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Analysis of PFAS in offshore sediment

Analys av PFAS i utsjösediment

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Sammanfattning

Per- och polyfluoralkylsubstanser (PFAS) utgör en grupp syntetiska kemikalier som orsakat miljöproblem på grund av deras persistens, bioackumulerande egenskaper och potentiella negativa hälsoeffekter. PFAS har identifierats i miljön globalt, och kontaminerat sediment kan fungera som en reservoar och utgöra ekologiska risker för akvatiska ekosystem. Studier av PFAS i ytsediment från Bottenhavet och Bottenviken har visat på betydligt högre PFAS-nivåer jämfört med havssediment från andra regioner. Dessutom visar analys av oxiderbara prekursorer i sediment från Västkusten förhöjda nivåer av prekursorer som kan brytas ned till skadliga PFAS. Denna studie syftar till att öka förståelsen för dessa mönster genom att analysera PFAS inklusive prekursorer i utvalda sedimentprover från olika platser, fokuserat på 56 PFAS, oxidativ omvandling av prekursorer och extraherbart organiskt fluor. Sedimentkärnor från fyra platser, inklusive ytskikt och två djupare kärnsediment, som representerar olika regioner och tidsperioder valdes ut för analys.

Perfluoroktansulfonsyra (PFOS) och långkedjiga perfluoralkylkarboxylsyror (PFCA) detekterades med den högsta frekvensen. Prekursorer detekterades i alla prover utom ett. En minskande trend av PFAS koncentrationen från ytsediment till djupare kärnprover observerades för proverna från östkusten, medan prover från Nordsjön visade konsekventa halter. PFAS-nivån i ytskiktet var i genomsnitt 5,5 gånger högre i norr, med summan PFAS 17,5 och 18,6 ng/g torrsubstans (t.s.). i Bottenviken respektive Bottenhavet jämfört med 3,3 ng/g t.s. i södra Östersjön. Nordsjön var mindre förorenad med en summa PFAS runt 1 ng/g t.s..

PFOS-prekursorer, inklusive FOSAA, EtFOSAA och diSAmPAP, hittades uteslutande i prover från Bottenviken och Bottenhavet. Flera fluortelomerprekursorer detekterades, inklusive 6:2 FTSA, 6:2 mono-PAP och långkedjiga diPAPs (8:2, 8:2/10:2, 10:2 och 10:2/12:2). Förändringen i koncentrationer efter oxidation, uttryckt som ΔPFCA, ökade med djupet vilket indikerar en högre historisk frisättning av prekursorer. Den största ΔPFCA observerades i södra Östersjön (93-716%) och Nordsjön (392-560%), jämfört med Bottenviken (38-224%) och Bottenhavet (5-160%). Den mest signifikanta ökningen efter oxidation observerades för kortkedjiga PFCA (C4-C7), vilket bidrog med 56-80% till ΔPFCA.

Sammantaget bekräftar studien högre PFAS-koncentrationer i Bottenviken och Bottenhavet jämfört med Östersjön och Nordsjön. Kortkedjiga PFCAs uppvisade betydande ökningar efter oxidation, vilket tyder på närvaron av kortkedjiga prekursorer i alla prover. Dessa prekursorer ingick inte i de 56 PFAS som studien inkluderade.

Summary

Per- and polyfluoroalkyl substances (PFAS) constitute a group of synthetic chemicals that has raised environmental concerns due to their persistence, bioaccumulative nature, and potential adverse health effects. PFAS have been identified in the environment globally, and sediments can act as reservoirs, posing ecological risks to aquatic ecosystems. Studies of PFAS in surface sediments from the Bothnian Sea and the Gulf of Bothnia have revealed significantly higher PFAS levels compared to offshore sediments from other regions. Moreover, analysis of oxidizable precursors in sediments from the West Coast of Sweden indicates elevated levels of precursors that can degrade into harmful PFAS. This study aims to increase the understanding of these patterns by analyzing precursors in selected sediment samples from various locations, targeting 56 PFAS compounds, oxidative conversion of precursors, and extractable organofluorine analysis. Sediment core samples from four locations, including surface layer and two deeper core sediments, were selected for analysis, representing different regions and time periods.

Perfluorooctane sulfonic acid (PFOS) and long-chain perfluoroalkyl carboxylic acids (PFCAs) were detected with highest frequency. Precursor compounds were detected in all samples except from one. A decreasing trend in concentration of PFAS from surface sediment to deeper core samples was observed for the East Coast samples, while the North Sea location showed consistent levels across the sediment core. The PFAS level in the surface layer was on average 5.5 times higher in the north, with sum PFAS of 17.5 and 18.6 ng/g d.w. in Bothnian Bay and Bothnian Sea, respectively, compared to 3.3 ng/g d.w. in Baltic South. Northern Sea was less contaminated with a sum of PFAS around 1 ng/g d.w..

