



PFAS contamination in the Western Seto Inland Sea of Japan: Substitution lag and TFA risk alert

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ABSTRACT

The western Seto Inland Sea (WSIS) is surrounded by major industrial zones in Japan and receives substantial inputs of pollutants. However, per- and polyfluoroalkyl substances (PFAS) contamination in the WSIS remains poorly characterized. In this study, we investigated the occurrence and potential sources of PFAS in seawater samples from the WSIS, and conducted a preliminary ecological risk assessment. A total of 24 PFAS were identified and quantified, with total concentrations ($\Sigma_{24}\text{PFAS}$) ranging from 519 to 916 ng/L. Trifluoroacetic acid (TFA) was the predominant compound (mean: 680 ± 156 ng/L), accounting for 96.9% of $\Sigma_{24}\text{PFAS}$. After excluding TFA, perfluoroalkyl sulfonic acids (PFASs) accounted for more than half of the remaining PFAS. Except for TFA, the remaining PFAS generally exhibited a spatial pattern suggestive of localized point-source inputs, with higher concentrations nearshore waters than in offshore waters. Principal component analysis-multiple linear regression (PCA-MLR) suggested that non-TFA PFAS were mainly associated with electroplating-related and surfactant-related sources, whereas correlation patterns and spatial distributions suggested that TFA was primarily associated with the atmospheric degradation of hydrofluorocarbons (HFCs) and hydrofluoroolefins (HFOs). The results further suggest that the transition from legacy PFAS to emerging alternatives remains incomplete. Ecological risk assessment based on risk quotients (RQs) indicated that the estimated risk posed by TFA varied substantially depending on the toxicity data used. Under the worst-case scenario, TFA posed a high risk ($\text{RQ} > 1$), whereas perfluorooctane sulfonate (PFOS) exhibited a low risk ($\text{RQ} < 0.01$). These findings underscore the need for multi-compartment and seasonal monitoring in the WSIS, inclusion of TFA monitoring programs across other marine regions, and expanded toxicity testing in marine organisms.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a class of synthetic chemicals characterized by unique physicochemical properties, including high thermal stability, chemical inertness, and surface activity (Lindstrom et al., 2011). Consequently, PFAS have been extensively used

for nearly seven decades as surfactants, lubricants, and processing aids in a wide range of industrial and consumer applications, including food packaging, cookware, textiles, electroplating processes, and other applications (Buck et al., 2011; Glüge et al., 2020). However, the exceptionally strong carbon–fluorine (C–F) bonds in PFAS make many of these compounds highly resistant to degradation under natural

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environmental conditions, contributing to their common designation as “forever chemicals” (Cousins et al., 2020). Perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) are among the most well-known PFAS. Both compounds have high bioaccumulation potential and can cause toxic effects in organisms, including humans. They were subsequently listed under the Stockholm Convention for global regulatory control (Stockholm Convention, 2019). This global regulatory framework has driven the fluorochemical industry toward two main substitution strategies. One involves shortening the carbon chain length, exemplified by short-chain homologues such as perfluorobutanoic acid (PFBA) and perfluorobutane sulfonate (PFBS). The other is the insertion of ether oxygen linkages into the carbon backbone, giving rise to novel perfluoroalkyl ether carboxylic acids (PFECAs) and perfluoroalkyl ether sulfonic acids (PFESAs) (Hamid et al., 2024; Wang et al., 2013). Both strategies aim to shorten the effective carbon chain length, thereby reducing the anticipated environmental persistence, bioaccumulation, and toxicity.

However, the environmental safety of these alternatives has increasingly been called into question. Although short-chain PFAS are considered to have lower bioaccumulation potential, their high water solubility and long-range transport capacity render them highly persistent in the marine environment. Perfluorohexane sulfonate (PFHxS), as a short-chain alternative, has also attracted increasing concern and is now subject to progressive restrictions (Bao et al., 2023). Consequently, novel ether-based alternatives, such as hexafluoropropylene oxide trimer acid (HFPO-TA) and 6:2 chlorinated polyfluoroether sulfonic acid (6:2 Cl-PFESA), have gained increasing use as replacements for legacy PFAS. Notably, PFECAs and PFESAs have been detected in various environmental matrices and biota in recent years, and have been demonstrated to exhibit bioaccumulation and hepatotoxicity comparable to those of legacy PFAS (Pan et al., 2017; Sheng et al., 2018; Yi et al., 2019). This underscores the urgent need for careful monitoring of the occurrence of these alternatives in the marine environment. More critically, trifluoroacetic acid (TFA), an ultrashort-chain PFAS (USC-PFAS) and a terminal atmospheric oxidation product of various hydrofluorocarbons (HFCs) and hydrofluoroolefins (HFOs), is continuously accumulating in the global oceans via dry and wet deposition, posing a potential threat to marine ecosystems (Frank et al., 2002). Although TFA is generally considered less toxic than many long-chain PFAS, its increasing environmental concentrations and persistence have raised concerns about potential ecological and human health effects (Arp et al., 2024; Maerten et al., 2026). Zhai et al. (2015) revealed that over 99% of TFA occurred in the water phase, with concentrations continuing to increase. A recent study of surface waters in the Yangtze River Delta reported an average TFA concentration of 8.7×10^4 ng/L, accounting for 90.1–99.6% of total PFAS and making it the most abundant compound in the aqueous environment (Miao et al., 2025). Moreover, Ma et al. (2025) investigated estuaries and adjacent coastal waters near a major fluorochemical industrial park, and similarly found that TFA dominated the PFAS profile (87–99%) and posed the greatest risk in those areas. Therefore, as regulatory-driven substitution continues, the co-occurrence patterns, extent of substitution, and ecological risks of legacy and emerging PFAS in the marine environment warrant comprehensive characterization.

The Seto Inland Sea (SIS) is the largest semi-enclosed sea in Japan and serves as a major marine corridor between western Japan and the Pacific Ocean. It is also an important mariculture region in Japan, with major industrial zones concentrated along its western coast (Gotoh et al., 1998). The SIS is characterized by relatively weak hydrodynamic exchange and long water residence times, which can facilitate pollutant retention in coastal waters and the persistence of chemical signatures associated with industrial discharges (Nishijima et al., 2016). PFAS released from these industrial activities may enter coastal waters through direct discharge and other transport pathways and subsequently be transported offshore by ocean currents (Li et al., 2026b; Miller et al., 2026). Although several studies have reported PFAS in

Japanese coastal waters such as Tokyo Bay (Ahrens et al., 2010) and Osaka Bay (Beškoski et al., 2017), most of these analyses have been limited to conventional PFAS with carbon chain lengths of C4 and above, with little attention paid to ultrashort-chain and emerging PFAS. In view of this, this study focused on the western Seto Inland Sea (WSIS) and systematically determined both legacy and emerging PFAS, including TFA. The investigation aimed to (1) characterize the concentration levels, spatial distribution, and compositional profiles of PFAS; (2) quantitatively apportion potential pollution sources and assess the extent of substitution from legacy to emerging PFAS; and (3) conduct a preliminary ecological risk assessment and identify substances and regions with elevated estimated risks. This study provides a scientific basis for PFAS contamination management in highly industrialized semi-enclosed marine systems and serves as a regional case study for assessing PFAS risks in global marine environments.

