PFPeA	262.97	3.174	263.0 [M-H] ⁻	218.9	70	5
PFHxA	312.97	4.047	313.0 [M-H] ⁻	118.8	60	25
				268.8	60	5
PFHpA	362.96	4.714	363.0 [M-H] ⁻	168.9	70	15
				318.8	70	5
PFOA	412.96	5.249	413.0 [M-H] ⁻	168.7	70	10
				368.8	70	5
PFNA	462.96	5.684	463.0 [M-H] ⁻	168.6	80	20
				418.9	80	5
PFDA	512.96	6.047	513.0 [M-H] ⁻	218.7	80	15
				468.9	80	5
PUnDA	562.95	6.357	563.0 [M-H] ⁻	268.9	80	10
				518.8	80	5
PDoDA	612.95	6.621	613.0 [M-H] ⁻	318.6	90	10
				568.8	90	5
PTriDA	662.95	6.852	663.0 [M-H] ⁻	219.1	90	20
				618.7	90	10
PTeDA	712.94	7.043	712.9 [M-H] ⁻	368.9	120	20
				668.9	120	10
<i>PFSA</i>s						
L-PFBS	298.94	3.551	298.9 [M-H] ⁻	80	130	40
				98.9	130	35
L-PFPeS	348.93	4.317	348.9 [M-H] ⁻	79.8	130	40
				98.5	130	35
L-PFHxS	398.93	4.746	398.9 [M-H] ⁻	79.8	140	40
				98.8	140	35
br-PFHxS	398.93	4.899	398.9 [M-H] ⁻	79.8	130	40
				98.7	130	40
L-PFOS	498.93	5.638	498.9 [M-H] ⁻	79.8	120	40
				98.6	120	40
	498.93	5.766	498.9 [M-H] ⁻	79.9	110	40
br-PFOS						
L-PFHpS	448.93	5.374	448.9 [M-H] ⁻	79.8	90	40
				98.8	90	40
L-PFNS	548.92	6.106	548.9 [M-H] ⁻	79.8	150	40
				98.8	150	40
L-PFDS	598.92	6.403	598.9 [M-H] ⁻	79.8	150	40
<i>Precursors</i>						
N-MeFOSAA	569.96	6.165	570.0 [M-H] ⁻	419.0	130	15
N-EtFOSAA	583.98	6.324	584.0 [M-H] ⁻	418.8	140	15
4:2 FTS	326.97	3.874	327.0 [M-H] ⁻	80.8	110	20
				306.0	110	20
6:2 FTS	426.96	5.155	427.0 [M-H] ⁻	81.1	120	30
				407.0	120	20
8:2 FTS	526.96	5.994	527.0 [M-H] ⁻	81.1	160	30
				506.8	160	25
<i>Alternatives</i>						
ADONA	376.96	4.799	377.0 [M-H] ⁻	84.8	70	20
				250.8	70	10

Gen-X	328.96	4.311	329.0 [M-H] ⁻	168.6	60	5
				285.0	60	5
F53B	530.89	5.994	530.9 [M-H] ⁻	350.3	130	25
<i>Internal standards</i>						
MPFDA	514.97	6.047	515.0 [M-H] ⁻	468.9	90	5
				514.8	90	5
MPFDoDA	614.96	6.621	615.0 [M-H] ⁻	268.9	100	20
				569.9	100	5
MPFHxA	314.98	4.047	315.0 [M-H] ⁻	269.9	70	5
				314.9	70	5
MPFNA	468.00	5.684	468.0 [M-H] ⁻	222.7	80	10
				422.8	80	5
MPFOA	416.99	5.248	417.0 [M-H] ⁻	171.8	80	20
				371.8	80	5
MPFUdA	564.97	6.357	565.0 [M-H] ⁻	318.7	90	15
				519.8	90	5

Table S7. Linear range, coefficient of determination (R^2), limit of detection (LOD), limit of quantification (LOQ), procedural blank concentration, and field blank concentration for the analysis of 28 PFASs using LC-MS/MS.

Compounds	Linear range (ng mL ⁻¹)	R ²	LOD (ng mL ⁻¹)	LOQ (ng mL ⁻¹)	Procedural blank concentration (ng mL ⁻¹)	Field blank concentration (ng mL ⁻¹)
PFBA	0.1–50	0.992	0.07	0.23	<LOD ^a	<LOD
PFPeA	0.1–50	0.997	0.02	0.08	<LOD	<LOD
PFHxA	0.1–50	0.998	0.01	0.05	<LOD	<LOD
PFHpA	0.1–50	0.998	0.01	0.03	<LOD	<LOD
PFOA	0.1–50	0.995	0.02	0.07	<LOD	<LOD
PFNA	0.1–50	0.997	0.02	0.06	<LOD	<LOD
PFDA	0.1–50	0.997	0.02	0.05	<LOD	<LOD
PFUnDA	0.1–50	0.996	0.01	0.04	<LOD	<LOD
PFDoDA	0.1–50	0.998	0.02	0.05	<LOD	<LOD
PFTriDA	0.1–50	0.998	0.01	0.05	<LOD	<LOD
PFTeDA	0.1–50	0.999	0.03	0.08	<LOD	<LOD
L-PFBS	0.1–50	0.999	0.03	0.10	<LOD	<LOD
L-PFPeS	0.1–50	0.998	0.02	0.07	<LOD	<LOD
L-PFHxS	0.1–50	0.995	0.07	0.22	<LOD	<LOD
br-PFHxS	0.1–50	0.996	0.03	0.10	<LOD	<LOD
L-PFOS	0.1–50	0.999	0.03	0.08	<LOD	<LOD
br-PFOS	0.1–50	0.997	0.03	0.10	<LOD	<LOD
L-PFHpS	0.1–50	0.999	0.04	0.14	<LOD	<LOD
L-PFNS	0.1–50	0.999	0.05	0.15	<LOD	<LOD
L-PFDS	0.1–50	0.990	0.06	0.19	<LOD	<LOD
N-MeFOSAA	0.1–50	0.999	0.04	0.14	<LOD	<LOD
N-EtFOSAA	0.1–50	0.997	0.07	0.21	<LOD	<LOD
4:2 FTS	0.1–50	0.998	0.18	0.57	<LOD	<LOD
6:2 FTS	0.1–50	0.997	0.14	0.45	<LOD	<LOD
8:2 FTS	0.1–50	0.993	0.10	0.33	<LOD	<LOD
ADONA	0.1–50	0.998	0.01	0.03	<LOD	<LOD
Gen-X	0.1–50	0.999	0.06	0.19	<LOD	<LOD
F53B	0.1–50	0.999	0.05	0.17	<LOD	<LOD

^a <LOD: Below limit of detected.