PFOS precursors, including FOSAA, EtFOSAA, and diSAmPAP, were exclusively found in Bothnian Bay and Bothnian Sea samples. Several fluorotelomer precursors were detected, including 6:2 FTSA, 6:2 mono-PAP, and long chain diPAPs (8:2, 8:2/10:2, 10:2 and 10:2/12:2). The change in concentrations post-oxidation, expressed as ΔPFCA, increased with depth which indicates a higher historical release of precursors. The largest ΔPFCA was seen for Southern Baltic (93-716%) and North Sea (392-560%), compared to Bothnian Bay (38-224%) and Bothnian Sea (5-160%). The most significant increase after oxidation was observed for short-chain PFCAs (C4-C7), contributing 56-80% to ΔPFCA.

Overall, the study confirms higher PFAS concentrations in Bothnian Bay and Bothnian Sea compared to Baltic Proper South and Northern Sea. Short-chain PFCAs exhibited significant increases after oxidation, suggesting the presence of unmonitored shorter chain precursors in all samples.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of synthetic chemicals that have been widely used in various industrial and consumer products due to their unique properties, such as water and oil repellency, heat resistance, and chemical stability (KemI, 2015). PFAS have become a significant environmental concern due to their persistence, bioaccumulative nature, and potential adverse health effects (Silva et al., 2021; Fenton et al., 2021).

PFAS has been detected in sediments worldwide, including the North and Baltic Sea (Joerss et al. 2019; Filipovic et al. 2013). These chemicals can enter sediments through various pathways, including direct discharge from industrial facilities, runoff from contaminated sites, atmospheric deposition, and wastewater effluent (Dasu et al. 2022). PFAS can persist in sediment for extended periods due to their chemical stability and resistance to degradation processes. Once deposited in sediment, PFAS can undergo sorption and desorption processes, leading to long-term environmental persistence (Mussabek et al. 2019). Sediment serves as a sink for PFAS, leading to potential ecological risks within aquatic ecosystems (Ankley et al. 2021).

Investigations of PFAS in surface sediments from the Bothnian Sea and the Gulf of Bothnia have revealed that PFAS levels in sediments in these sea areas are, on average, eight times higher compared to offshore sediments from the Baltic Sea proper and from the West Coast (Josefsson 2022). On the contrary, analysis of oxidizable precursors for samples from the West Coast indicates that sediments from this area contain precursors that can break down into PFHxA, PFOA, and PFDA.

The aim of this study was to further investigate the pattern of PFAS including oxidizable precursors in selected sediment samples from the Bothnian Sea, Bothnian Bay, Baltic Sea, and from the West Coast.

2. Material and methods

2.1 Target analytes and chemicals

A total of 54 non-polymer PFAS were targeted in the study, divided into the subgroups perfluoroalkyl acids (PFCAs, n=15), perfluoroalkyl sulfonic acids (PFSAs, n=11), perfluoroalkylsulfonamides (FASA, n=7), fluorooctanesulfonamidoacetic acids (FOSAA, n=3), polyfluorotelomer sulfonic acids (FTSA, n=6), polyfluoroalkyl phosphate diesters (diPAP, n=8), perfluoroether acids/sulfonic acids (PFECA/PFESA, n=4), N-ethyl perfluorooctane sulfonamido ethyl phosphate diester (diSAmPAP), and perfluoroethylcyclohexane sulfonic acid (PFECHS). For full list of target analytes, see Appendix A. In addition, EOF was targeted. The analysis after oxidation also included ultra-short chain PFAS, TFA, PFPrA, TFMS, PFEtS, PFPrS). Native and labeled standards were from Wellington Laboratories (Guelph, Canada).

2.2 Samples

Sediment core samples taken during the environmental monitoring program 2020-2021 were selected and retrieved from frozen archives, for details see Josefsson (2022). Four locations were jointly selected for this study by the Swedish Environmental Protection Agency and Geological Survey of Sweden (SGU), representing Bothnian Bay, Bothnian Sea, Baltic Sea and West Coast (Table 1). Three samples from sediment cores representing surface sediments (recent years), approximately year 2000, and dated back to mid-1980s and the early 1990s, were selected from each of the four sites. Sediment was stored at – 20 °C and was freeze-dried prior to analysis.

Table 1. Information on the sediment samples included in the study

Station	Sea area	Sea basin	Coordinates *		Depth (m)
SE-17	Bothnian Bay	N Bothnian Bay	N 7255864	E 892219	87
SE-4	Bothnian Sea	Åland Deep	N 6679643	E 724318	230
SE-12	Baltic Proper South	Arkona Basin	N 6095142	E 433358	47
SE-16	North Sea	E Skagerrak	N 6504578	E 236241	197

* SWEREF 99TM

2.3 Sample preparation

Each sediment sample was extracted in duplicate, with one replicate analysed for target analysis, while the other replicate was extracted for EOF analysis, oxidative conversion (also referred to as the Total Oxidizable Precursor Assay, TOPA), and mass balance analysis. Samples for target analysis were fortified with internal standards prior to extraction. In brief, 0.5 g of freeze-dried sediment was digested with sodium hydroxide in methanol, followed extraction using methanol and sonication. Clean-up for target analysis was performed using graphitized carbon (ENVI-carb, 100 mg, 1 mL, Supelco), while additional clean-up was made for EOF, TOPA, and mass balance analysis using Oasis WAX cartridges (60 mg, 3 cc, 30 µm; Water Corporation, Milford, USA. The final sample extracts were concentrated to 500 µL under nitrogen.