2. Materials and methods

2.1. Sample collection

The sampling stations in the WSIS are shown in Fig. 1. A research cruise was conducted aboard the R/V NAGASAKI MARU in November 2025. Seawater samples (500 mL) were collected from depths of 0.5–1 m below the surface using a stainless-steel sampler and stored in polypropylene bottles that had been pre-cleaned with methanol and ultrapure water. To minimize potential PFAS loss due to adsorption, all samples were ultrasonicated to desorb any PFAS that may have adhered to the bottle walls and then filtered through glass fiber filters (0.7 μ m, 47 mm, Whatman, UK). The processed samples were stored at -20 °C until analysis. Detailed information on the sampling stations is provided in Table S1 in the Supporting Information. It should be noted that this study is based on a single research cruise conducted in November 2025 and a limited number of sampling stations. The dataset therefore represents a temporal snapshot of PFAS contamination in the WSIS and does not capture seasonal variations in precipitation, river discharge, water-mass mixing, or industrial and atmospheric emissions. Consequently, the spatial distribution, source apportionment, and ecological risk assessment presented below should be regarded as preliminary and interpreted with caution. Future multi-season and multi-compartment monitoring is needed to assess the representativeness of these findings.

2.2. Standards and reagents

A total of 55 PFAS were included in the study. Based on chain structure and functional groups, the target compounds can be further categorized into perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkyl sulfonic acids (PFSA), *Per*- and polyfluoroether carboxylic acids (PFECAs), *per*- and polyfluoroalkyl ether sulfonic acids (PFESAs), fluorotelomer sulfonic acids (FTSAs), *per*- and polyfluoroalkyl sulfonamides (PFASAs), *per*- and polyfluoroalkyl sulfonamidoacetic acids (PFASAAAs). According to carbon chain length, analytes were further classified as ultrashort-chain (with <4 carbons in their molecular structures, i.e., TFA, pentafluoropropionic acid (PFPrA), perfluoromethanesulfonic acid (PFMeS), perfluoroethanesulfonic acid (PFEtS), perfluoropropanesulfonic acid (PFPrS), bis(trifluoromethane)sulfonimide (Ntf2) and perfluoro-2-methoxyacetic acid (PFMOAA)), short-chain (C4–C7 for carboxylates and C4–C6 for sulfonates), or long-chain ($\geq$ C8 for carboxylates and $\geq$ C7 for sulfonates). Details of analyte standards, mass-labeled internal standards, and reagents are provided in Text S1.

2.3. Sample preparation and analysis

Sample preparation prior to analysis was minimal. Samples were thawed at room temperature and ultrasonicated to homogenize the seawater and facilitate the desorption of any PFAS that may have

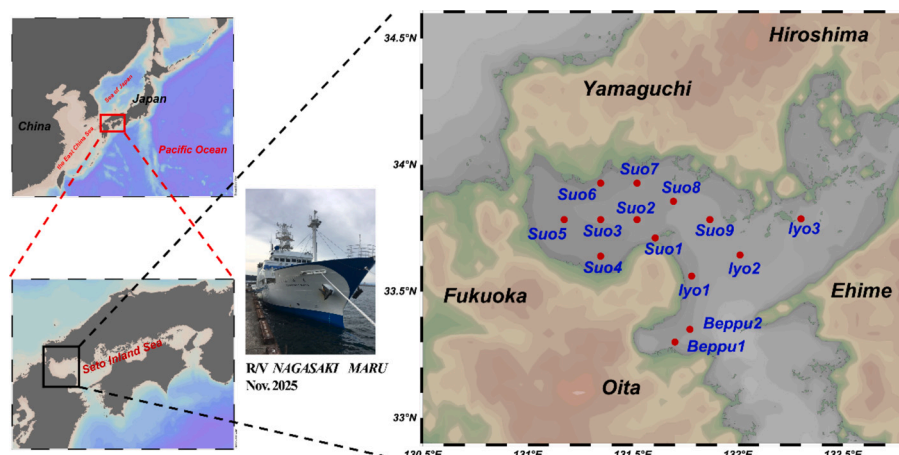


Fig. 1. Map of the sampling stations.

adsorbed to the bottle walls. Three 5 mL subsamples were then taken from the upper, middle, and lower portions of the bottle and combined to form a 15 mL composite aliquot. This composite aliquot was transferred to a new tube and amended with ammonium acetate to a final concentration of 5 mM. After spiking with isotope-labeled internal standards, a 500 μ L aliquot was injected by direct injection. Compounds were quantified using a multidimensional LC platform (1290 Infinity II $\times$ 1290 Infinity II) coupled with a triple-quadrupole MS (6495D, Agilent Technologies, USA). Detailed instrumental parameters are provided in Tables S2–3. A detailed description of the methodology can be found in our previous publication (Ma et al., 2025).

2.4. Quality assurance and quality control

Any fluorine-containing materials were rigorously avoided during sample collection, transportation, pretreatment, and instrumental analysis. All laboratory equipment and solvents were ensured to be free of contamination prior to use. Quality control samples, procedural blanks, and matrix recovery tests were prepared during the experiment to guarantee the accuracy of the data. Three procedural blanks were analyzed to assess any potential laboratory background contamination. Only TFA showed a relatively high procedural blank concentration (mean: 103 ± 17.5 ng/L); therefore, the reported TFA concentrations were blank-corrected by subtracting the corresponding blank concentrations. Detailed information regarding the limits of quantification (LOQs) and spiked recoveries is provided in Tables S4–5.

2.5. Principal component analysis-multiple linear regression

Principal component analysis-multiple linear regression (PCA-MLR) is an effective tool for apportioning pollutant sources and quantifying their contributions in a study area (Salim et al., 2019; Xu et al., 2013). In this study, PCA-MLR was applied to analyze the potential sources of PFAS with a detection frequency exceeding 40%. Concentrations below the LOQ were treated as half the LOQ. PCA was used to qualitatively identify the pollution sources of PFAS in the water column. MLR was then conducted using the principal component scores as independent variables and the Z-score normalized pollutant concentrations as the dependent variable, to estimate the contributions from multiple sources. The regression coefficients (β) obtained from MLR were used to estimate the contribution of each source, according to Eq. (1).

$$P_i(\%) = \left(\beta_i / \sum \beta_i \right) \times 100 \quad (1)$$

where P_i represents the percentage contribution of each source, and β_i denotes the regression coefficient for the i -th source. Further details on

the PCA-MLR methodology can be found in Salim et al. (2019) and Xu et al. (2013).

2.6. Ecological risk assessment

The ecological risk quotient (RQ) was calculated using Eq. (2):

$$RQ = MEC/PNEC \quad (2)$$

In this assessment, MEC (ng/L) represents the measured environmental concentration, and PNEC (ng/L) represents the predicted no-effect concentration. The risk levels were defined according to the calculated RQ values: $RQ > 1.0$ indicates high ecological risk; $0.1 < RQ \leq 1.0$ indicates medium risk; $0.01 < RQ \leq 0.1$ indicates low risk; and $RQ \leq 0.01$ indicates negligible risk (Tsui et al., 2014). The PNEC values of PFAS in seawater are provided in Table S6.

