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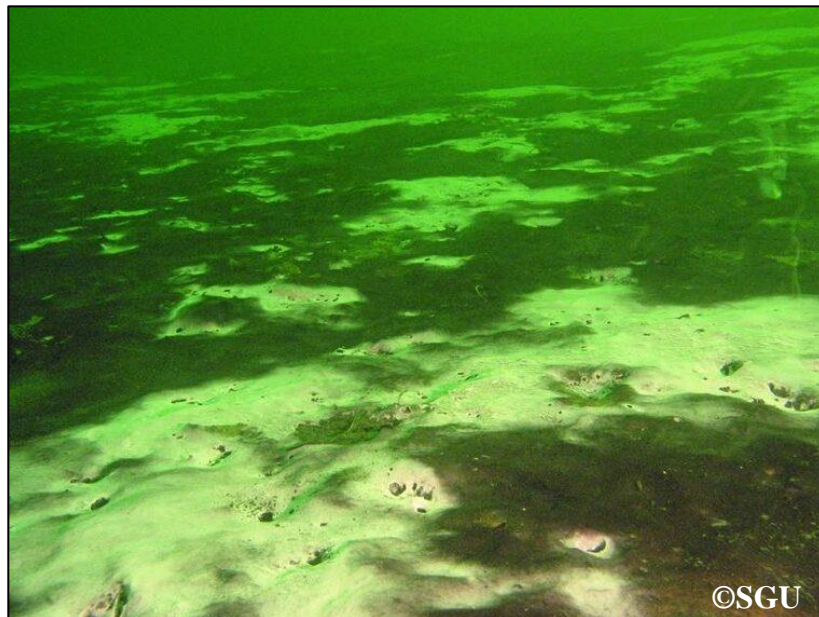
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PFAS distribution and sorption dynamics in Swedish offshore sediments

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PFAS distribution and sorption dynamics in Swedish offshore sediments

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Sammanfattning <p>Förekomsten av PFAS i utsjösediment runt Sverige har blivit tydlig på senare år och väckt frågor om de underliggande orsakerna. Denna studie undersökte förekomsten och sorptionsmönstren av PFAS i svenska utsjösediment, med fokus på regionala skillnader genom sorptionstester. Förhöjda PFAS-koncentrationer påträffades i Bottenviken, där kortkedjade PFCAs var dominerande. Inget enhetligt bevis för ökad sorptionsförmåga i norra sediment observerades, vilket tyder på närvaron av distinkta och potentiellt oidentifierade PFAS-källor i regionen, vilket motiverar ytterligare undersökning. Framtida övervakningsstudier är avgörande för att lokalisera potentiella inre källor till PFAS-förorening, med hänsyn till gränsöverskridande förorening från olika baltiska regioner. Även om ytterligare sorptionsstudier kan förbättra förståelsen, bör prioritet ges åt att undersöka PFAS-ingångar i Östersjön, inklusive tillflöde från biflöden.</p>	

Summary

The prevalence of per- and polyfluoroalkyl substances (PFASs) in offshore sediments around Sweden has become evident in recent years raising questions on the underlying reasons. This study examined the occurrence and sorption patterns of PFASs across Swedish offshore sediments, focusing on regional disparities through sorption tests. Elevated PFAS concentrations were found in the Bothnian Gulf, confirming previous investigations. No consistent evidence of heightened sorption affinity in northern sediments was observed, suggesting the presence of distinct and potentially unidentified PFAS sources in the region, warranting further investigation. Future monitoring studies are crucial for pinpointing potential inland sources of PFAS contamination, considering transboundary contamination from different Baltic regions. While additional sorption studies could enhance understanding, priority should be given to investigating PFAS influxes into the Baltic, including tributary loading.

Sammanfattning

Förekomsten av PFAS i utsjösediment runt Sverige har blivit tydlig på senare år och väckt frågor om de underliggande orsakerna. Denna studie undersökte förekomsten och sorptionsmönstren av PFAS i svenska utsjösediment, med fokus på regionala skillnader genom sorptionstester. Förhöjda PFAS-koncentrationer påträffades i Bottenviken. Inget enhetligt bevis för ökad sorptionsförmåga i norra sediment observerades, vilket tyder på närvaron av distinkta och potentiellt oidentifierade PFAS-källor i regionen, vilket motiverar ytterligare undersökning. Framtida övervakningsstudier är avgörande för att lokalisera potentiella inre källor till PFAS-förorening, med hänsyn till gränsöverskridande förorening från olika baltiska regioner. Även om ytterligare sorptionsstudier kan förbättra förståelsen, bör prioritet ges åt att undersöka PFAS-ingångar i Östersjön, inklusive tillflöde från biflöden.