The TOPA was based on the method developed by Houtz and Sedlak in 2012 (Houtz and Sedlak 2012) but modified by increasing the dose of oxidant and base to strive for complete oxidation. In brief, sample extracts were evaporated to near dryness in a 50 mL PP tube and was amended with 45 mL of 120 mM potassium persulphate (Sigma Aldrich, Germany) in Milli-Q and 1.35 mL 10N NaOH solution. Samples were placed at 85 °C water bath for 6 h. After the reaction, solutions were cooled to room temperature and acidified to pH 2 with formic acid (Thermo Fischer scientific, USA). Post-oxidation extraction followed a similar procedure described above, with Oasis WAX SPE cartridges of larger sorbent material and particle (500 mg, 60 µm) to reduce the elution of salt. Only Milli-Q and ammonium acetate buffer were used for washing steps, to improve recovery of trifluoroacetate (TFA).

2.4 Instrumental analysis and quality control

2.4.1 Target PFAS

Targeted analysis was carried out on an Acquity UPLC coupled to a Xevo TQ-S triple quadrupole mass spectrometer, equipped with a BEH C18 column (1.7 µm, 100 × 2.1 mm). All instrumentations and columns were from Waters Corporation, Milford, Massachusetts, USA. The mobile phase consisted of 2 mM ammonium acetate in water and methanol. For the PAP analysis 5 mM 1-methylpiperidine was used as an additional additive. An isolator column was inserted after the solvent mixer before the injector to separate any potential contamination from the UPLC system from the injected sample. The system was operated in negative electrospray ionization (ESI-) mode. The source and desolvation temperatures were set at 150 °C and 400 °C, respectively. The desolvation and cone gas flows (nitrogen) were set at 800 L/h and 150 L/h, respectively. The capillary voltage was set at 0.7 kV. Some modifications were done for the PAP analysis; desolvation temperatures and capillary voltage were set at 200 °C and 2.9 kV, respectively. Quantification was performed using labeled internal standards and minimum a four-point linear calibration curve, that ranged from 0.02 to 50 ng/mL, depending on the analyte. Each batch of samples contained one or several procedural blanks and a fortified sample. The procedural blank was used for calculating limit of detection, together with the lowest concentration standard in the calibration curve. Recovery of all analytes were monitored in the fortified sample, together with the internal standard recoveries.

2.4.2 EOF

A combustion ion chromatography (CIC) system with a combustion module from Analytik Jena, (Germany), and an ion chromatograph from Metrohm (Switzerland) was used to quantify extractable organofluorine (EOF). The anions were separated with an ion exchange column (Metrosep A Supp 5–150/4), carbonate buffer (64 mmol/L sodium carbonate and 20 mmol/L sodium bicarbonate) as eluent and isocratic elution. The autosampler injected 100 µL of the extract on a quartz boat. The boat was inserted into the oven (1000–1050 °C) under a flow of oxygen (300 mL/min), argon (100 mL/min), and argon mixed with water vapor (100 mL/min) under hydropyrolytic conditions monitored by a flame sensor followed by 2 minutes of post-combustion time with the flow of oxygen (400 mL) only. The hydrogen fluoride (HF) formed during combustion was absorbed in ultrapure water (in the absorber module). The F^{-} concentration was measured via conductivity. A five-point calibration curve of 50-1000 µg/L using PFOA as standard was used for quantification. Empty boat combustions were made before and after each sample and the average background was subtracted from the sample signal. Procedural blanks were run for monitoring background contamination.

3. Results and discussion

3.1 Target analysis

The highest sum concentration of detected PFAS was found in surface sediments of Bothnian Sea (18.6 ng/g d.w.) and Bothnian Bay (17.6 ng/g d.w.). A decreasing trend in concentration of PFAS from surface sediment to deeper core samples was observed for the East Coast samples, while the North Sea location showed consistent levels across the sediment core. The concentration difference between surface and core sediments from the East Coast may indicate increasing PFAS contamination from recent years compared to approximately year 2000, and mid-1980s and the early 1990s.

PFOS was detected in most samples (detection frequency (df) 92%), followed by PFOA, PFNA, and PFUnDA (df = 83%). The percentage contribution of PFOS within the sediment core was similar in Bothnian Bay and North Sea samples, while decreasing with depth for Bothnian Sea and Southern Baltic. PFOS was the predominating compound in most of the East Coast samples, except for the deepest layer of the Bothnian Sea samples, which the composition was mainly contributed by PFAS precursors. Only PFCA with chain length greater than 6 were detected, with long chain PFCAs predominating the profile.