2.7. Statistical analysis

Statistical analyses were performed using IBM SPSS Statistics 27, Origin 2024, and Ocean Data View (version 5.8.6). The relationships between PFAS concentrations and carbon chain length were analyzed using linear regression for PFAS and PFASs, and quadratic polynomial regression for PFCAs and PFECAs. PFAS concentrations were $\log_{10}$ -transformed prior to regression analysis. Regression analyses were performed using Origin. The coefficient of determination (R^2) and p -values were used to evaluate model goodness of fit and statistical significance. Redundancy analysis (RDA) was performed using Origin to explore the potential relationships between PFAS concentrations and environmental factors. PFAS concentrations were $\log_{10}$ -transformed prior to analysis. Environmental factors, including nutrients and total organic carbon (TOC), were used as explanatory variables. Spearman's rank correlation analysis was employed to examine the potential correlations among different PFAS in the samples. Visualization was conducted using SPSS, Origin, and Ocean Data View.

3. Results and discussion

3.1. Occurrence of PFAS in the WSIS

Of the 55 target PFAS analyzed, 24 were reliably quantified in seawater from the WSIS. The concentration ranges and detection frequencies of the target compounds are presented in Fig. 2 (a and b) and Tables S7–8. TFA, PFOS and its alternative, 6:2 Cl-PFESA, were detected at all sampling stations. Meanwhile, the legacy C3–C14 PFCAs, including perfluoroundecanoic acid (PFUnDA), PFBA, and PFOA, were also frequently detected, with detection frequencies ranging from 85.7%

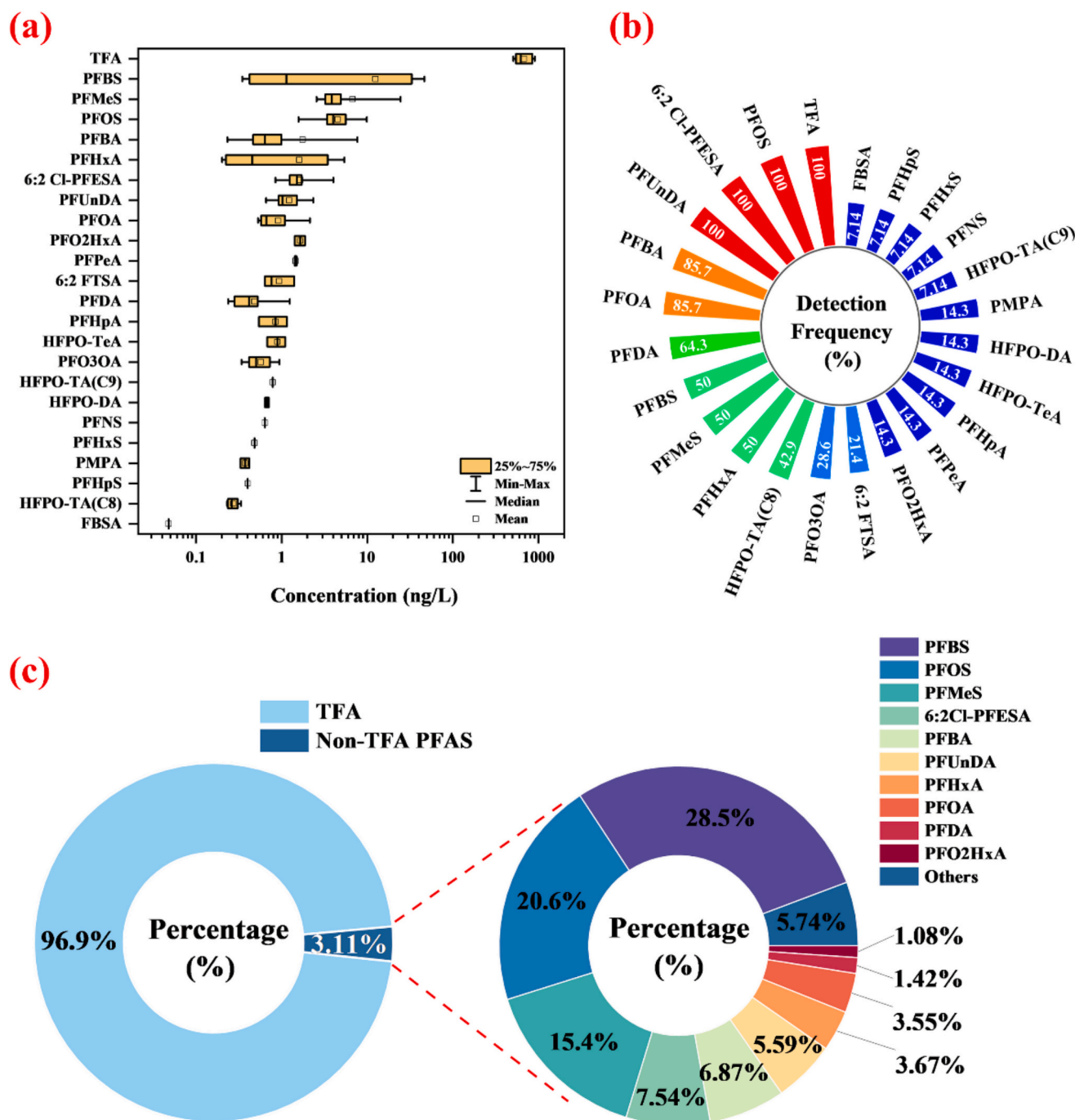


Fig. 2. Pollution characteristics of individual PFAS. (a) Concentration ranges, (b) detection frequencies, and (c) compositional profiles.

to 100%. These results indicate widespread PFAS contamination in the study area, including the continued occurrence of legacy PFAS. The total concentrations of the 24 quantified PFAS (Σ_{24} PFAS) ranged from 519 to 916 ng/L. Among them, TFA exhibited the highest concentrations, ranging from 509 to 908 ng/L with a mean of 680 ± 156 ng/L. The mean TFA concentration was more than 50-fold higher than that of PFBS (range: 0.35–46.6 ng/L; mean: 12.5 ± 19.2 ng/L), which had the second-highest concentration. In contrast, C3–C14 PFCAs and PFSAs occurred at higher concentrations than the emerging alternatives, such as HFPO-TA and 6:2 Cl-PFESA (Fig. S1).