Table S8. Average recovery rate and standard deviation of internal standards in seawater samples.

Compound	Recovery rates (% , n = 204)	
	Mean	SD^a
MPFDA	111	11
MPFDoA	87	12
MPFNA	100	9.4
MPFOA	107	8.7
MPFUdA	101	9.9
MPFHxA	105	11

^a SD: standard deviation.

Table S9. Average recovery rate and standard deviation of internal standards in procedural blanks.

Compound	Recovery rates (% , n = 9)	
	Mean	SD^a
MPFDA	110	11
MPFDoA	107	12
MPFNA	107	7.1
MPFOA	113	13
MPFUdA	110	11
MPFHxA	107	7.9

^a SD: standard deviation.

Table S10. Average recovery rate and standard deviation of internal standards in field blanks.

Compound	Recovery rates (% , n = 9)	
	Mean	SD ^a
MPFDA	113	6.4
MPFDoA	110	14
MPFNA	103	5.9
MPFOA	112	8.6
MPFUdA	111	6.8
MPFHxA	103	10

^a SD: standard deviation.

Table S11. Concentration range and detection rates of 28 PFASs in seawater from the YS, ECS, SS, and ES.

Compounds	YS			ECS			SS			ES		
	DR (%) ^a	Min to Max (ng L ⁻¹)	Mean (ng L ⁻¹)	DR (%)	Min to Max (ng L ⁻¹)	Mean (ng L ⁻¹)	DR (%)	Min to Max (ng L ⁻¹)	Mean (ng L ⁻¹)	DR (%)	Min to Max (ng L ⁻¹)	Mean (ng L ⁻¹)
<i>PFCA</i> s												
PFBA	79	<LOQ ^b to 0.50	0.25	39	<LOQ to 0.51	0.15	0	- ^c	-	0	-	-
PFPeA	98	<LOQ to 0.79	0.46	64	<LOQ to 0.79	0.27	56	<LOQ to 0.18	0.06	0	-	-
PFHxA	98	<LOQ to 1.0	0.72	100	0.10 to 1.3	0.54	85	<LOQ to 0.26	0.14	68	<LOQ to 0.18	0.08
PFHpA	98	<LOQ to 0.67	0.39	76	<LOQ to 0.65	0.23	47	<LOQ to 0.12	0.04	9	<LOQ to 0.09	0.01
PFOA	100	0.19 to 12	6.8	100	0.16 to 12	4.3	100	0.08 to 1.5	0.73	81	<LOQ to 0.79	0.47
PFNA	84	<LOQ to 0.20	0.09	42	<LOQ to 0.17	0.05	0	-	-	0	-	-
PFDA	0	-	-	0	-	-	0	-	-	0	-	-
PFUnDA	0	-	-	0	-	-	0	-	-	0	-	-
PFDoDA	0	-	-	0	-	-	0	-	-	0	-	-
PFTriDA	0	-	-	0	-	-	0	-	-	0	-	-
PFTeDA	0	-	-	0	-	-	0	-	-	0	-	-
<i>PFS</i> A												
L-PFBS	98	<LOQ to 0.69	0.47	82	<LOQ to 0.65	0.31	12	<LOQ to 0.16	0.02	65	<LOQ to 0.39	0.10
L-PFPeS	0	-	-	0	-	-	0	-	-	0	-	-
L-PFHxS	0	-	-	0	-	-	0	-	-	0	-	-
br-PFHxS	2	<LOQ to 0.12	-	0	-	-	0	-	-	0	-	-
L-PFOS	98	<LOQ to 0.64	0.35	58	<LOQ to 0.63	0.18	21	<LOQ to 0.22	0.03	0	-	-
br-PFOS	0	-	-	0	-	-	0	-	-	0	-	-
L-PFHpS	0	-	-	0	-	-	0	-	-	0	-	-
L-PFNS	0	-	-	0	-	-	6	<LOQ to 0.96	0.05	0	-	-
L-PFDS	0	-	-	0	-	-	0	-	-	0	-	-
<i>Precursors</i>												
N-MeFOSAA	0	-	-	0	-	-	0	-	-	0	-	-
N-EtFOSAA	0	-	-	0	-	-	0	-	-	0	-	-
4:2 FTS	0	-	-	0	-	-	0	-	-	0	-	-
6:2 FTS	0	-	-	0	-	-	0	-	-	0	-	-
8:2 FTS	0	-	-	0	-	-	0	-	-	0	-	-
<i>Alternatives</i>												
ADONA	0	-	-	0	-	-	0	-	-	0	-	-
Gen-X	45	<LOQ to 0.38	0.11	52	<LOQ to 0.38	0.13	0	-	-	0	-	-
F53B	0	-	-	0	-	-	0	-	-	0	-	-
ΣPFASs		0.19 to 17	10		0.26 to 17	6.1		0.08 to 3.4	1.1		- to 1.4	0.66

^a DR: Detection rate (%).

^b <LOQ: Below limit of quantification.

^c -: <LOD (Below limit of detection).

Table S12. Global comparison of PFASs in seawater measured in this study with those reported in previous studies.