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Abbreviations list

Term	Definition
PFAS	Per- and polyfluoroalkyl substances
PFAAs	Perfluoroalkyl acids
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonic acid
PFCA	Perfluorocarboxylic acid
PFDA	Perfluorodecanoic acid
PFHpA	Perfluoroheptanoic acid
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexane sulfonic acid
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFOSA	Perfluorooctanesulfonamide
PFPeA	Perfluoropentanoic acid
PFSA	Perfluorosulfonic acid
PFUnDA	Perfluoroundecanoic acid
SAR	Sediment accumulation rate
TC	Total carbon
TN	Total nitrogen
TOC	Total organic carbon
TS	Total solids

1 INTRODUCTION

Marine sediments hold significant value as indicators of environmental quality and anthropogenic pressures. Sweden's extensive coastal expanse is dominated by sediment deposits, providing both historical data on environmental degradation, as well as predictions for future changes.

Swedish offshore sediments are delineated by the Baltic Sea to the east and the North Sea to the west (Figure 1). The Baltic Sea, is a partially confined basin of brackish water, that emerged some 10,000 years ago during the last glacial era (Tuuling et al., 2011). Spanning an area of roughly 415,000 km² and a mean depth of only 55 meters, it is bordered by Denmark, Estonia, Finland, Germany, Latvia, Lithuania, Poland, Russia, and Sweden. Its only connection to the Atlantic Ocean is through the Danish straits to the North Sea. Unique features such as low salinity, pronounced vertical stratification, and substantial freshwater inflows, contribute to its distinctive ecological profile. The Baltic Sea is home to around 1,000 marine species in the west, decreasing to only around 50 species in the northern and eastern reaches. This biodiversity gradient is driven by the Baltic Sea's brackish nature, as salinity declines from west to east.



Figure 1. Overview of the Baltic and North Seas, surrounding the Scandinavian peninsula (source: EMODnet).

Over the last several decades, the Baltic and North Seas have experienced significant environmental deterioration, marked by eutrophication, algal blooms, and the accumulation of persistent organic pollutants and heavy metals (Kanwischer et al., 2022). This degradation stems from various sources, including historical conflicts like World Wars I and II, industrial activities, and agricultural practices, which have introduced harmful substances into the sea

through its tributaries. The catchment zone of the Baltic Sea covers an area of 1,720,270 km², which is approximately four times larger than the sea itself and spans roughly half of Europe. This vast expanse of the catchment zone amplifies the terrestrial influences on the marine environment (HELCOM, 2004). Specifically, river influxes are one of the most significant contributors to the Baltic Sea's contamination, as they funnel an annual average of around 660 km³ of freshwater into the sea, carrying substantial quantities of fertilizers and other pollutants (Nguyen et al., 2017). Deposition of contaminants, including metals and organic contaminants in the Baltic by airborne transport has been reported previously (Kanwischer et al., 2022). In addition, the coastal regions of the Baltic continue to host a significant presence of chemical businesses, including the paper industry and the metallurgy sector, contributing to ongoing environmental challenges.

Seabed sediments have an important role in storing and retaining pollutants, acting as contamination sinks. Nevertheless, there is a lack of thorough understanding of the concentration and distribution of contaminants within these sediments. The potential resuspension of sediments, such as through dredging activities, poses a risk of remobilizing contaminants, thereby threatening marine life and ecosystem quality (Moreira et al., 2021; Vanavermaete et al., 2023). In addition, climate change is expected to incur increased sedimentation rates, temperature changes, acidification, and deoxygenation (Meier et al., 2022). All of these shifts will have uncertain consequences on the fate and transport of the contaminants stored within seabed sediments.

Known for its substantial pollution, the Baltic Sea has been extensively monitored over the years. While the presence of legacy pollutants has been established in the Baltic Sea, there is also presence of emerging contaminants, such as per- and polyfluoroalkyl substances (PFASs) (Ahrens et al., 2010; Filipovic et al., 2013). PFASs are a large group of synthetic chemicals that have become ubiquitous environmental contaminants worldwide. Within the Baltic Sea region, PFASs have emerged as a concerning environmental pollutant, reflecting anthropogenic activities prevalent in the surrounding countries. For instance, high PFAS concentrations have been reported in fish of the Bothnian Gulf (Kumar et al., 2022). Understanding the occurrence and fate of PFASs in sediments of the Baltic Sea and North Sea is crucial for developing effective management strategies to mitigate their impacts.