As shown in Fig. 2c, TFA dominated the PFAS compositional profile in the WSIS, accounting for 96.9% of Σ_{24} PFAS, while the other 23 PFAS collectively accounted for 3.11%. This is consistent with findings from

other study areas where TFA was included, regardless of whether the water bodies were fresh or marine (Ma et al., 2025; Miao et al., 2025). Our results suggest that TFA warrants particular attention in the WSIS because of its exceptionally high concentration and dominant contribution to total PFAS concentrations. To better characterize the regional PFAS fingerprint, the composition of PFAS excluding TFA was further analyzed. Among the remaining PFAS, PFSAs, including PFBS (28.5%), PFOS (20.6%), and PFMeS (15.4%), together with the alternative 6:2 Cl-PFESA (7.54%), accounted for relatively large proportions, followed by PFCAs, emerging PFECAs, and other PFAS, each contributing less than 7%. This compositional pattern differs from that reported by Li et al. (2024) in the Yangtze River Estuary and by Tian et al. (2026) in Qinzhou Bay, China, where PFCAs were the dominant compounds. Previous

studies have indicated that PFASs, especially PFOS, are important components in most rivers and coastal areas of Japan (Muir and Miaz, 2021; Murakami et al., 2008; Murakami et al., 2009; Yamazaki et al., 2019). Interestingly, further analysis of the composition of different PFAS categories (Fig. S2) revealed that C4 short-chain PFASs, such as PFBS (43.9%) and PFBA (30.4%), were predominant. This is consistent with findings from some Japanese rivers (Kosaka et al., 2025). Therefore, the prevalence of short-chain PFASs in the non-TFA profile may reflect the combined influence of legacy industrial residues and the subsequent use of short-chain alternatives.

Compared with other regions, the Σ_{24} PFAS concentrations in the WSIS (range: 519–916 ng/L; mean: 702 ± 147 ng/L; median: 644 ng/L) were higher than those reported in Tokyo Bay (range: 16.7–42.3 ng/L) (Ahrens et al., 2010), Osaka Bay (maximum: 53.4 ng/L) (Beškoski et al., 2017), Qinzhou Bay (mean: 7.96 ng/L) (Tian et al., 2026), Beibu Gulf (mean: 2.04 ng/L) (Xiao et al., 2021) and Laizhou Bay (44.2–543.98 ng/L) in China (Shi et al., 2025), as well as the North and Baltic Seas in Germany (4.7–7.4 ng/L) (Joeris et al., 2019). Among the compared regions, the Σ_{24} PFAS concentration in the WSIS was relatively high. However, as noted above, the exceptionally high contribution of TFA may substantially influence this comparison. After excluding TFA, the mean Σ_{23} PFAS concentration in the WSIS was 21.9 ± 25.2 ng/L, which was comparable to the concentrations reported for Jiaozhou Bay, China (mean: 16.1 ng/L) (Han et al., 2020), and the Bay of Bengal coast, Bangladesh (10.6–46.8 ng/L) (Habibullah-Al-Mamun et al., 2016), suggesting that overall PFAS contamination in the WSIS was not exceptionally high when TFA was excluded.

For TFA alone, the concentrations in the WSIS were lower than those at the Xiaoqing River estuary and its adjacent waters in China

(2023–2024 data, > 1000 ng/L) (Ma et al., 2025), but higher than those reported in Chinese coastal bays such as the Bohai Sea and Hangzhou Bay (1999–2001 data, 4.2–190 ng/L) (Zhang et al., 2005), as well as in the Arctic, Pacific, and Atlantic Oceans (2005 data, up to 150 ng/L) (Scott et al., 2005). These comparisons highlight how the dominant contribution of TFA can substantially cross-region comparisons of total PFAS concentrations. Despite growing concern over the environmental concentrations and potential toxicity of USC-PFASs, data on their occurrence in seawater remain scarce, particularly in recent years. Cahill (2022) reported that TFA concentrations in surface waters increased approximately sixfold over a 23-year period and continue to rise, indicating that elevated TFA concentrations may warrant continued attention because of their potential ecological implications. If routine monitoring continues to exclude TFA from target analyte lists, the risks associated with PFAS contamination in bays may be substantially underestimated. Therefore, we recommend the inclusion of representative USC-PFASs in future PFAS monitoring and research efforts to comprehensively characterize PFAS contamination profiles in the environment.

3.2. Spatial distribution characteristics of PFAS

The spatial distribution and compositional profiles of Σ_{24} PFASs are presented in Fig. 3a and b. PFAS concentrations in the study area were higher near the coast than offshore, a pattern similar to that observed in many other coastal and semi-enclosed marine regions (Ma et al., 2025; Shi et al., 2025). Specifically, relatively high Σ_{24} PFAS concentrations were observed along both the northern and southern coasts of the study area. TFA dominated the PFAS profile at all sampling stations,