Location	Sampling Year	<i>n</i> ^a	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFBS	PFOS	Gen-X	other PFASs	∑PFASs	References
<i>Seawater (ng L⁻¹)</i>														
Geum River, Korea	2012	2	<LOD ^b	0.88–3.8 (2.3)	0.92–0.94 (0.93)	0.49–0.67 (0.58)	5.9–8.5 (7.2)	0.41–0.79 (0.6)	0.81–1.9 (1.4)	0.81–1.9 (1.4)	NA ^c	1.2	17–20 (19)	Hong et al. (2015)
Bohai Sea, China	2017	20	3.6–18 (10)	1.7–8.9 (4.7)	0.58–12 (5.3)	0.73–10 (4.0)	3.6–630 (110)	0.56–8.7 (1.9)	0.61–31 (4.8)	0.95–14 (4.1)	<LOD–4.0 (1.0)	5.9	20.5–680 (150)	Liu et al. (2019)
Bukhan river	2012	3	NA	NA	0.11–0.31 (0.18)	0.12–0.27 (0.19)	0.56–1.4 (0.94)	0.29–0.52 (0.38)	NA	0.83–1.8 (1.3)	NA	0.89	2.3–5.7 (3.9)	Lam et al. (2014)
Nakdong river	2012	3	NA	NA	0.51–7.9 (3.8)	0.71–3.4 (1.9)	3.6–8.3 (6.5)	0.83–4.5 (2.3)	NA	6.3–8.5 (7.4)	NA	4.2	15–41 (26)	Lam et al. (2014)
Yeongsan river	2012	3	NA	NA	0.93–1.3 (1.1)	0.41–0.79 (0.60)	2.4–4.7 (4.0)	0.54–1.1 (0.85)	NA	1.2–15 (11)	NA	1.1	8.5–25 (19)	Lam et al. (2014)
Yellow Sea	2017	5	NA	0.79–2.7 (1.4)	0.39–0.73 (0.57)	<LOD–0.23 (0.05)	2.1–4.5 (3.0)	NA	0.68–1.3 (0.94)	<LOD–0.12 (0.03)	NA	0.17	4.9–7.4 (6.2)	Wang et al. (2022)
East China Sea	2017	7	NA	0.97–2.8 (1.5)	<LOD–0.93 (0.46)	<LOD	<LOD–2.9 (1.2)	NA	0.42–1.6 (1.1)	<LOD–0.1 (0.03)	NA	0.29	1.5–8.6 (4.5)	Wang et al. (2022)
South China Sea	2017	6	NA	0.49–1.2 (0.73)	<LOD–0.39 (0.29)	<LOD	0.03–0.50 (0.24)	NA	0.29–1.5 (1.0)	<LOD–0.37 (0.19)	NA	0.19	1.4–3.6 (2.6)	Wang et al. (2022)
Lianyungang, South Yellow Sea, China	2018	12	NA	4.8–6.9 (5.7)	6.4–11 (8.6)	1.7–2.1 (1.8)	21–25 (23)	0.61–0.89 (0.77)	4.9–10 (6.4)	0.43–0.59 (0.52)	0.20–0.32 (0.25)	9.8	46–70 (57)	Feng et al. (2020)
Yancheng, South Yellow Sea, China	2018	11	NA	5.0–8.2 (6.6)	6.0–11 (8.5)	1.4–2.2 (1.8)	17–31 (24)	0.49–0.86 (0.70)	2.3–16 (8.4)	0.39–1.4 (0.73)	0.11–0.28 (0.18)	10	38–95 (61)	Feng et al. (2020)
Nantong, South Yellow Sea, China	2018	7	NA	1.9–8.0 (4.4)	3.4–8.2 (5.4)	1.0–1.6 (1.3)	11–25 (16)	0.47–0.94 (0.57)	1.9–11 (5.3)	0.49–1.8 (0.89)	0.06–0.11 (0.09)	7.5	24–69 (42)	Feng et al. (2020)

^a n: Sample numbers analyzed.^b <LOD: Below limit of detected.^c NA: Not available.

Table S13. Concentrations (ng L⁻¹) of individual PFASs in seawater samples.

Sites	Y1	Y1	Y1	Y1	Y1	Y1	Y2	Y2	Y2	Y2	Y2	Y2
	0m	10m	20m	30m	50m	72m	0m	10m	20m	30m	50m	74m
PFBA	0.31	0.30	0.32	0.36	0.29	0.25	<LOQ ^c	0.30	<LOQ	<LOQ	0.25	0.26
PFPeA	0.45	0.44	0.47	0.42	0.44	0.43	0.38	0.35	0.51	0.37	0.42	0.42
PFHxA	0.67	0.69	0.65	0.70	0.67	0.66	0.65	0.62	0.60	0.62	0.69	0.64
PFHpA	0.36	0.36	0.38	0.35	0.35	0.35	0.34	0.33	0.33	0.33	0.36	0.33
PFOA	6.4	6.3	6.3	6.2	6.1	6.0	5.7	5.5	5.4	5.2	5.7	5.4
PFNA	0.09	0.09	0.09	0.09	0.10	0.09	0.09	0.08	0.08	<LOQ	0.08	<LOQ
PFDA	<LOD ^b	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	0.48	0.48	0.46	0.43	0.43	0.40	0.53	0.43	0.45	0.41	0.49	0.47
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD
br-PFHxS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
L-PFOS	0.37	0.37	0.45	0.31	0.30	0.32	0.35	0.21	0.38	0.33	0.20	0.37
br-PFOS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	<LOQ	<LOQ	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOQ	0.24	<LOQ
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	9.1	9.0	9.2	8.9	8.7	8.4	8.1	7.9	7.7	7.3	8.4	7.9

Sites	Y3	Y3	Y3	Y3	Y3	Y4	Y4	Y4	Y4	Y4	Y5	Y5
	0m	10m	20m	30m	47m	0m	10m	20m	30m	60m	0m	10m
PFBA	0.32	0.37	0.50	0.49	0.38	0.34	0.42	0.44	0.44	0.38	0.31	0.30
PFPeA	0.68	0.69	0.68	0.79	0.72	0.64	0.65	0.62	0.68	0.63	0.51	0.53
PFHxA	0.98	1.0	0.94	1.0	1.0	0.98	0.98	0.95	0.95	0.86	0.83	0.86
PFHpA	0.63	0.64	0.61	0.66	0.67	0.55	0.54	0.58	0.57	0.53	0.48	0.48
PFOA	11	11	10	12	11	9.7	9.9	9.9	9.9	9.4	8.3	8.4
PFNA	0.16	0.18	0.16	0.19	0.20	0.18	0.14	0.15	0.19	0.15	0.13	0.12
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	0.56	0.60	0.61	0.69	0.62	0.57	0.61	0.59	0.56	0.54	0.53	0.52
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
br-PFHxS	<LOQ	<LOQ	<LOQ	0.12	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
L-PFOS	0.41	0.59	0.46	0.53	0.37	0.53	0.52	0.64	0.30	0.50	0.43	0.35
br-PFOS	<LOQ	<LOQ	<LOD	<LOQ	<LOD	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOD	<LOQ
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	0.37	0.29	<LOQ	0.36	0.38	0.28	0.26	0.19	<LOQ	0.22	0.23	0.24
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	15	16	14	16	16	14	14	14	14	13	12	12