One of the latest monitoring efforts in the region, conducted by the Geological Survey of Sweden (SGU) and published in May 2022, scrutinized the presence of pollutants, including PFASs, in offshore sediments along Sweden's coastline (Josefsson, 2022). The research focused primarily on the analysis of organic pollutants and components present in sediment samples collected from 16 monitoring stations situated in offshore regions (see also Section 2.1). The selection of these sampling locations was made in order to mitigate the direct impact of specific pollution sources. The SGU report emphasized a substantial variation in PFAS levels among the sites, with particularly elevated concentrations observed in the sediments of the Bothnian Gulf (ranging from 12-26 µg kg⁻¹ of dry weight (dw)) in comparison to other areas (ranging from 0.41-2.4 µg kg⁻¹ dw). This conclusion was unexpected, as the levels of organic contaminants in the Gulf of Bothnia are generally lower than those in the Baltic Proper. Additionally, prior research has demonstrated that the levels of PFASs in saltwater or biota within the northern Baltic Sea are comparatively lower than those found in the southern parts (Mikolajczyk et al., 2023).

According to SGU, there was a preliminary investigation of PFASs in 2014, which indicated the presence of sediment contamination, and additional assessments done in 2020/2021 further confirmed this finding. The SGU then conducted a more in-depth examination of the dispersion of various PFASs in the sediments, observing notable disparities between the Gulf of Bothnia and other locations. Specifically, perfluorononanoic acid (PNFA), perfluorooctanoic acid (PFOA), and perfluorooctanesulfonate (PFOS) were the prevailing compounds in the Gulf of Bothnia, whereas perfluoroundecanoic acid (PFUnDA) and PFOS were the main substances in other regions.

These disparities prompted questions about their underlying reasons, leading to the aim of this study: to elucidate why PFAS concentrations in Bothnian Gulf sediments surpass those in the southern Baltic Sea. The primary research question addressed the sorption capacity of different sediment samples for PFASs. To answer this, a simple sorption batch study tested the hypothesis that sediments in the southern Baltic have a higher sorption capacity. Sediment – pore-water partitioning coefficients (K_d values) and organic-carbon distribution coefficients (K_{OC}) were determined for each Baltic region.

2 METHODOLOGY

2.1 SAMPLING AND SAMPLE PREPARATION

SGU conducted sampling operations in 2014 and 2020/2021 with their research vessel. At various stations, a 50-meter radius circle was established, within which surface sediments were collected from seven locations (one at the circle's center and six distributed randomly). For the assessment of organic contaminants, the seven samples from each station were combined to create a single composite sample per site.

Sampling conditions were recorded, including environmental factors such as dissolved oxygen levels, conductivity, temperature, salinity, and others. Due to storage issues, several samples (from SE-5 to SE-16) collected in 2020 were compromised, prompting a follow-up sampling campaign in 2021 for stations 6 to 16; station 5 however, could not be revisited for sampling at the time.

SGU supplied a preliminary collection of wet surface sediment samples gathered in 2020/2021. These samples were kept in polypropylene (PP) bottles at a temperature of -20°C . The samples were subjected to an 8-day freeze-drying procedure to remove the water content. The freeze-drying process, conducted at temperatures below -40°C and pressure below 0.6 mbar, enabled the elimination of water by sublimation.

Due to the scarcity of sediment material, it was necessary to combine the samples for the purposes of the batch sorption tests (see Section 2.2). Composite samples were prepared by combining equal mass of samples of different locations. The grouping was determined according to the respective areas in the Baltic and North Seas, as follows (see also Figure 2):

- Bothnian Bay: SE-1 and SE-17
- Bothnian Sea: SE-2 and SE-3
- Baltic Proper: SE-6, SE-7, SE-8, SE-9, and SE-10
- Southern Baltic: SE-11 and SE-12
- North Sea: SE-13, SE-14, and SE-15

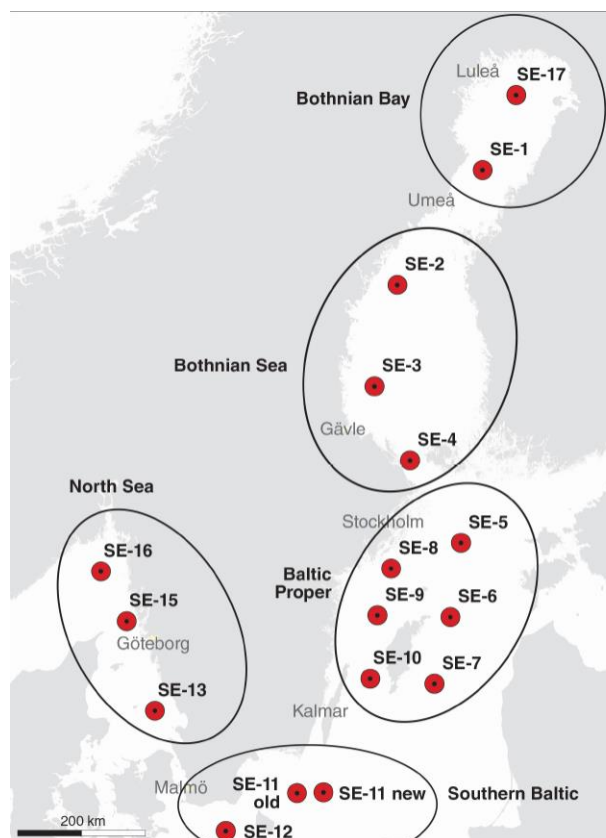


Figure 2. The national-level monitoring of offshore sediments encompasses 16 stations distributed along Sweden's coastline, as depicted by the encircled red markers representing their respective sea areas. Note that Station SE-11 was relocated between 2008 and 2014. Source: (Josefsson, 2022).