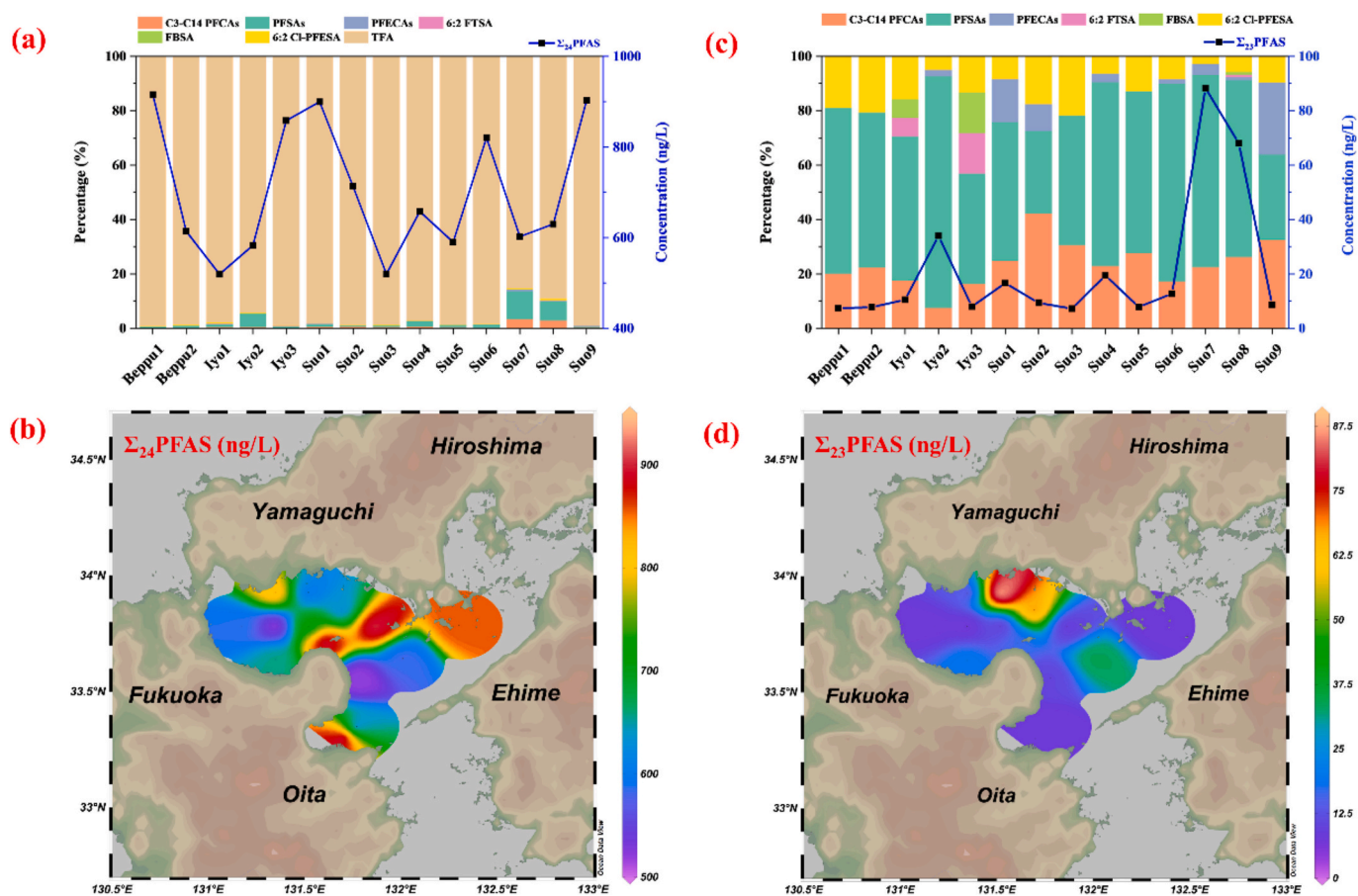


Fig. 3. Spatial distribution and compositional characteristics of PFAS. (a) and (c) show the percentage contributions of different PFAS classes to Σ_{24} PFAS and Σ_{23} PFAS (excluding TFA) at each station, respectively; the corresponding spatial distributions of Σ_{24} PFAS and Σ_{23} PFAS (excluding TFA) are shown in (b) and (d), respectively.

contributing over 85%. Therefore, this pattern may largely reflect the spatial distribution of TFA and obscure the distribution patterns of other PFAS. To this end, TFA was excluded from the total to more accurately characterize the spatial distribution and composition of the remaining PFAS (Fig. 3c and d). The results showed that PFASs (30.3–85.1%) and C3–C14 PFCAs (7.55–42.2%) accounted for relatively large proportions at most sampling stations. The Σ_{23} PFAS concentrations ranged from 7.27 to 88.3 ng/L, with the highest levels observed near stations Suo7 and Suo8 on the northern coast. Moreover, the highest concentrations of most PFAS classes and individual compounds were found in this area (Fig. S3). This pattern may be attributable to the influence of industrial zones along the northern coast, where PFAS and other pollutants may enter coastal waters through riverine inputs and surface runoff (Li et al., 2026b; Tanaka et al., 1991). In contrast, the distribution of TFA may be primarily influenced by atmospheric wet deposition and surface runoff, which may explain its distinct spatial pattern relative to other PFAS (Frank et al., 2002; Solomon et al., 2016).

Diagnostic ratios are commonly used to infer potential point-source influences on PFAS contamination, with ratios exceeding 1 generally indicating PFOS-dominated profiles in source identification studies (Armitage et al., 2009; So et al., 2004). Here, the calculated PFOS/PFOA ratios were used to identify areas potentially influenced by PFOS releases (Fig. S4). The results showed that all stations had PFOS/PFOA ratios greater than 1, indicating a PFOS-dominated profile throughout the study area, consistent with the widespread occurrence of PFOS documented above. Among them, stations Iyo1, Suo6, and Beppu1 exhibited ratios exceeding 10, indicating stronger PFOS enrichment relative to PFOA and suggesting potential PFOS source influences in the vicinity of these locations. Notably, these stations are located near several major rivers that discharge into the sea, such as the Asa River, Kotou River, and Saba River on the southern coast of Yamaguchi Prefecture, as well as the Oita River and Ono River north of Oita City (Amano et al., 2011; Magome and Isobe, 2003; Zhu et al., 2019). These rivers flow through industrial zones, airports, ports, and densely populated areas, and have historically been associated with elevated concentrations of heavy metals, brominated flame retardants (BFRs), and polychlorinated biphenyls (PCBs) (Amano et al., 2011; Hoang et al., 2021; Li et al., 2025; Zhu et al., 2019). Therefore, these rivers may represent important pathways for pollutant inputs to the WSIS. Although PFOS has long been listed under the Stockholm Convention for regulatory control, its environmental persistence means that legacy PFOS may still enter the water column through riverine transport and sediment resuspension, contributing to PFOS contamination in the region.

Given the limitations of the single-cruise sampling design, the present results represent a snapshot of PFAS contamination in the WSIS, and seasonal variations driven by precipitation and river discharge were not captured. Nevertheless, these findings provide a comprehensive baseline dataset on legacy, emerging, and ultrashort-chain PFAS (USC-PFAS) in this region and highlight the need for future multi-season investigations.

3.3. Environmental correlation and source analysis

3.3.1. Environmental behavior and internal correlation of PFAS

The relationships between PFAS concentrations and the number of carbon atoms were analyzed using linear and quadratic polynomial regression (Fig. S5). The results showed that PFAS concentrations were significantly and negatively correlated with the number of carbon atoms ($p < 0.001$, $R^2 = 0.27$). When analyzed by PFAS class, PFASs showed no significant linear relationship with carbon chain length ($p > 0.05$, $R^2 = 0.01$). In contrast, PFCAs exhibited a pronounced U-shaped trend, with concentrations decreasing and then increasing with carbon chain length ($p < 0.001$, $R^2 = 0.80$). Notably, PFECAs, as alternatives to PFCAs, displayed a similar but weakened relationship ($p < 0.05$, $R^2 = 0.30$). Previous studies have indicated that PFECAs exhibit different characteristics from PFCAs in terms of environmental

behavior, bioaccumulation, and trophic transfer (Li et al., 2023; Li et al., 2022). This underscores the importance of incorporating emerging PFAS into future studies to better understand and characterize their environmental behavior.

RDA was applied to examine the relationships between PFAS detected at frequencies exceeding 40% and environmental factors, such as nutrients and TOC. Except for PFMeS, which was associated with $\text{NH}_4\text{-N}$, no other PFAS exhibited strong correlations with the environmental factors (Fig. S6). This finding is consistent with a dominant influence of industrial inputs on PFAS distributions in the WSIS. Spearman's rank correlation analysis was then performed to examine correlations among PFAS (Fig. 4). The results showed that PFCAs, including PFBA, PFUnDA, PFOA, and perfluorohexanoic acid (PFHxA), were significantly positively correlated with each other ($p < 0.05$), with correlation coefficients (r) ranging from 0.55 to 0.83. In particular, several pairs, including PFBA–PFHxA, showed highly significant correlations ($p < 0.001$). Strong positive correlations among contaminants generally suggest shared sources or similar environmental behavior, supporting further source apportionment analysis (Hua et al., 2021; Zhang et al., 2022). Interestingly, TFA was negatively correlated with most individual PFAS ($r < 0$), particularly with 6:2 Cl-PFESA ($r = -0.64$); however, none of these correlations remained statistically significant after multiple-comparison correction. These results suggest that TFA in the study area may originate from sources distinct from those of other PFAS, potentially including atmospheric deposition. This is consistent with the findings of Taniyasu et al. (2008) and Wang et al. (2024), who reported TFA in rainwater samples from Japan, China, and South Korea. This view is also supported by Frank et al. (2002), who detected TFA in precipitation over the Southern Ocean. Together, these results support atmospheric deposition as a potential contributor to elevated TFA concentrations in the study area. TFA is known to be a terminal degradation product of several fluorinated gases, such as HFC-134a, HCFC-123, and HFO-1234yf, which are released from industrial activities, refrigeration, and air conditioning systems. Atmospheric oxidation of these precursors leads to the formation of TFA, which is subsequently removed through wet and dry deposition (Burkholder et al., 2015; Wang et al., 2024). Therefore, regional emissions of these F-gases may serve as a potential source of TFA in the WSIS.