Sites	Y5	Y5	Y5	Y5	Y6	Y6	Y6	Y6	Y6	Y6	Y7	Y7
	20m	30m	50m	68m	0m	10m	20m	30m	50m	72m	0m	10m
PFBA	0.28	0.28	0.28	0.28	0.26	0.32	0.30	0.39	<LOQ	0.25	0.27	0.27
PFPeA	0.56	0.51	0.47	0.49	0.44	0.43	0.44	0.43	0.47	0.38	0.48	0.44
PFHxA	0.79	0.86	0.84	0.82	0.68	0.75	0.74	0.69	0.73	0.68	0.76	0.76
PFHpA	0.46	0.44	0.44	0.43	0.41	0.38	0.35	0.38	0.38	0.33	0.39	0.39
PFOA	8.0	7.9	8.0	7.3	6.7	6.6	6.8	6.9	6.6	5.8	6.8	7.3
PFNA	0.12	0.12	0.12	0.12	0.09	0.10	0.10	0.11	0.09	0.10	0.10	0.12
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	0.50	0.59	0.46	0.50	0.50	0.46	0.45	0.50	0.45	0.46	0.47	0.52
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOD	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOQ
br-PFHxS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
L-PFOS	0.49	0.27	0.41	0.35	0.41	0.43	0.45	0.31	0.33	0.25	0.25	0.55
br-PFOS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	0.22	0.22	0.26	0.23	0.19	0.24	<LOQ	0.23	<LOQ	0.20	<LOQ	<LOQ
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	11	11	11	10	9.7	9.7	9.7	9.9	9.0	8.4	9.6	10

Sites	Y7	Y7	Y7	Y7	Y8	Y8	Y8	Y8	Y8	Y8	Y8	Y9
	20m	30m	50m	75m	0m	10m	20m	30m	50m	75m	87m	0m
PFBA	0.26	0.24	0.33	<LOQ	0.25	0.24	<LOQ	0.27	0.26	0.24	<LOQ	<LOQ
PFPeA	0.48	0.42	0.44	0.44	0.41	0.39	0.37	0.36	0.42	0.36	0.39	0.37
PFHxA	0.75	0.70	0.70	0.71	0.68	0.68	0.65	0.63	0.65	0.57	0.63	0.60
PFHpA	0.40	0.37	0.40	0.36	0.36	0.30	0.30	0.34	0.32	0.29	0.33	0.33
PFOA	6.9	6.3	6.7	6.8	5.6	5.8	5.6	5.6	5.7	5.2	5.5	6.2
PFNA	0.10	0.09	0.09	0.08	<LOQ	0.08	<LOQ	<LOQ	0.08	<LOQ	0.09	0.08
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	0.50	0.51	0.47	0.43	0.36	0.39	0.42	0.46	0.43	0.24	0.38	0.50
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOD	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOQ
br-PFHxS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
L-PFOS	0.33	0.52	0.42	0.47	0.28	0.21	0.36	0.23	0.21	0.10	0.23	0.29
br-PFOS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	0.28	<LOQ	<LOQ	<LOQ	0.23	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.22	0.22
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	10	9.2	9.5	9.2	8.2	8.0	7.7	7.9	8.0	7.0	7.8	8.6

Sites	Y9	Y9	Y9	Y9	Y9	Y10	Y10	Y10	EC1	EC1	EC1	EC1
	10m	20m	30m	50m	83m	0m	10m	27m	0m	10m	20m	30m
PFBA	<LOQ	0.32	0.27	0.32	0.27	<LOQ	<LOQ	<LOD	0.47	0.40	0.41	0.39
PFPeA	0.42	0.41	0.41	0.39	0.42	0.30	0.13	<LOD	0.79	0.48	0.70	0.32
PFHxA	0.70	0.70	0.64	0.66	0.71	0.51	0.20	<LOD	1.1	0.84	1.1	0.52
PFHpA	0.37	0.37	0.35	0.32	0.34	0.24	0.10	<LOD	0.54	0.42	0.60	0.29
PFOA	5.9	5.9	6.1	5.7	6.0	3.8	1.8	0.19	11	7.6	10	5.3
PFNA	0.08	0.09	0.09	0.08	0.10	<LOQ	<LOQ	<LOD	0.16	0.12	0.15	0.08
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	0.51	0.56	0.49	0.49	0.47	0.37	0.16	<LOD	0.52	0.43	0.52	0.20
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOQ	<LOD	<LOQ	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD
br-PFHxS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOD
L-PFOS	0.23	0.38	0.44	0.26	0.24	0.28	0.12	<LOD	0.33	0.36	0.52	0.09
br-PFOS	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	0.19	0.21	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOD	0.29	0.38	0.36	0.20
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	8.4	8.9	8.8	8.3	8.6	5.5	2.5	0.19	15	11	15	7.4

Sites	EC1	EC2	EC2	EC2	EC2	EC3	EC3	EC3	EC3	EC3	EC4	EC4
	37m	0m	10m	20m	38m	0m	10m	20m	30m	47m	0m	10m
PFBA	0.40	0.38	0.51	0.42	0.46	0.28	0.29	0.30	0.31	<LOQ	<LOD	<LOD
PFPeA	0.50	0.73	0.62	0.62	0.61	0.54	0.44	0.53	0.48	0.49	0.19	0.19
PFHxA	0.87	1.3	1.2	1.0	1.1	0.92	0.75	0.94	0.75	0.94	0.45	0.45
PFHpA	0.45	0.65	0.54	0.54	0.59	0.45	0.38	0.47	0.39	0.38	0.13	0.13
PFOA	7.8	12	10	10	10	8.1	6.8	8.3	7.1	6.9	2.3	2.5
PFNA	0.12	0.17	0.14	0.15	0.16	0.12	0.08	0.10	0.12	0.12	<LOQ	<LOQ
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	0.32	0.65	0.47	0.45	0.51	0.57	0.47	0.62	0.33	0.46	0.43	0.45
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOQ	<LOQ	<LOQ	<LOD	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
br-PFHxS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOD
L-PFOS	0.27	0.44	0.20	0.37	0.63	0.47	0.30	0.42	0.27	0.39	0.23	0.16
br-PFOS	<LOD	<LOD	<LOQ	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	0.21	0.35	0.28	0.24	0.27	<LOQ	0.24	0.28	0.22	0.26	<LOQ	<LOQ
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	11	16	14	14	15	11	9.8	12	10	9.9	3.8	3.9