2.2 SORPTION UPTAKE TESTS

To assess the adsorption capacity of sediments across different regions, sorption uptake batch tests were conducted, following a methodology outlined elsewhere (Niarchos et al., 2023), with a few modifications detailed in the following.

To mimic natural conditions, artificial seawater was formulated by mixing sodium chloride (NaCl) with Millipore water, achieving a salinity of 5 g L^{-1} , representative of median concentrations in the bottom of the Bothnian Gulf (Apler and Josefsson, 2016). However, it should be noted that the salinity is lowest in the northern and eastern parts of the Baltic Sea, such as the Bothnian Bay, due to high freshwater input from rivers (Radtke et al., 2020).

For estimating the sorption of various PFASs, a spiking solution was prepared, combining equimolar concentrations of: PFPeA, PFHxA, PFOA, PFNA, PFDA, PFUnDA, PFBS, PFHxS, PFOS, PFOSA. The selection of the spiking concentration was based on literature data, to reflect the higher end of environmentally relevant concentrations. According to bibliographic data, PFBA concentrations of up to 640 ng L^{-1} have been observed in the water phase of sediment studies, whereas PFBA values of $10.7 \text{ ng g}^{-1} \text{ dw}$ have been found in riverine sediments (Campo et al., 2016). This value has been chosen as a representative concentration for individual PFAS concentrations, using equal molarities for each PFAS in the spiking

mixture¹. Each PFAS in the final spiking solution had a molar concentration of 0.5 $\mu\text{mol L}^{-1}$. These spiking concentrations were much higher than the native contamination concentrations, as can be seen from the screening results in Section 3.1.

To perform the sorption tests, 1 g (± 0.17) of freeze-dried sediment was added into a 15 mL PP-tube, followed by mixing with 9.9 mL of synthetic seawater and 100 μL of the equimolar-spiking-mix, resulting in a 10:1 liquid:solid ratio. Each sample was triplicated and each tube was vigorously shaken in an end-over-end motion for a duration of 7 days at a speed of 64 revolutions per minute, to reach equilibrium (Kupryianchyk et al., 2016). Subsequently, the tubes were centrifuged at a speed of 3000 rpm for a duration of 15 minutes and after centrifugation, resulting in separation of the sediment and liquid portion (supernatant). The sediment and aqueous phases were then analysed for their PFAS content respectively, as described in Section 2.3.

Sediment – pore water distribution coefficients (K_d values) were determined, reflecting sorption between sediment and liquid phases at equilibrium. The sorption coefficients were calculated as:

$$K_d = \frac{C^*}{C_w} \quad (\text{L kg}^{-1}),$$

where C^* is the individual PFAS concentration in the sediments, and C_w is the individual PFAS concentration in the water phase. The native contamination estimated from the sediment screening was subtracted from the final calculations of K_d values.

Table 1. Total nitrogen (TN), total carbon (TC), and total organic carbon (TOC) concentrations as % of total solids (TS) in the different regions investigated. The standard deviation of TOC (TOC SD) is also presented, along with the carbon to nitrogen ratio (C/N). Source: (Josefsson, 2022).

Region	TN	TC	TOC	TOC SD	C/N
	(% TS)	(% TS)	(% TS)		
North Sea	0.2	3.2	2.2	0.1	10.8
Southern Baltic	0.8	6.7	6.6	0.7	8.0
Baltic Proper	1.5	12.8	12.7	1.0	8.4
Bothnian Sea	0.3	2.9	3.0	0.0	10.7
Bothnian Bay	0.4	4.6	4.5	0.2	11.2

Additionally, organic carbon-normalised partitioning coefficients were calculated. Variations in organic carbon content at different regions of the Baltic have been observed (Table 1) and K_{OC} values take such variations into account.

The sorption to organic carbon was estimated through organic carbon distribution coefficients (K_{OC}), according to:

$$K_{OC} = \frac{K_d}{f_{OC}} \quad (\text{L kg}^{-1}),$$

¹ Although PFBA was not included in the intended PFAS selection, PFPeA was chosen as the compound with the most similar molecular weight in order to calculate the molarity of each PFAS.

where f_{oc} (kg kg^{-1}) describes the % fraction of organic carbon in the sediments.