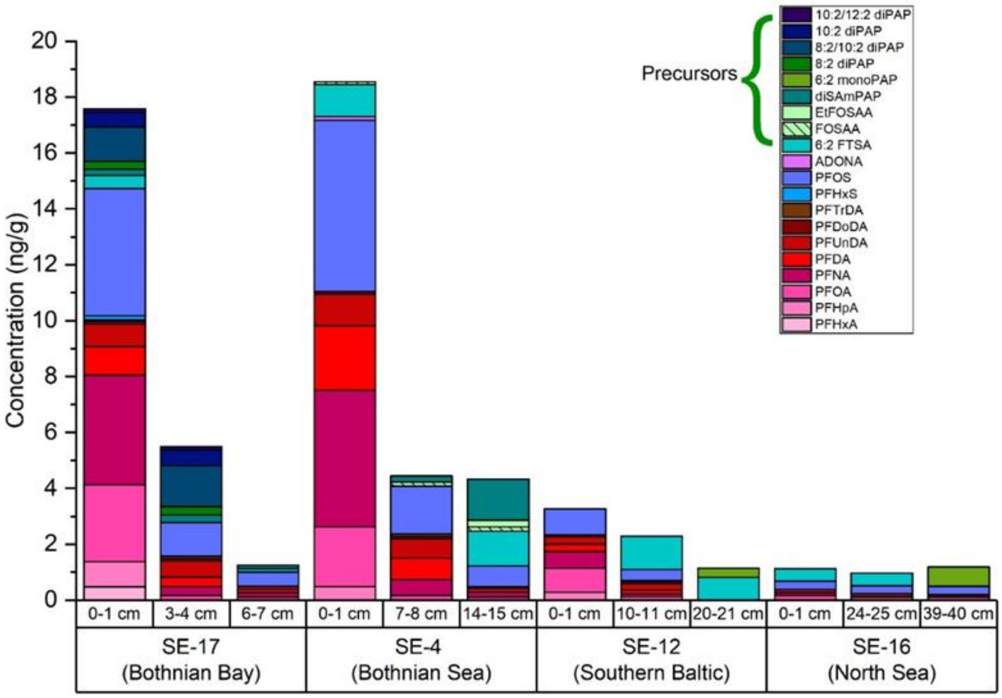

Figure 1. Concentrations (ng/g d.w.) of PFAS detected in sediment core samples

PFAS precursors were detected in all samples except of the surface layer in Southern Baltic (Figure 2). FOSAA, EtFOSAA, and diSAmPAP, which are precursors manufactured by electrochemical fluorination (ECF), were exclusively detected in the Bothnian Bay and Bothnian Sea. FOSAA and Et-FOSAA were only found in the core from Bothnian Sea. FOSAA was detected in all three layers while Et-FOSAA was only detected in the deepest layer (14-15 cm). EtFOSAA and FOSAA have been observed to be transformation products of diSAmPAP in marine sediment and soil (Benskin et al. 2013; Bugsel and Zwiener 2020). Highest concentration of diSAmPAP was found in the 14-15 cm layer at Bothnian Sea (1.5 ng/g d.w.).

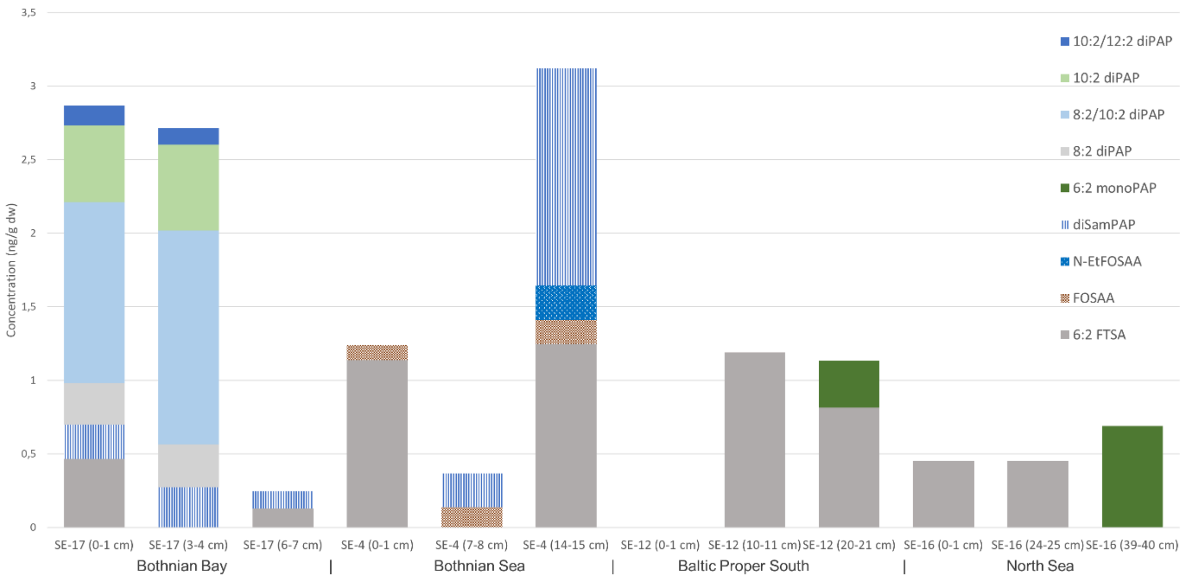

Figure 2. Concentrations (ng/g d.w.) of precursor compounds detected in sediment core samples. Bars with patterns represent PFOS precursors.