Sites	EC4 20m	EC4 30m	EC4 50m	EC4 66m	EC5 0m	EC5 10m	EC5 20m	EC5 30m	EC5 50m	EC5 87m	EC6 0m	EC6 10m
PFBA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFPeA	0.15	0.14	0.14	0.12	<LOQ	<LOQ	0.10	<LOQ	<LOQ	<LOQ	<LOD	<LOQ
PFHxA	0.43	0.40	0.39	0.35	0.30	0.24	0.22	0.25	0.24	0.13	0.12	0.10
PFHpA	0.12	0.11	0.11	0.09	0.05	0.06	0.04	0.05	0.05	<LOQ	<LOQ	<LOQ
PFOA	2.3	2.2	2.0	2.0	0.72	0.74	0.68	0.73	0.69	0.36	0.30	0.29
PFNA	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	0.37	0.32	0.26	0.27	0.28	0.24	0.25	0.24	0.22	<LOQ	0.12	<LOQ
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
br-PFHxS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFOS	0.18	<LOD	0.18	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.09	<LOQ
br-PFOS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	0.22	0.21	<LOQ	0.19	0.19	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOQ
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	3.8	3.4	3.0	3.0	1.5	1.3	1.3	1.3	1.2	0.5	0.63	0.39

Sites	EC6 20m	EC6 30m	EC6 50m	EC6 75m	EC6 103m	S1 5m	S1 10m	S1 20m	S1 30m	S1 50m	S1 75m	S2 5m
PFBA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFPeA	<LOD	<LOQ	<LOQ	<LOQ	<LOD	<LOQ	0.12	0.13	0.11	<LOQ	<LOD	0.12
PFHxA	0.10	0.10	0.10	0.14	0.11	0.11	0.20	0.26	0.21	0.08	<LOQ	0.21
PFHpA	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOQ	0.08	0.12	0.08	<LOQ	<LOD	0.09
PFOA	0.31	0.30	0.33	0.31	0.16	0.40	1.1	1.5	1.1	0.39	0.08	1.0
PFNA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	<LOQ	0.15	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.16	<LOQ	<LOQ	<LOD	0.13
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ
br-PFHxS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOQ
L-PFOS	<LOD	<LOQ	<LOQ	<LOD	<LOD	<LOQ	<LOQ	<LOQ	0.12	0.09	<LOD	<LOQ
br-PFOS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	<LOD	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	0.41	0.56	0.43	0.45	0.26	0.51	1.5	2.2	1.7	0.56	0.08	1.6

Sites	S2	S2	S2	S2	S2	S2	S2	S3	S3	S3	S3	S3
	10m	20m	30m	50m	75m	100m	110m	5m	10m	20m	30m	50m
PFBA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFPeA	<LOQ	<LOQ	<LOQ	<LOD	<LOQ	<LOD	<LOQ	0.18	0.13	0.09	0.09	<LOQ
PFHxA	0.16	0.10	0.10	0.07	0.08	<LOQ	<LOQ	0.19	0.23	0.19	0.17	<LOQ
PFHpA	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOD	0.08	0.12	0.10	0.07	<LOQ
PFOA	0.76	0.51	0.28	0.17	0.17	0.11	0.08	1.3	1.5	1.2	1.0	0.29
PFNA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOQ	0.15	0.15	<LOQ	<LOD
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD
br-PFHxS	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOQ	<LOD	<LOQ	<LOD
L-PFOS	<LOQ	<LOQ	<LOD	<LOQ	<LOD	<LOD	<LOQ	0.19	<LOQ	<LOQ	<LOQ	<LOQ
br-PFOS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOQ	<LOD
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	0.93	0.61	0.38	0.24	0.25	0.11	0.08	1.9	2.2	1.8	1.4	0.29

Sites	S3	S4	S4	S4	S4	S4	S4	S4	S5	S5	S5	S5
	70m	5m	10m	20m	30m	50m	75m	85m	5m	10m	20m	30m
PFBA	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFPeA	<LOQ	0.10	0.10	0.10	0.11	<LOQ	0.09	0.10	0.16	0.11	0.09	0.09
PFHxA	<LOQ	0.18	0.17	0.16	0.19	0.15	0.17	0.14	0.19	0.18	0.21	0.19
PFHpA	<LOD	0.07	0.10	0.09	0.07	<LOQ	<LOQ	0.07	0.08	0.08	0.09	<LOQ
PFOA	0.13	0.98	0.94	0.92	1.1	0.80	0.84	0.80	0.95	0.98	1.0	0.93
PFNA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDoDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTriDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	<LOD	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOQ	<LOD
br-PFHxS	<LOD	<LOQ	<LOD	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOQ	<LOQ	<LOQ	<LOD
L-PFOS	<LOD	0.17	0.22	<LOQ	0.18	<LOQ	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	0.09
br-PFOS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	0.86	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.96
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	<LOQ	<LOD	<LOD	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOQ	<LOD
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	0.13	1.5	1.5	1.3	2.6	0.95	1.1	1.1	1.4	1.3	1.4	2.3

Sites	E7 0m	E7 10m	E7 20m	E7 30m	E7 50m	E7 75m	E7 100m	E7 200m	E7 500m	E7 1000m	E7 1500m	E7 2142m
PFBA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFPeA	<LOD	<LOQ	<LOQ	<LOD	<LOQ	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD
PFHxA	0.11	0.10	0.13	0.13	0.11	0.12	0.11	<LOQ	<LOD	<LOD	<LOD	<LOD
PFHpA	0.09	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOD
PFOA	0.71	0.65	0.70	0.71	0.63	0.68	0.65	0.33	0.07	<LOD	<LOD	<LOD
PFNA	<LOQ	<LOQ	<LOD	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD
PFDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFUnDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFDODA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTrDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PFTeDA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFBS	0.17	0.13	0.12	0.16	0.13	0.14	0.14	0.11	<LOD	<LOD	<LOD	<LOD
L-PFPeS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHxS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
br-PFHxS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFOS	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
br-PFOS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFHpS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFNS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
L-PFDS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N- MeFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
N-EtFOSAA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
4:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
6:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
8:2FTS	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ADONA	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Gen-X	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
F53B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ΣPFASs	1.1	0.88	0.95	1.0	0.87	0.94	0.89	0.44	0.07	N.D	N.D	N.D

^a N.D: Not detected.

^b LOD: Limit of detection.

^c LOQ: Limit of quantification.