3.3.2. Quantitative source analysis based on PCA-MLR

The correlation results preliminarily suggested that PFAS in the WSIS may have identifiable and quantifiable source contributions. Therefore, PCA-MLR was employed to identify and quantify the major sources and their contributions. TFA was excluded from the PCA-MLR analysis because the RDA and correlation analysis results suggested that its sources differed from those of the other PFAS and may have been influenced primarily by atmospheric deposition rather than direct industrial inputs. Principal components (PC) with eigenvalues greater than 1 were extracted using the PCA model, and the factor matrix was rotated using the varimax orthogonal rotation method. Two principal components were retained (Table 1 and Fig. S7), cumulatively explaining 69.0% of the total variance. Notably, the third principal component had an eigenvalue of 0.95 (< 1) and was therefore not retained according to the Kaiser criterion. Thus, the two-component solution was retained for its simplicity and interpretability. The remaining 31.0% of the variance may reflect unidentified mixed sources, measurement noise, collinearity among PFAS variables, and the limited sample size. PC1 explained 37.8% of the total variance and was mainly characterized by high loadings of PFOS, 6:2 Cl-PFESA, and perfluorodecanoic acid (PFDA). PFOS-containing mist suppressants were commonly used in the electroplating industry for controlling hexavalent chromium emissions. Following restrictions on PFOS, 6:2 Cl-PFESA was subsequently produced and used as an alternative (Xiao, 2017; Xiao et al., 2012). Therefore, PC1 was interpreted as an electroplating-related source (Source 1). PC2 explained 31.2% of the total variance, with high loadings for HFPO-TA (C8) (0.83) and PFOA

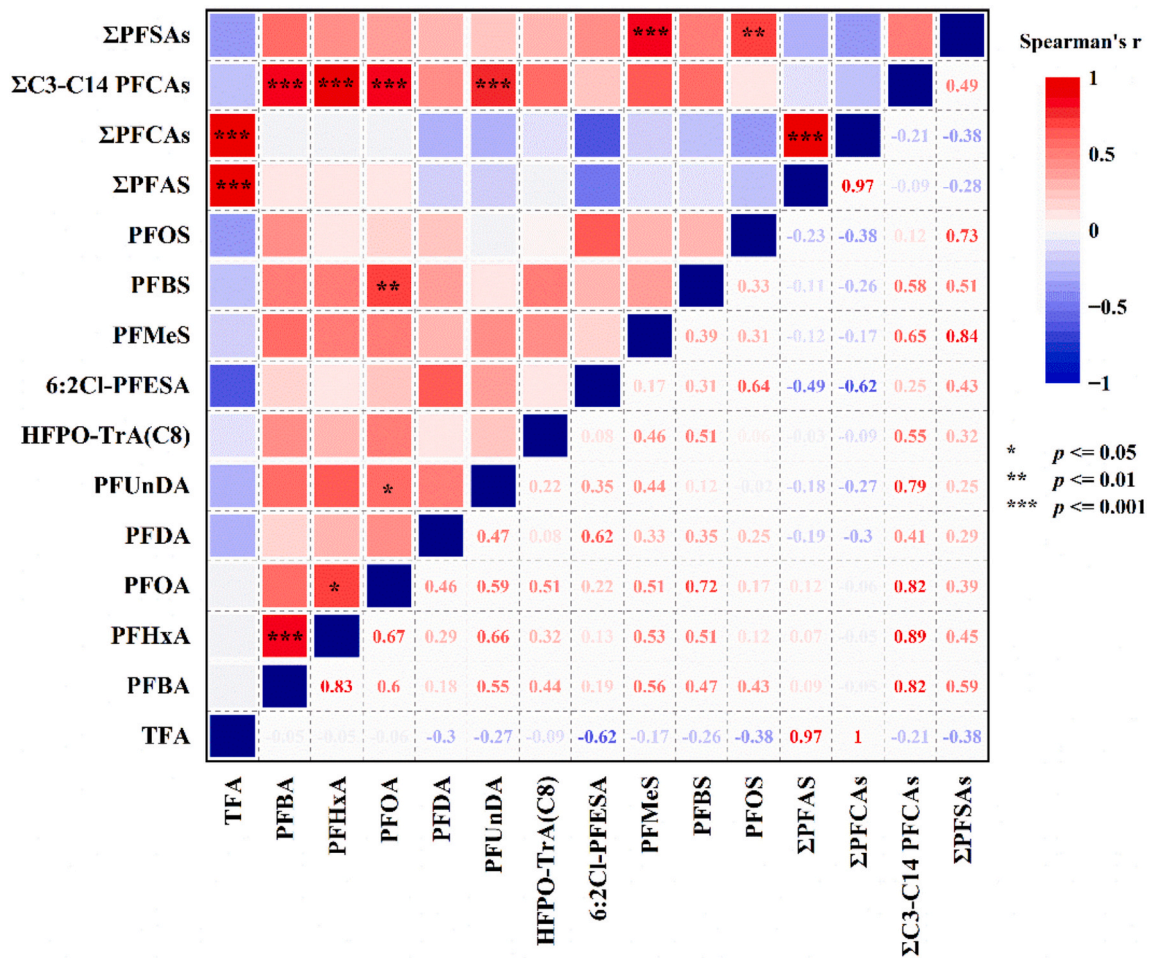


Fig. 4. Heatmap of Spearman's rank correlation coefficients among individual PFAS. The numbers in the cells represent Spearman's r values. The color scale indicates the strength and direction of the correlations, ranging from -1 (dark blue, strong negative correlation) to 1 (dark red, strong positive correlation). Asterisks denote statistical significance: * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
Factor loadings of individual PFAS on the extracted principal components.

PFAS	PC1 (37.8%)	PC2 (31.2%)
PFBA	0.62	0.66
PFHxA	0.60	0.67
PFOA	0.49	0.70
PFDA	0.79	0.07
PFUnDA	0.61	0.31
HFPO-TA(C8)	-0.23	0.83
6:2 Cl-PFESA	0.96	0.07
PFMeS	0.19	0.67
PFBS	0.57	0.68
PFOS	0.70	0.21

Note: Loadings with absolute values ≥ 0.70 are shown in bold.

(0.70). PFOA was widely used as a surfactant and processing aid. Following restrictions on PFOA, HFPO-based chemicals such as hexafluoropropylene oxide dimer acid (HFPO-DA) and HFPO-TA, were increasingly used as alternatives (Buck et al., 2011; Pan et al., 2017). Therefore, PC2 was interpreted as a source associated with surfactant-related applications (Source 2).