Several fluorotelomer precursors were detected, including 6:2 FTSA, 6:2 mono-PAP, and long chain diPAPs (8:2, 8:2/10:2, 10:2 and 10:2/12:2). DiPAPs were detected at the surface and middle (3-4 cm) layer of the sediment core from Bothnian Bay, while <MDL in the other three sampling locations, suggesting specific sources in the Bothnian Bay region. 6:2 FTSA was detected at all sampling locations, but not in all samples (df = 67%). However, no trend for the concentration nor distribution could be observed. It should be noted that a discrepancy between the target analysis and the mass balance analysis for comparison with EOF was observed. No 6:2 FTSA was observed in the mass balance analysis, in which an extra clean-up step was introduced. 6:2 monoPAP was detected at the bottom core samples of Southern Baltic and North Sea at a low concentration close to the MDL.

The precursor profile and the trend observed in sediment core could have connection to paper industry as one point source that uses diSAmPAP, FOSAAs and diPAPs. It has previously been observed that sediments collected downstream of paper factories could have high concentrations of PFAS precursors, especially diSAmPAP and FOSAAs (Kärrman et al. 2022; Langberg et al. 2021).

3.2 Oxidative conversion

After oxidative conversion, a change in PFCA (ΔPFCA) concentrations was detected in all sediment samples, with a percentage increase of total molar concentration (nM) between 38% and 716% (Figure 3), and ΔPFCA concentration increase of 1.7 ng/g d.w. to 4.5 ng/g d.w.. Labeled FOSA was added to the samples as an indicator for oxidation efficiency. Despite a 94% conversion observed in the positive control, the average recoveries in the oxidation batches were 58% ± 10%, which was lower than expected. Therefore, the possibility of underestimating ΔPFCA cannot be ruled out. Concentration increase of each chain length was determined by the ratio between pre- and post-oxidation PFCA concentrations, in which only a ratio greater than 1.2 of each chain length was included as an increase in concentration. No significant loss of PFCAs during oxidative conversion and the subsequent extraction was also concluded by a ratio of no less than 0.8.

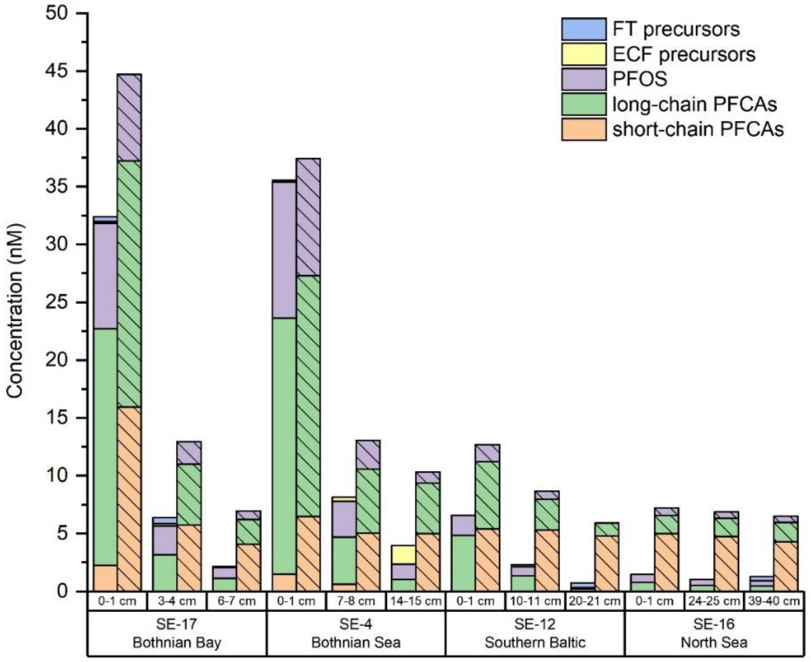

Figure 2 The total concentrations (nM) of PFAS pre- (non-striped) and post-TOPA (striped) in sediments core samples from offshore Baltic Sea.

The post-oxidation concentrations displayed the same trend as pre-oxidation target analysis, with a decrease in concentration from surface to deeper core sediment samples from Bothnian Bay, Bothnian Sea, and Southern Baltic. On the contrary, the ΔPFCA increased with depth (Appendix C) which indicates a higher historical release of precursors. The largest ΔPFCA was seen for

Southern Baltic (93-716%) and North Sea (392-560%), compared to Bothnian Bay (38-224%) and Bothnian Sea (5-160%). The post-oxidation concentrations for the three core samples from North Sea were similar with no increasing or decreasing trend which is aligned with the pre-oxidation concentrations.

Overall, the homologue profile of oxidation products was similar for all samples, and consisted of mainly C4 - C8 PFCAs, with the most significant increase after oxidation observed for short-chain PFCAs (C4-C7), contributing 56% to 80% to ΔPFCA. (Figure 4) The results suggest the presence of unmonitored shorter chain precursors in all of the samples.