Supplementary Figures

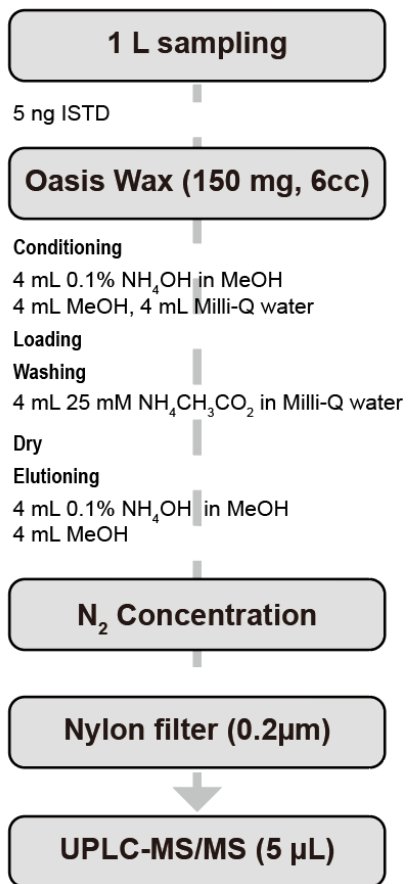


Fig. S1. Analytical procedures for per- and polyfluoroalkyl substances (PFASs) in seawater samples.

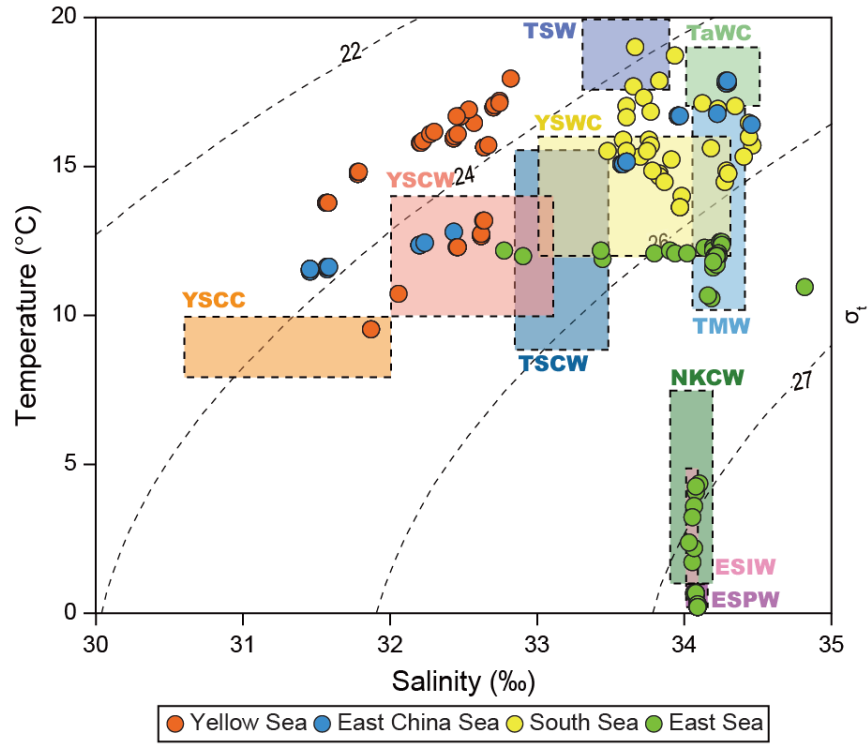


Fig. S2. Water masses of the Yellow Sea, East China Sea, South Sea, and East Sea based on the temperature-salinity (T-S) diagram obtained in this study.

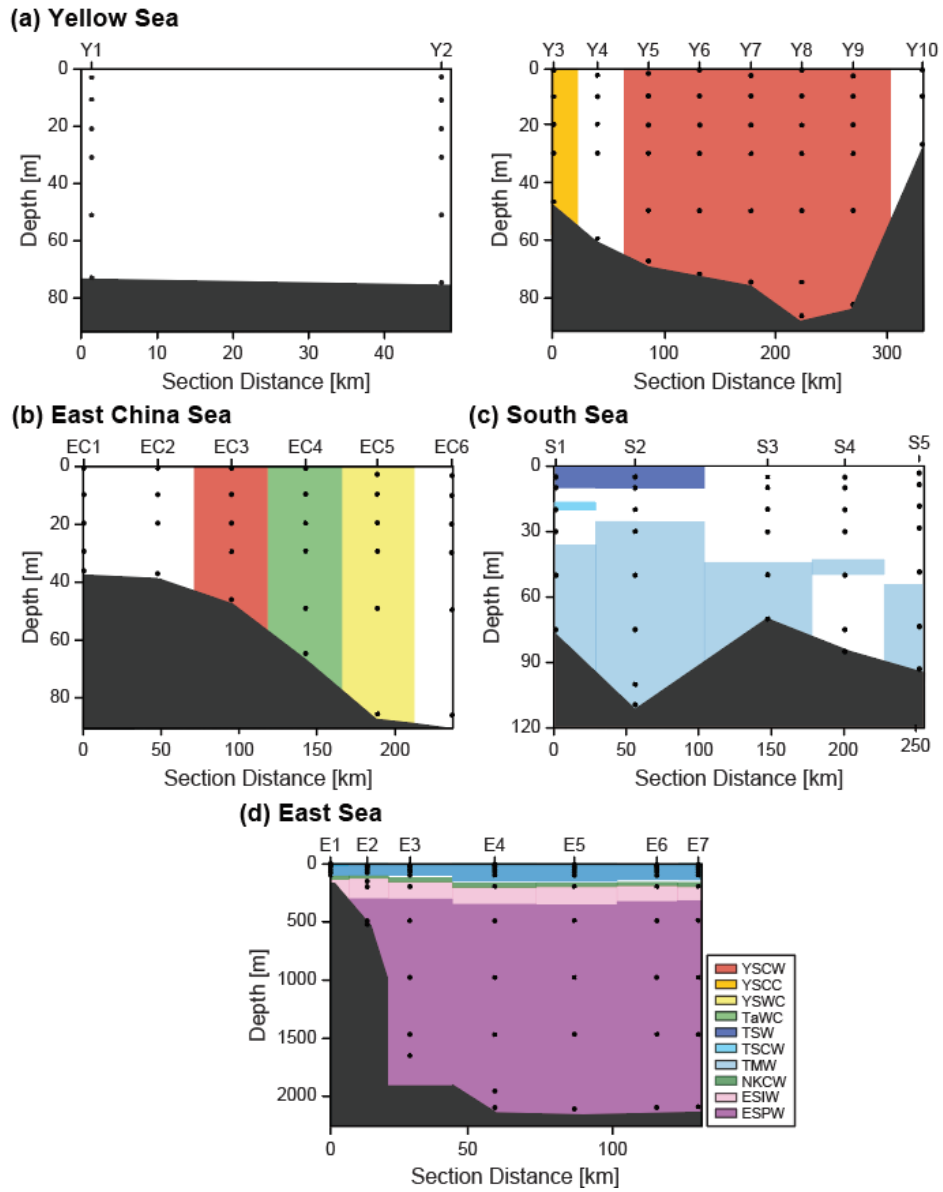


Fig. S3. Distribution of water masses in the (a) Yellow Sea, (b) East China Sea, (c) South Sea, and (d) East Sea. Black circles in the figure represent the depths at which sampling was conducted.