2.3 PFAS ANALYSIS

The PFAS analysis targeted a range of 28 compounds, including: seven perfluoroalkane sulfonic acids (C4-9 PFASs) (PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFNS, and PFDS), 11 perfluoroalkyl carboxylates (C3-13 PFCAs) (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA), three n:2 fluorotelomersulfonic acids (4:2 FTSA, 6:2 FTSA, 8:2 FTSA) two compounds of F-53 B (9Cl-PF3ONS, 11Cl-PF3OUdS), GenX (HFPO-DA), FOSA, MeFOSAA, EtFOSAA, and NaDONA.

For the assessment of sorption experiments, only the spiked PFAS compounds were considered, including PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFBS, PFHxS, PFOS, and PFOSA.

After the batch tests, samples underwent centrifugation at 3000 rpm to separate solid and liquid phases. Subsequently, a portion of the resulting liquid, spiked with an internal standard (IS) and high-performance liquid chromatography (HPLC)-grade methanol, was utilized for PFAS quantification. The mixture then underwent filtration using a $0.45 \mu\text{m}$ recycled cellulose (RC) filter (Sartorius®), before injection and ultra-high-pressure liquid chromatography coupled with tandem mass spectrometry (UHPLC-MS/MS) analysis.

The sediment phase underwent freeze-drying for three days before methanol extraction and analysis. In this process, 1 g of sediment (representing the entire sample) was freeze-dried and then subjected to an extraction process involving HPLC-grade methanol and sodium hydroxide (NaOH) to solubilize and extract PFASs from the sediment matrix. The resulting extracts were concentrated using N₂ blow-down.

Finally, the water and sediment extracts were subjected to analysis using UHPLC-MS/MS. The chromatographic separation was performed on a SCIEX ExionLC AC system equipped with a Gemini® $3 \mu\text{m}$ C18 100 Å column (50 x 2 mm), while detection was carried out using a triple-quadrupole mass spectrometer (SCIEX Triple Quad™ 3500) equipped with an electrospray ionization (ESI) interface operating in negative ionization mode.

3 RESULTS

3.1 PFAS DISTRIBUTION IN OFFSHORE SEDIMENTS

PFAS levels were determined for the composite sediment samples from different regions in duplicates. Out of the 28 screened PFAS compounds, the following were detected in the reference sediments at varying concentrations: PFOA, PFNA, PFDA, PFUnDA, PFBS, PFHxS, PFHpS, and PFOS (Figure 3). Although short-chain PFCAs (PFBA, PFPeA, PFHxA, and PFHpA) were also detected, they were excluded from the analysis due to high levels in the negative blanks. Previous investigations at the site recorded the same compounds (Josefsson, 2022), including PFDS at low levels, which was below detection limits in our study.

The sum of PFAS concentrations ($\sum\text{PFAS}$) in the reference samples varied by region, with the highest concentrations observed in the following order: Bothnian Sea ($33 \pm 21 \mu\text{g kg}^{-1}$) > Bothnian Bay ($27 \pm 11 \mu\text{g kg}^{-1}$) > North Sea ($14 \pm 3.4 \mu\text{g kg}^{-1}$) > Southern Baltic ($13 \pm 6.9 \mu\text{g kg}^{-1}$)

kg⁻¹) > Baltic Proper (12 ± 0.71 µg kg⁻¹) (Figure 3). Elevated PFAS concentrations were particularly notable in the Bothnian Gulf, encompassing the Bothnian Sea and Bothnian Bay, corroborating previous findings by SGU (Josefsson, 2022). It should be noted that, while no standard deviation was reported in the SGU study, our screening revealed high standard deviations for duplicate sediment samples at higher concentration profiles.

The minor differences in PFAS levels between the Bothnian Sea and Bothnian Bay can primarily be attributed to the elevated levels of PFDA in the Bothnian Sea region. Additionally, the Bothnian Sea has a slightly higher sediment accumulation rate (SAR) of 2.2 ± 0.51 yr cm⁻¹, than the Bothnian Bay at 5.95 ± 0.35 yr cm⁻¹ (Josefsson, 2022). This suggests that the Bothnian Sea is more influenced by recent changes in PFAS influxes.