No ultra-short-chain PFCAs (C1-C3) were detected post-oxidation. An increase in ΔPFBA was observed only at the surface sediments of Bothnian Bay and contributed to 44% of the total ΔPFCA in the sample. Contribution of PFHpA was similar across all samples (6% - 10%). Linear PFOA contributed 11% – 36% of the ΔPFCA, with the highest observed increase at the bottom core of Bothnian Sea, where highest concentrations of C8 ECF precursors was detected in target analysis. Branched isomers of PFOA were also detected in all samples and contributed between 2 - 8% of the total concentration. The branched isomers in Bothnian Bay and Bothnian Sea can partially be explained from the isomers of the detected ECF precursors diSAmPAP and EtFOSAA, while for Southern Baltic and North Sea samples this may be originated from other unknown ECF manufactured precursors. An increase in PFNA was observed at the 3-4 cm core of Bothnian Bay, and an increase in PFDA was observed in the 10-11 cm core of Southern Baltic. At Bothnian Bay, this may be explained from the longer chain diPAPs detected in target analysis, while the source of PFDA at Southern Baltic samples are not known from detected precursors.

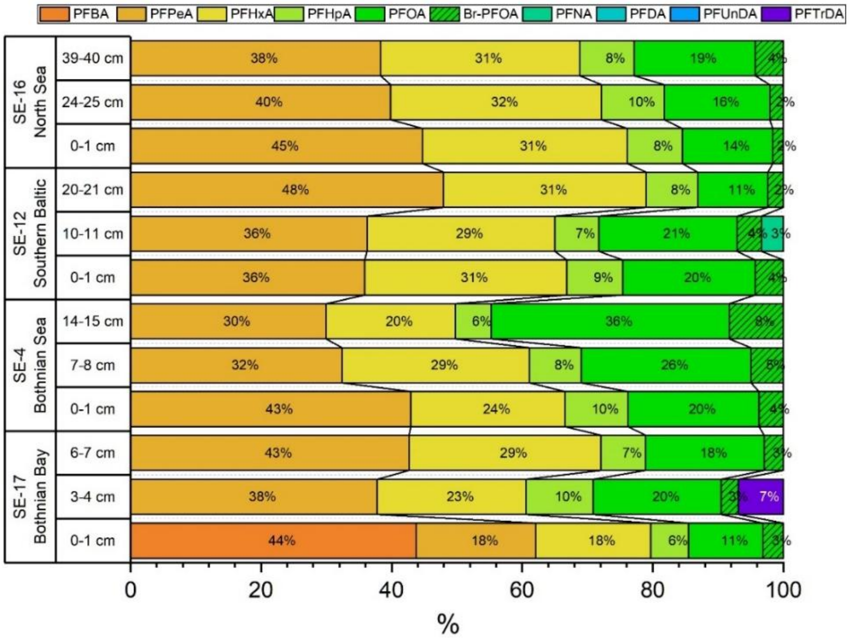

Figure 3 Oxidation profiles of linear and branched PFCAs post-oxidation in sediment core samples.

3.3 Extractable organofluorine analysis

Only one out of the twelve samples, the deepest core sample from Southern Baltic, had an EOF level (220 ng/g F) above the reporting limit of 206 ng/g F. The higher level of EOF was unexpected at that location, as it showed a low concentration of PFAS from target analysis and a similar magnitude of ΔPFCA from oxidative conversion in comparison to other samples. Target analysis initially accounted for only 0.1% of the EOF. Although oxidative conversion was used in the hope of identifying a larger fraction of EOF, the post-oxidation ΔPFCA in EOF only contributed to an additional 0.5%. This results in a total identified EOF of less than 1%.

4. Conclusion

This study confirms previous reports on higher PFAS concentrations in Bothnian Bay and Bothnian Sea compared to Baltic Proper South. The PFAS level in the surface layer was on average 5.5 times higher in the north, with sum PFAS 17.5 and 18.6 ng/g d.w. in Bothnian Bay and Bothnian Sea, respectively, compared to 3.3 ng/g d.w. in Baltic South. However, the deeper sediment samples displayed a lower concentration and the PFAS levels were only 1-3.8 times higher in the northern samples compared to the Baltic South. The study also concludes that Northern Sea is less contaminated with a sum of PFAS around 1 ng/g d.w. for all depths. The homologue profile analysis show that the Bothnian Sea site could be influenced by point sources using diPAPs, FTSA, and PFOS-precursors diSAmPAP, FOSAA and Et-FOSAA. These PFAS were previously seen in sediments close to paper manufacturing (Kärrman et al. 2022; Langberg et al. 2021). However, there is a higher portion of unidentified precursors, relative the known PFAS concentration, in samples from the North Sea and Baltic South. The oxidation products points at primarily short chain precursors (C6) but long chain precursors are also present.