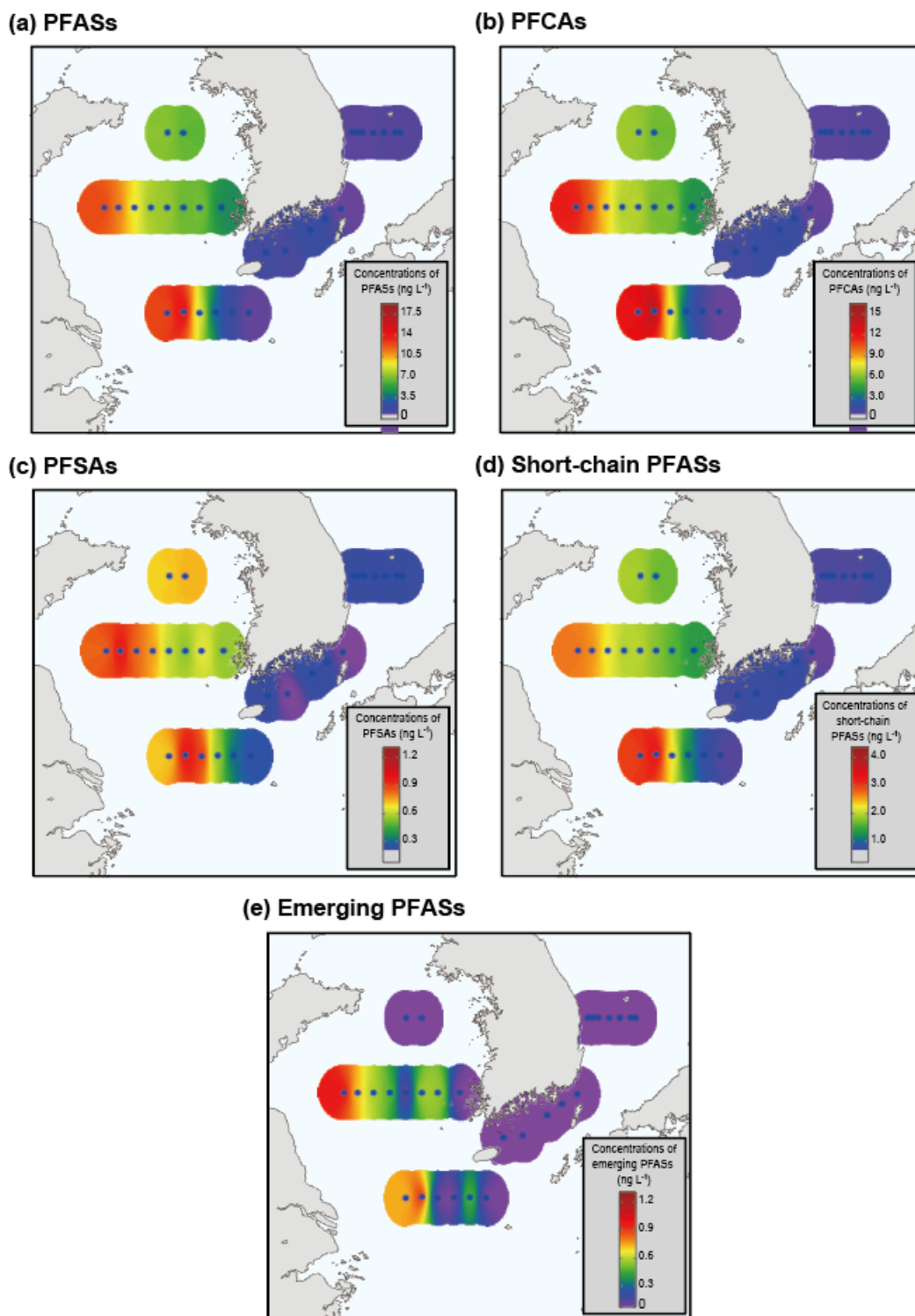


Fig. S4. Spatial distributions of (a) PFASs, (b) PFCAs, (c) PFSAs, (d) short-chain PFASs, and (e) emerging PFASs in surface waters of regional seas of South Korea.