The MLR model was then used to calculate the contribution rates of each factor, and the results are presented in Table 2. Within the PCA-MLR model, electroplating-related and surfactant-related sources accounted for 51.2% and 48.8% of the apportioned PFAS, respectively. Japan is an important global producer of fluorochemicals, which may

Table 2
Source apportionment results and contribution rates from PCA-MLR.

Source	Source type	Marker PFAS	Contribution (%)
1	Electroplating-dominated	PFOS, 6:2 Cl-PFESA and PFDA	51.2
2	Surfactant-dominated	PFOA and HFPO-TA(C8)	48.8

contribute to the observed industrial source pattern (Miller et al., 2026). Thus, the distribution of PFAS-related industries and facilities may help explain the observed source pattern.

3.4. Substitution progress of emerging PFAS

The ratio of emerging PFAS to legacy PFAS has been increasingly used as a semi-quantitative indicator of PFAS substitution in the environment. Previous studies have shown that concentration ratios of emerging to legacy PFAS can reflect substitution trends and may also serve as indicators for distinguishing among water masses (Li et al., 2026a; Pan et al., 2018). Studies of PFAS contamination associated with industrial sources in China further support the link between fluorochemical industrial parks and PFAS contamination in receiving rivers and bays, such as the Xiaoqing River-Laizhou Bay and Daling River-Bohai Sea systems, where PFAS concentrations and mass loadings

have been strongly associated with industrial discharges (Ma et al., 2025; Song et al., 2018; Wang et al., 2016). Some short-chain PFAS were excluded from this analysis because they may originate not only from the direct use of PFCA-containing products but also from indirect sources. Therefore, only PFECAs and PFESAs were classified as emerging PFAS and included in the analysis, as these groups represent major ether-oxygen-containing PFAS alternatives. Overall, the ratio of emerging to legacy PFAS was less than 1, indicating that emerging alternatives remained less abundant than legacy PFAS and suggesting that substitution was incomplete in this region (Fig. 5). Subcategory analysis further revealed differences between the two substitution pathways. For the PFCA pathway, the ratio of the representative emerging alternative HFPO-TA to PFOA was 0.15, indicating that HFPO-TA remained substantially less abundant than PFOA and suggesting that substitution along this pathway was still limited. For the PFSA pathway, the ratio of the representative alternative 6:2 Cl-PFESA to PFOS was 0.37, suggesting a greater degree of substitution than that observed for the PFCA pathway, although the abundance of the emerging alternative remained substantially lower than that of PFOS.

The replacement ratios for PFOA and PFOS calculated for each subregion revealed distinct apparent substitution patterns among different stations and areas (Fig. S8). Specifically, the relative abundance of the emerging PFOA alternative remained low at both the northern and southern coasts (e.g., stations Suo4 and Suo7). For PFOS, the emerging-to-legacy PFAS ratios at the northern coast (e.g., stations Suo6 and Suo7) reached their lowest values, with replacement ratios of only 0.19 and 0.25, indicating pronounced substitution lags. In contrast, at offshore stations farther from the potential industrial source areas (e.g., Suo2 and Suo9), the corresponding ratios were relatively higher (> 0.5). This pattern does not necessarily indicate faster substitution offshore. Rather, the higher ratios offshore may result from the preferential association of legacy long-chain PFAS (e.g., PFOS) with particles and their removal from the water column through nearshore settling, together with dilution of the point-source plume with increasing distance offshore. Consequently, their relative concentrations in the offshore water column decrease significantly, which passively elevates the relative proportions of emerging alternatives (Li et al., 2024; Pan and You, 2010).

Importantly, this ratio is a semi-quantitative proxy rather than a direct measure of industrial production or use. It is influenced by source strength, riverine and atmospheric inputs, transport, dilution, degradation, and partitioning between the water and particle phases. Therefore, it reflects the relative occurrence of emerging and legacy PFAS and

their apparent substitution trend in the water column, but cannot be directly converted into absolute industrial replacement rates. Nonetheless, several studies have reported that emerging PFAS such as HFPO-TA and 6:2 Cl-PFESA are persistent in the environment and may undergo bioaccumulation and trophic transfer through food webs (Li et al., 2022; Pan et al., 2017; Shi et al., 2018). Therefore, the potential environmental risks associated with current substitution strategies warrant continued attention, and long-term monitoring of these compounds.

3.5. Preliminary ecological risk assessment

The risk quotient (RQ) approach was applied to conduct a preliminary ecological risk assessment for the study area. In accordance with the principle of protecting the most sensitive species, the lowest chronic no-observed-effect concentration (NOEC) values available in the toxicity database were selected to derive the predicted no-effect concentration (PNEC), and RQs were calculated using the maximum measured PFAS concentrations at each station (European Chemicals Agency, 2008). Substantially different PNEC values for TFA in the toxicity database introduced considerable uncertainty into the ecological risk assessment. As shown in Fig. S9, the RQs for TFA calculated based on four different PNEC values spanned multiple orders of magnitude. The highest RQ value reached 7.57, indicating high ecological risk, while the lowest fell into the negligible risk range ($RQ < 0.01$). This substantial discrepancy highlights the large variation in species sensitivity to TFA across trophic levels, as well as the limited toxicity data available for TFA in marine environments. Therefore, to ensure adequate protection for organisms across all trophic levels, we adopted the most sensitive PNEC value for subsequent risk characterization.

The RQ results (Fig. 6) indicated that TFA was associated with high ecological risk ($RQ > 1.0$), PFOS was associated with low ecological risk ($0.01 < RQ \leq 0.1$), and the remaining PFAS were associated with negligible risk ($RQ \leq 0.01$). Notably, 6:2 Cl-PFESA, an emerging alternative, had the third-highest RQ value among the analyzed PFAS, after TFA and PFOS. Given that the substitution of PFOS remains incomplete, its continued environmental presence may make it a potential future risk contributor despite its relatively low current concentrations. At the station level (Fig. S10), most PFAS, except TFA and PFOS, had $RQs \leq 0.01$ at all stations. PFOS posed a low ecological risk ($0.01 < RQ \leq 0.1$) at all stations, whereas TFA posed with high ecological risk ($RQ > 1$) at all stations. Based on the total of RQ (ΣRQs), all sampling stations exhibited high ecological risk, with TFA