Acknowledgements

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Appendix A

List of included PFAS in the study

Perfluoroalkyl acid (PFCA)	*Trifluoroacetic acid (TFAA, also called trifluoroacetate (TFA))
	*Perfluoropropanoic acid (PFPrA)
	Perfluorobutanoic acid (PFBA)
	Perfluoropentanoic acid (PFPeA)
	Perfluorohexanoic acid (PFHxA)
	Perfluoroheptanoic acid (PFHpA)
	Perfluorooctanoic acid (PFOA)
	Perfluorononanoic acid (PFNA)
	Perfluorodecanoic acid (PFDA)
	Perfluoroundecanoic acid (PFUnDA)
	Perfluorododecanoic acid (PFDoDA)
	Perfluorotridecanoic acid (PFTrDA)
	Perfluorotetradecanoic acid (PFTeDA)
	Perfluoropentadecanoic acid (PFPeDA)
	Perfluorohexadecanoic acid (PFHxDA)
	Perfluoroheptadecanoic acid (PFHpDA)
	Perfluorooctadecanoic acid (PFOcDA)
Perfluoroalkyl sulfonic acid (PFSA)	*Trifluoromethyl sulfonic acid (TFMS)
	*Perfluoroethane sulfonic acid (PFEtS)
	*Perfluoropropane sulfonic acid (PFPrS)
	Perfluorobutane sulfonic acid (PFBS)
	Perfluoropentane sulfonic acid (PFPeS)
	Perfluorohexane sulfonic acid (PFHxS)
	Perfluorooctane sulfonic acid (PFOS)
	Perfluorononane sulfonic acid (PFNS)
	Perfluorodecane sulfonic acid (PFDS)
	Perfluoroundecane sulfonic acid (PFUnDS)
	Perfluorododecane sulfonic acid (PFDoDS)

	Perfluorotridecane sulfonic acid (PFTrDS)
	Perfluorotetradecane sulfonic acid (PFTeDS)
Fluoroalkylsulfonamide (FASA)	Perfluorobutane sulfonamide (FBSA)
	N-Methyl perfluorobutane sulfonamide (MeFBSA)
	Perfluorohexane sulfonamide (FHxSA)
	N-Methyl perfluorohexane sulfonamide (MeFHxSA)
	Perfluorooctane sulfonamide (FOSA)
	N-Methyl perfluorooctane sulfonamide (MeFOSA)
	N-Ethyl perfluorooctane sulfonamide (EtFOSA)
Fluorooctanesulfonamidoacetic acid (FOSAA)	Perfluorooctane sulfonamidoacetic acid (FOSAA)
	N-Methyl perfluorooctane sulfonamidoacetic acid (MeFOSAA)
	N-Ethyl perfluorooctane sulfonamidoacetic acid (EtFOSAA)
Fluorotelomer sulfonic acid (FTSA)	4:2 Fluorotelomer sulfonic acid (4:2 FTSA)
	6:2 Fluorotelomer sulfonic acid (6:2 FTSA)
	8:2 Fluorotelomer sulfonic acid (8:2 FTSA)
	10:2 Fluorotelomer sulfonic acid (10:2 FTSA)
	12:2 Fluorotelomer sulfonic acid (12:2 FTSA)
	14:2 Fluorotelomer sulfonic acid (14:2 FTSA)
Polyfluoroalkyl phosphate diesters (diPAP)	4:2 Fluorotelomer phosphate diester (4:2 diPAP)
	6:2 Fluorotelomer phosphate diester (6:2 diPAP)
	8:2 Fluorotelomer phosphate diester (8:2 diPAP)
	6:2/8:2 Fluorotelomer phosphate diester (6:2/8:2 diPAP)
	8:2/10:2 Fluorotelomer phosphate diester (8:2/10:2 diPAP)
	10:2 Fluorotelomer phosphate diester (10:2 diPAP)
	10:2/12:2 Fluorotelomer phosphate diester (10:2/12:2 diPAP)
	12:2 Fluorotelomer phosphate diester (12:2 diPAP)
N-ethyl perfluorooctane sulfonamido ethyl phosphate diester	diSAmPAP
Perfluoro ether acids / sulfonic acids (PFECA/PFESA)	Hexafluoropropylene oxide dimer acid (HFPO-DA)(GenX)
	3H-perfluoro-3-[(3-methoxy-propoxy)propanoic acid] (ADONA)

	6:2 chlorinated polyfluorinated ether sulfonate (6:2 Cl-PFESA)
	8:2 chlorinated polyfluorinated ether sulfonate (8:2 Cl-PFESA)
Perfluoroethylcyclohexane sulfonic acid	PFECHS