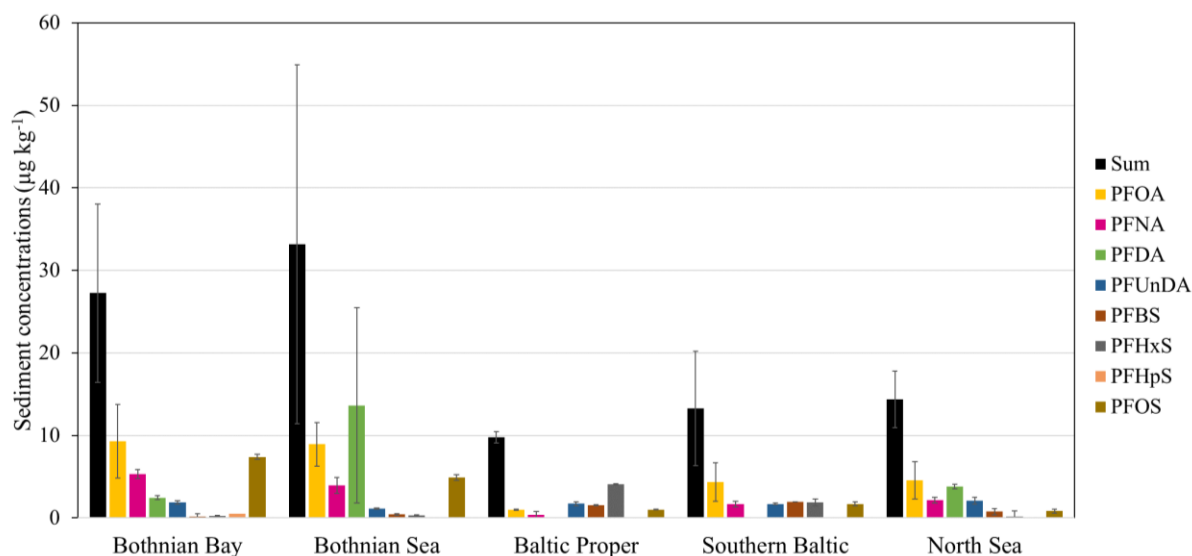


Figure 3. Estimated PFAS concentrations in Swedish offshore sediments (per dw), using composite sediment samples for each region.

3.2 PFAS PARTITIONING ANALYSIS

The PFAS partitioning study aimed to explain trends contributing to heightened PFAS levels in the Bothnian Gulf. Based on the sediment – pore water partitioning coefficients (K_d values) higher sorption affinity in sediments of the north was not consistently evident (Figure 4). Notably, the Baltic Proper exhibited higher K_d values for several PFASs, including PFOS, indicating higher PFAS retention in this region compared to others. However, despite this enhanced retention capacity, sediment-phase PFAS concentrations were the lowest in the Baltic Proper (Figure 3). Consequently, the observed high concentrations in the Bothnian Gulf cannot be directly attributed to inherent differences in sorption capacity of the sediments.