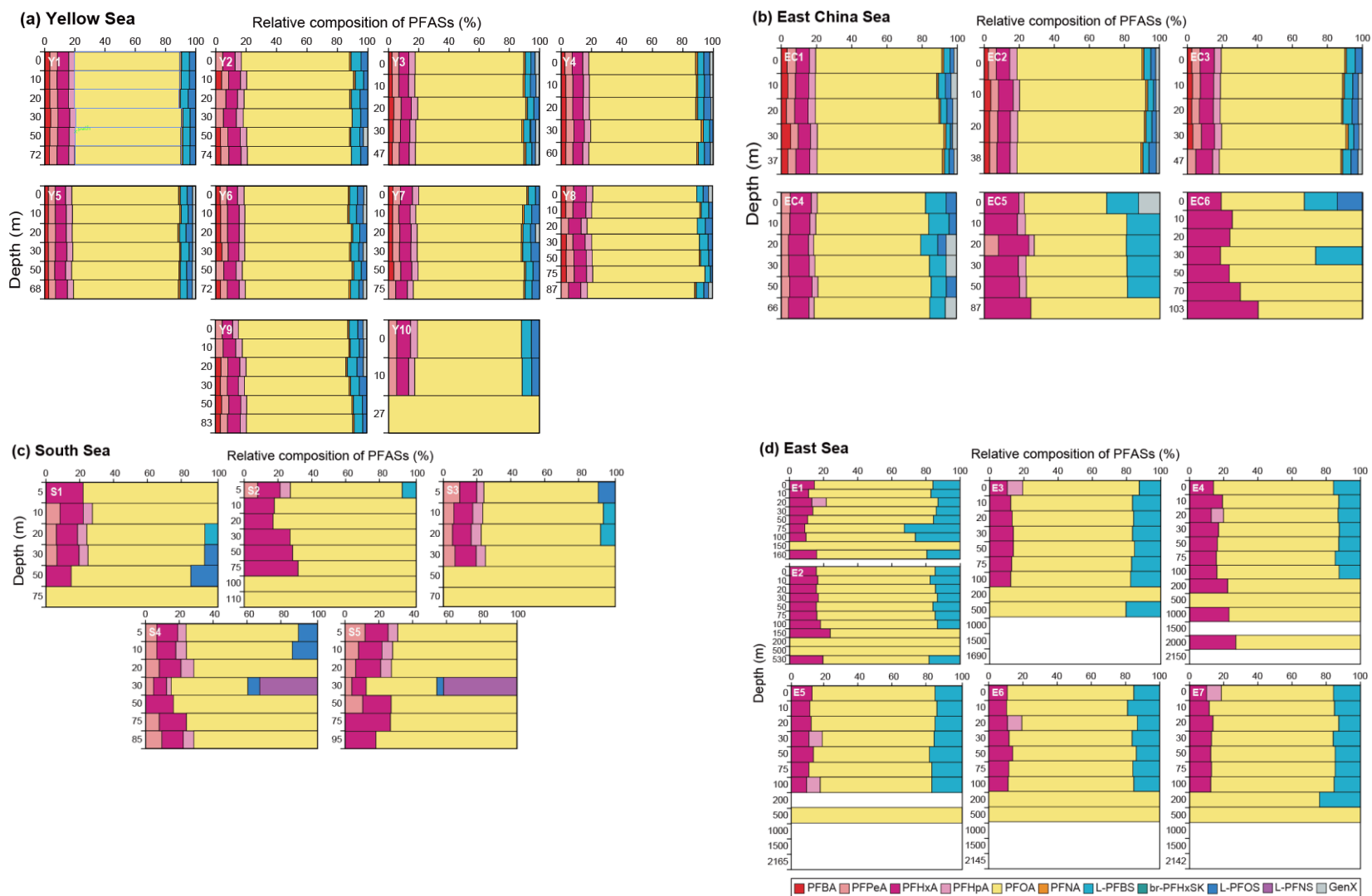


Fig. S6. Vertical profiles of PFASs compositions in the water columns of the (a) Yellow Sea, (b) East China Sea, (c) South Sea, and (d) East Sea.

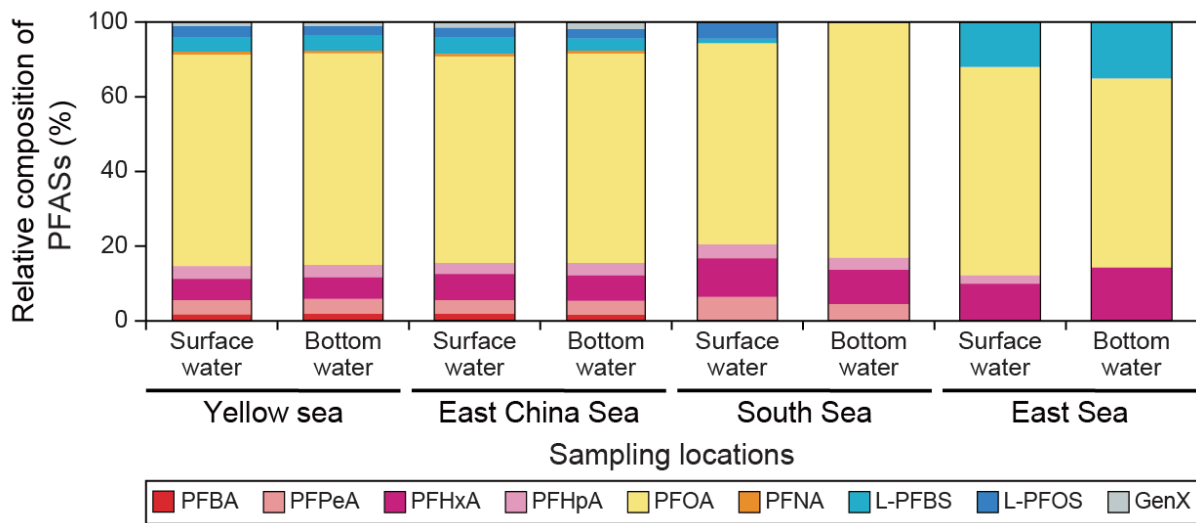


Fig. S7. Concentration profiles of legacy and emerging PFASs in surface water and bottom water.

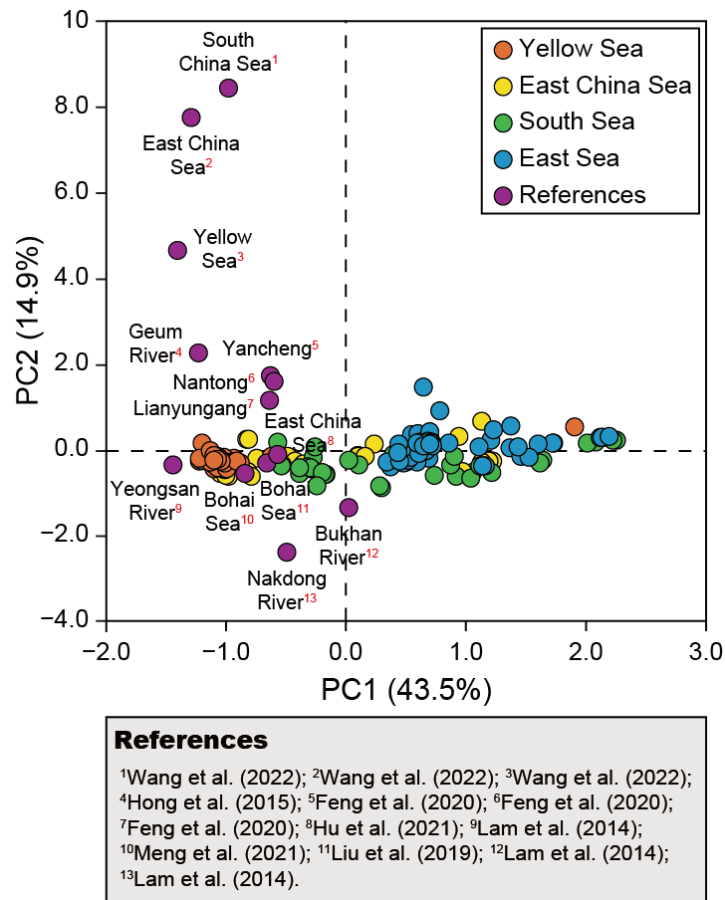


Fig. S8. Principal component analysis of PFASs compositions in water samples collected from freshwater, coastal areas, and regional seas of China and South Korea.

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