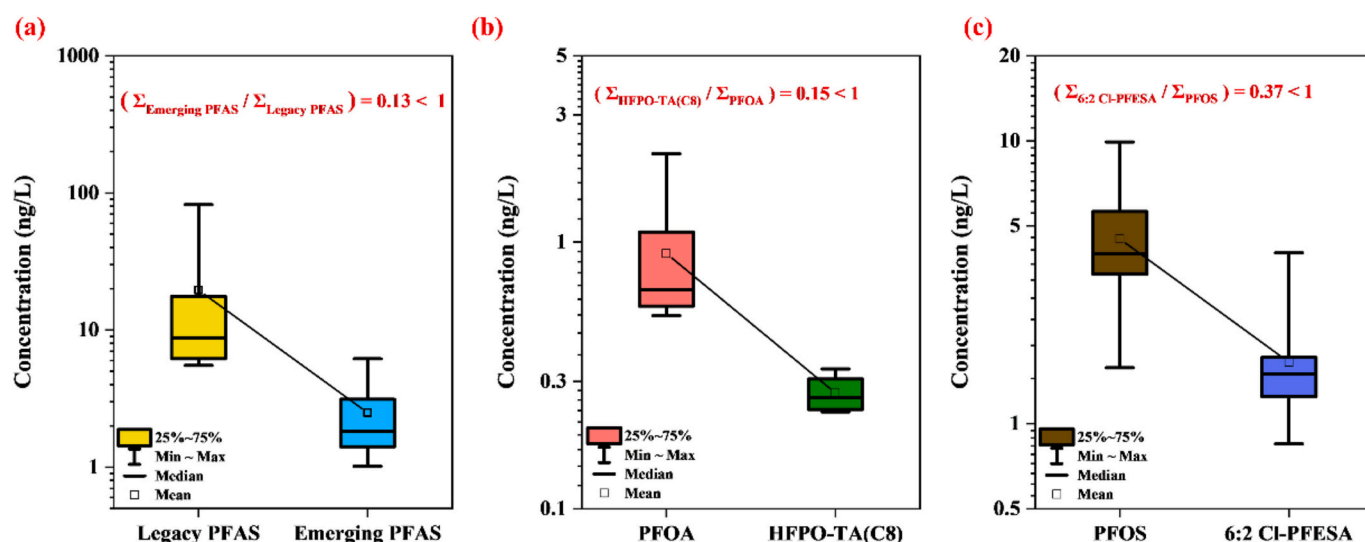


Fig. 5. Concentration ratios of emerging to legacy PFAS. (a) $\Sigma_{\text{Emerging PFAS}} / \Sigma_{\text{Legacy PFAS}}$, (b) $\Sigma_{\text{HFPO-TA(C8)}} / \Sigma_{\text{PFOA}}$ and (c) $\Sigma_{\text{6:2 Cl-PFESA}} / \Sigma_{\text{PFOS}}$.

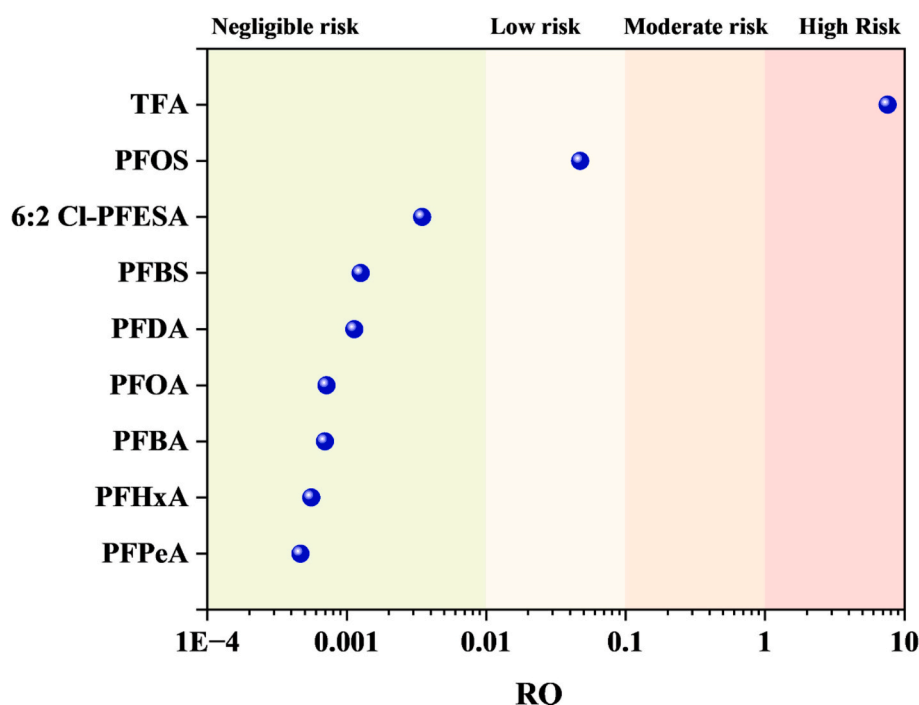


Fig. 6. RQ and risk levels of individual PFAS.

representing the dominant contributor to the summed risk. Similar findings have been reported in other TFA-contaminated regions in China, including the Xiaoqing River estuary and the Yangtze River Delta (Ma et al., 2025; Miao et al., 2025). Together with previous studies, our findings highlight the need for continued assessment of the potential ecological implications of TFA in aquatic environments, particularly coastal ecosystems.

In addition, the toxic effects of emerging PFAS such as HFPO-TA and 6:2 Cl-PFESA on marine organisms are being increasingly revealed (Deng et al., 2018; Wu et al., 2022; Yi et al., 2019). Therefore, the potential combined ecological effects of these compounds warrant further attention.

4. Conclusions

This study systematically investigated the occurrence of PFAS in the WSIS, Japan, revealing distinctive contamination patterns, potential sources, and an ecological risk profile characteristic of this highly industrialized semi-enclosed marine system. A total of 24 PFAS were identified and quantified, with TFA overwhelmingly dominating the total PFAS concentrations and highlighting a potential gap in current marine PFAS monitoring. After excluding TFA, PFSA were the predominant PFAS group in the seawater of the study area. TFA showed a relatively widespread and spatially heterogeneous distribution across the study area, potentially influenced by atmospheric deposition and surface runoff. In contrast, the other PFAS showed a clear decreasing gradient from nearshore to offshore waters, with higher concentrations predominantly observed along the northern coast. PCA-MLR identified electroplating-related and surfactant-related sources as the major potential sources of non-TFA PFAS. By contrast, TFA was excluded from the PCA-MLR analysis, and its potential atmospheric influence was inferred from correlation patterns, spatial distribution, and supporting literature evidence. This distinction highlights the need for source-specific monitoring and management strategies for legacy, emerging, and ultrashort-chain PFAS (USC-PFAS). The ratios of emerging to legacy PFAS indicated incomplete and spatially heterogeneous substitution in the study area. A conservative risk assessment based on the most sensitive PNEC values showed that TFA was posed a high ecological risk

($RQ > 1.0$) at all sampling stations and was the dominant contributor to the total RQ. Future efforts should include multi-season and multi-compartment monitoring in this region and incorporate representative USC-PFAS, such as TFA, into routine monitoring programs in other marine areas. Furthermore, expanded toxicity testing, particularly with marine organisms, is needed to better characterize the ecological risks of TFA and support risk management.

CRediT authorship contribution statement

Wenke Dou: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Wei Ma:** Methodology, Formal analysis, Data curation. **Zhen Qiao:** Methodology, Investigation, Data curation. **Jun Zhao:** Methodology, Investigation. **Xinyu Guo:** Supervision, Resources, Project administration, Investigation. **Yitao Pan:** Methodology, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Jianhui Tang:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, 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.marpolbul.2026.120466>.

Data availability

Data will be made available on request.

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