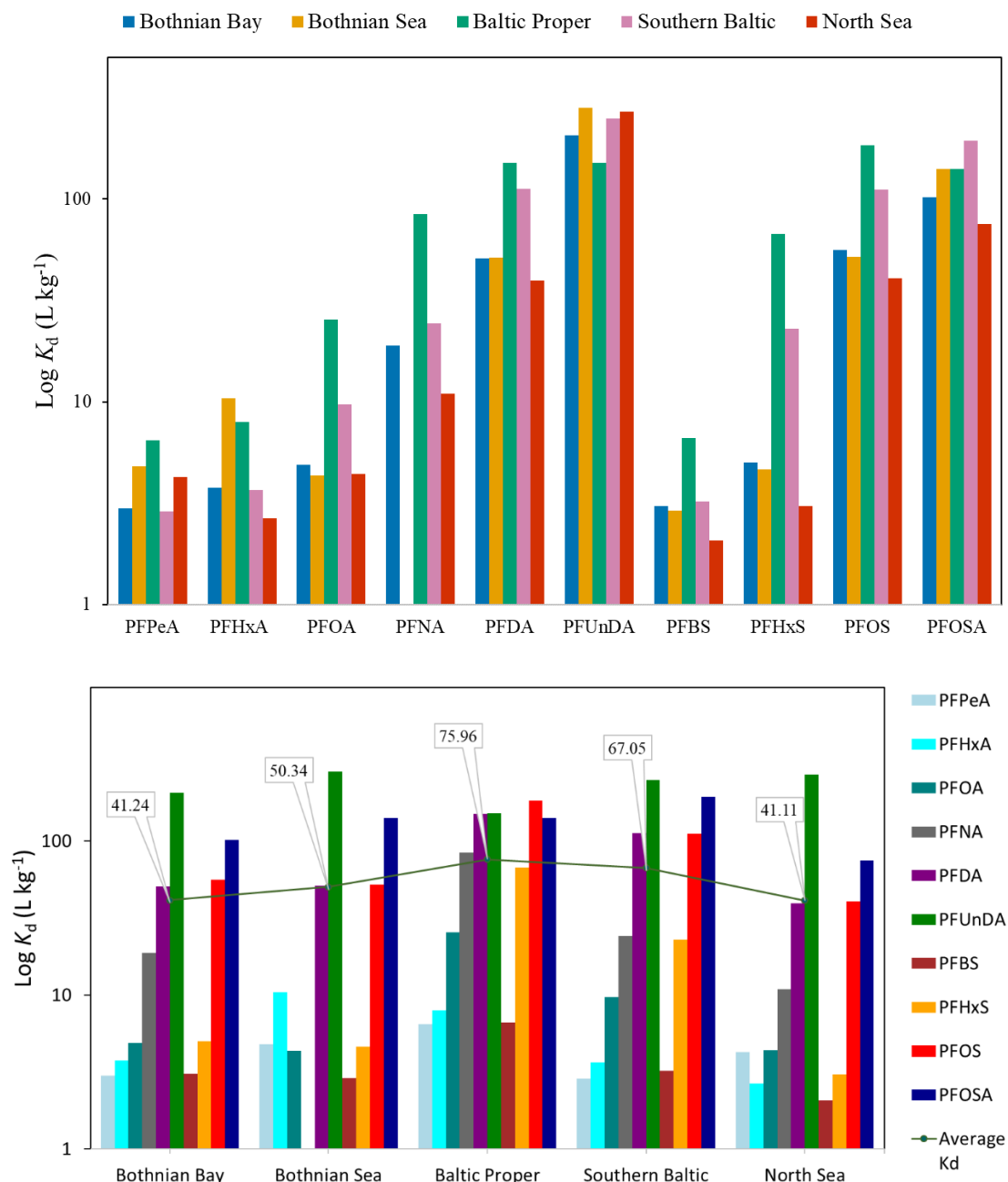


Figure 4. Sediment – pore-water partitioning coefficients (K_d values) for the different Swedish offshore sediment regions. Top: grouped for each individual PFAS. Bottom: aggregated by region with callouts indicating average K_d values per region.

The highest log K_d values were noticed for PFUnDA at 150 L kg^{-1} , followed by PFOSA at 140 L kg^{-1} . These K_d values are exceptionally high for natural sediments. Mussabek et al. (2020) investigated naturally contaminated sediments in Lake Sänksjön in Ronneby, south Sweden, revealing K_d values ranging from $0.6 - 2.3 \text{ L kg}^{-1}$ for perfluoroalkyl carboxylates (PFCAs) and $0.9 - 5.6 \text{ L kg}^{-1}$ for perfluoroalkane sulfonates (PFSAs). Their findings highlighted a significant correlation between mineral content (specifically Fe, Pb, Rb, As) and sorption affinity and low influence of the organic carbon content. Notably, the study underscored that higher mineral content has a greater impact on PFAS sorption to sediments compared to organic matter content. However, it should be noted that lake sediments typically

have a higher content of inorganic compounds, like iron and arsenic compared to offshore sediments, due to more reducing conditions and higher sedimentation rates, leading to the release of inorganic compounds from mineral phases (Barrett et al., 2019). Another study by Oliver et al. (2020), who used spiked samples with PFOS, PFOA and PFHxS at $10 \mu\text{g L}^{-1}$, estimated K_d values of up to 30.4 L kg^{-1} for PFOS. The high K_d values in this study are likely due to an overestimation of the PFAS concentrations in the solid phase, an issue that was also evident when high concentrations were detected in the sediment screening (Figure 3). Nonetheless, patterns can be observed among the different regions and PFASs.

A variety of mechanisms can influence PFAS sorption capacities in environmental systems (Higgins and Luthy, 2006). These mechanisms may depend on the intrinsic properties of PFASs, such as their chain length and functional groups, affecting physicochemical properties like hydrophobicity and electrostatic interactions. Additionally, environmental factors, including pH, temperature, salinity, organic and inorganic compounds can be crucial. Evidently, the carbon content is one of the most determinant factors of the ability of a medium to retain PFASs (Kookana et al., 2022). Furthermore, the specific type of organic carbon (e.g., fulvic and humic acids) can significantly influence PFAS sorption dynamics (Campos Pereira et al., 2018).

The highest K_d values were observed in samples from the Baltic Proper and the Southern Baltic, coinciding with the carbon content of the sediments in these regions (Table 1). To examine the influence of organic carbon on the observed partitioning behavior, the partitioning coefficients were normalized for the organic carbon content and K_{OC} values were calculated (Figure 5). Overall, K_{OC} values exhibited lower variability compared to K_d values, indicating the significance of organic carbon in influencing PFAS sorption. PFPeA, PFHxA, PFBS, PFUnDA, and PFOSA exhibited higher K_{OC} values in the Bothnian and North Seas, with PFUnDA demonstrating a particularly pronounced difference at an order of magnitude; however, this pattern was not consistent across all PFAS compounds.