* Only determined after oxidation

Appendix B

Concentrations (ng/g d.w.) of PFAS and EOF in sediment samples

	Bothnian Bay			**Bothnian Sea**			**Baltic Proper South**			**North Sea**		
Sample name	**SE-17**	**SE-17**	**SE-17**	**SE-4**	**SE-4**	**SE-4**	**SE-12**	**SE-12**	**SE-12**	**SE-16**	**SE-16**	**SE-16**
Sample depth (cm)	**0-1**	**3-4**	**6-7**	**0-1**	**7-8**	**14-15**	**0-1**	**10-11**	**20-21**	**0-1**	**24-25**	**39-40**
EOF	<206	<206	<206	<206	<206	<206	<206	<206	220	<206	<206	<206
TFA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PFPrA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PFBA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PFPeA	<0.16	<0.16	<0.16	<0.16	<0.16	<0.16	<0.16	<0.16	<0.16	<0.16	<0.16	<0.16
PFHxA	0.47	<0.21	<0.21	<0.21	<0.21	<0.21	<0.21	<0.21	<0.21	<0.21	<0.21	<0.21
PFHpA	0.90	<0.10	<0.10	0.48	<0.10	<0.10	0.28	<0.10	<0.10	<0.10	<0.10	<0.10
PFOA	2.75	0.17	0.11	2.14	0.17	0.11	0.86	0.12	<0.10	0.17	0.10	0.10
PFNA	3.93	0.31	0.16	4.89	0.56	0.18	0.61	0.10	<0.03	0.10	0.04	0.03
PFDA	1.01	0.34	0.11	2.30	0.77	0.11	0.26	0.13	<0.10	<0.10	<0.10	<0.10
PFUnDA	0.81	0.57	0.11	1.13	0.69	0.07	0.26	0.23	<0.02	0.10	0.10	0.07
PFDoDA	0.10	0.08	<0.05	0.09	0.08	<0.05	0.07	0.07	<0.05	<0.05	<0.05	<0.05
PFTrDA	0.05	0.09	<0.02	<0.02	0.10	<0.02	<0.02	0.05	<0.02	<0.02	<0.02	<0.02
PFTDA	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04	<0.04
TFMS	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PFEtS	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PFPrS	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
PFBS	<0.09	<0.09	<0.09	<0.09	<0.09	<0.09	<0.09	<0.09	<0.09	<0.09	<0.09	<0.09
PFPeS	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
PFHxS	0.15	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15
PFHpS	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
L-PFOS	4.54	1.22	0.51	6.14	1.70	0.74	0.92	0.39	<0.05	0.30	0.28	0.30
Br-PFOS	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
PFDS	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
11Cl-PF3OUdS	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
9Cl-PF3ONS	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
PFECHS	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
HFPO-DA	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
ADONA	<0.10	<0.10	<0.10	0.14	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
4:2 FTSA	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
6:2 FTSA	0.47	<0.23	0.13	1.14	<0.23	1.25	<0.23	1.19	0.81	0.45	0.45	<0.23
8:2 FTSA	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
10:2 FTSA	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
FOSA	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
N-MeFOSA	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
N-EtFOSA	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
FOSAA	<0.10	<0.10	<0.10	0.10	0.14	0.16	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
N-MeFOSAA	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10

N-EtFOSAA	<0.10	<0.10	<0.10	<0.10	<0.10	0.23	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
diSamPAP	0.23	0.27	0.12	<0.10	0.23	1.48	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
6:2 monoPAP	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	0.32	<0.10	<0.10	0.69
8:2 monoPAP	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
6:2 diPAP	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
8:2 diPAP	0.28	0.29	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
6:2/8:2 diPAP	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
8:2/10:2 diPAP	1.23	1.45	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
10:2 diPAP	0.52	0.58	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
10:2/12:2 diPAP	0.13	0.11	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10

NA: Not analyzed

Appendix C

Results from oxidative conversion

Table C1. Increase (%) of PFCAs after oxidation

Sample	Change after oxidation
SE-17 (0-1 cm)	38%
SE-17 (3-4 cm)	103%
SE-17 (6-7 cm)	224%
SE-4 (0-1 cm)	5%
SE-4 (7-8 cm)	60%
SE-4 (14-15 cm)	160%
SE-12 (0-1 cm)	93%
SE-12 (10-11 cm)	277%
SE-12 (20-21 cm)	716%
SE-16 (0-1 cm)	392%
SE-16 (24-25 cm)	560%
SE-16 (39-40 cm)	399%

Only PFCAs with an increase ratio of 1.2 after oxidation were included.

Table C2. Ratio of concentrations post- oxidation / pre-oxidation

Sample	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFHxS	PFOS
SE-17 (0-1 cm)	1.95	4.22	2.69	1.27	1.18	0.89	0.93	0.86	0.99		0.74	0.82
SE-17 (3-4 cm)					3.43	1.00	1.06	0.78		7.00		0.78
SE-17 (6-7 cm)					2.99	0.97	0.88	0.83				0.77
SE-4 (0-1 cm)		6.62	2.78	1.48	1.21	0.84	0.82	0.74				0.86
SE-4 (7-8 cm)		5.10			4.19	0.86	0.96	0.93				0.80
SE-4 (14-15 cm)					8.06	0.96	0.88					0.74
SE-12 (0-1 cm)				1.69	1.51	0.85	0.91					0.84
SE-12 (10-11 cm)					3.84	1.73		0.83				0.88
SE-12 (20-21 cm)					2.96							0.00
SE-16 (0-1 cm)					2.42							0.98
SE-16 (24-25 cm)					3.71							1.04
SE-16 (39-40 cm)					3.72							1.14

Ratios between 0.8 and 1.2 are considered as no change after oxidation