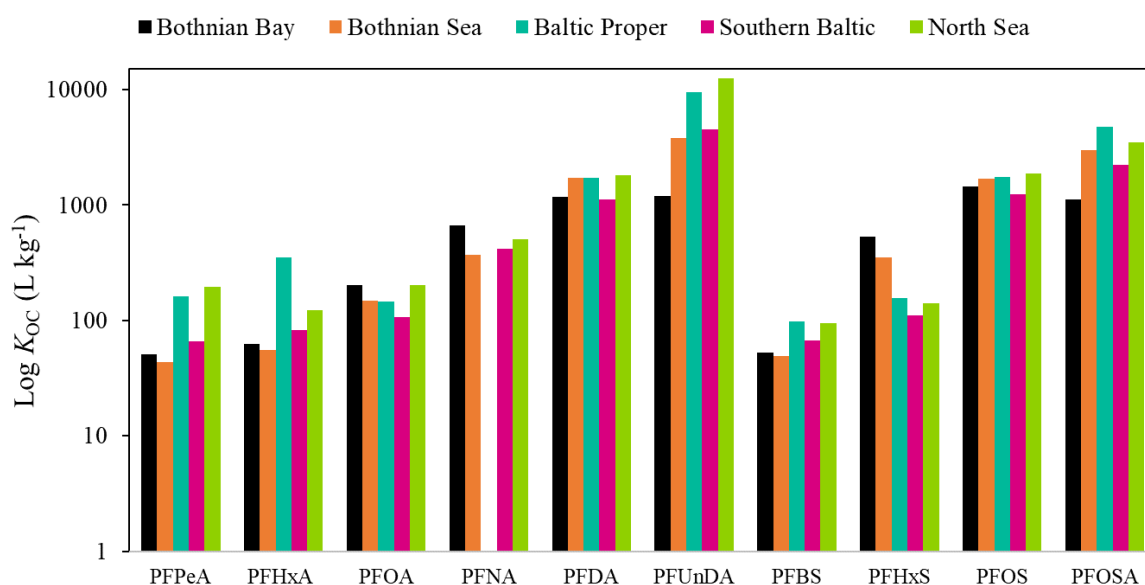


Figure 5. Logarithmic values of the organic carbon partitioning coefficients for different PFAS at various regions of the Baltic Sea and the North Sea.

4 IMPLICATIONS AND OUTLOOK

This study investigated the occurrence and sorption behavior of PFASs across Swedish offshore sediments, aiming to explain regional disparities in PFAS levels. Significantly elevated PFAS concentrations were identified in sediments of the northern Swedish marine region, particularly in the Bothnian Gulf. The similarity of the PFAS composition at the Bothnian Sea and Bothnian Bay regions suggest the influence of similar sources of PFASs. Minor differences in contaminant levels can likely be attributed to variations in SAR. Further analysis of core samples is expected to provide deeper insights into the historical deposition patterns and source contributions over time.

Contrary to initial hypotheses, no consistent evidence of heightened sorption affinity in northern sediments was observed. The absence of elevated sorption capacity in offshore sediments of the Bothnian Gulf suggests the presence of distinct PFAS sources in this region. Consequently, the unusually high PFAS concentrations in the northern Baltic Sea cannot be solely attributed to sediment properties, which necessitates the consideration of additional sources.

The estimated K_d values in this study did not reveal any strong trends, and a rather increased sorption affinity at the Southern Baltic and Baltic Proper, which contrasts with initial expectations. This increased sorption affinity was attributed to higher organic carbon content, as the Southern Baltic and Baltic Proper had higher TOC concentrations. The K_{OC} values exhibited lower variability and did not reveal significant differences or trends among the different regions.

It is worth noting that the estimated partitioning coefficients were exceptionally high, indicating some analytical uncertainty in the estimations. It is also essential to acknowledge that both K_d and K_{OC} values provide simplified representations of the sorption process. While the influence of organic matter on PFAS sorption was confirmed, other environmental factors are also likely to affect the fate and transport of PFASs in offshore sediments, including the presence of oxides, salinity, pH, and more. While additional sorption studies, such as adsorption isotherms, could enhance understanding, their necessity is debatable given the absence of significant patterns in this study and the apparent dominance of organic carbon content in driving sorption processes.

Further investigations are warranted to uncover the potential PFAS sources contributing to elevated levels in the Bothnian Gulf. Source identification poses challenges, as it necessitates comprehensive monitoring efforts. Exploring temporal trends, particularly related to snowmelt or seasonal variations, could provide valuable insights. Tributary PFAS loading also needs to be further characterized. Additionally, it is noteworthy that contamination of offshore sediments can be a transboundary issue, influenced by multiple regions of the Baltic and even long-range transport processes. For instance, three major rivers of Finland have been reported to contribute high levels of PFAS into the Bothnian Bay: River Oulujoki: 12 kg y⁻¹, River Kemijoki: 15 kg y⁻¹ and River Tornionjoki: 3.4 kg y⁻¹ (Junttila et al., 2019). Therefore, multiple point and diffuse sources should be considered across a widened geographic area.

In summary, ongoing research efforts should prioritize the investigation of PFAS inputs into the Baltic, with a focus on source identification, especially in the regions of the Bothnian

Gulf. This will provide crucial insights to enhance mitigation strategies and inform environmental management practices effectively.

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