Passive Sampler Derived Profiles and Mass Flows of Perfluorinated Alkyl Substances (PFASs) across the Fram Strait in the North Atlantic

Matthew Dunn, Simon Vojta, Thomas Soltwedel, Wilken-Jon von Appen, and Rainer Lohmann*

ACCESS | Metrics & More | Article Recommendations | SI Supporting Information

ABSTRACT: Per- and polyfluorinated alkyl substances (PFAS) are a family of pollutants of high concern due to their ubiquity and negative human health impacts. The long-range marine transport of PFAS was observed during year-long deployments of passive tube samplers in the Fram Strait across three depth transects. Time weighted average concentrations ranged from 2.4 to 360 pg L^{-1}, and 10 different PFAS were regularly observed. PFAS profiles and concentrations were generally similar to those previously characterized for polycyclic aromatic hydrocarbons (PAHs) at these sites. The detection of several anionic PFAS in "old" water demonstrated that they are not perfect water mass tracers but are also transported to depth via settling particles. Mass flows of PFAS through the Fram Strait in and out of the Arctic Ocean were basically similar (112 ± 82 Mg year^{-1} northward flow, 100 ± 54 Mg year^{-1} southward flow). For perfluorooctane sulfonamide (FOSA), export from the Arctic Ocean via the Fram Strait exceeded import by Atlantic Water, likely due to preferential transport and deposition in the Arctic Ocean. These observations suggest that PFAS in the Arctic are governed by the feedback loop previously described for polycyclic aromatic hydrocarbons (PAHs) in the region—with additional atmospheric transport delivering volatile PFAS to the Arctic, which then get exported via Arctic water masses.

KEYWORDS: PFAS, Fram Strait, PFAS Export, Long-Term Monitoring, Arctic, POPs, Passive Sampling
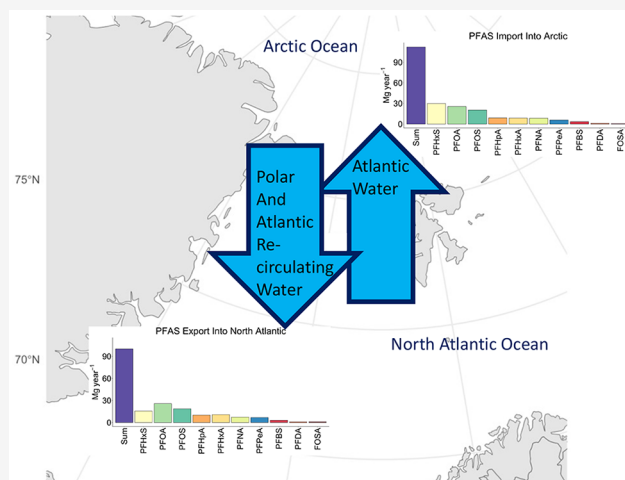
INTRODUCTION

One of the defining characteristics of persistent organic pollutants (POPs) is their ability to undergo long-range transport.[1] Per- and polyfluorinated alkyl substances, or PFAS, are a specific POP class of great concern due to their negative human health effects and global presence, even in remote regions including the Arctic.[2−8] Due to this long-range transport and ability to bioaccumulate, great effort has been put into characterizing the impact of PFAS in the Arctic.[5−7,9−14] While many previous studies have detected the most common PFAS, perfluorooctanoic acid (PFOA) and perfluorooctanoic sulfonic acid (PFOS), in water, sediment, and biota, there is new concern about their replacements, as industry has phased out PFOA and PFOS.[6,9,13] Past research has shown that increases in legacy PFAS deposition to the Arctic are tied to increases in production, something that may be replicated again with replacement compounds such as 6:2 fluorotelomer alcohol (FTOH), perfluorobutanesulfonate (PFBS), and other compounds.[5,8,9,12]

Understanding the fate and transport of PFAS to the Arctic is made further difficult by the region's remoteness. However, the Fram Strait, which has been previously characterized for other POPs, offers a unique opportunity to study PFAS dynamics through this "gateway of the Arctic".[15−17] Previous research has observed a feedback loop through the Fram Strait, where similar mass flows of POPs are imported from Atlantic Water masses in the eastern Fram Strait and exported from the Arctic Ocean to the North Atlantic in western parts of the strait.[16] However, there have been observations of depletions with depth for all POPs in the Fram Strait, potentially caused by degradation processes that govern some legacy POPs or might reflect changing emissions over time.[15,16] Due to inherent persistence of perfluorinated acids, they constitute an ideal tracer to further examine the complex interplay of the hydrological processes that intersect with chemistry and biology at depth.[12,15−18]

Ample evidence suggests that the dominant sources of PFAS in the remote northern hemisphere are via long-range transport

Figure 1. Sampling locations within the Fram Strait monitored from 2021−2022 using passive samplers.

from emissions to air and subsequent atmospheric deposition, coupled with oceanic long-range transport.[12,13,19−21] Prior studies have highlighted the presence and changing composition of PFAS contamination in the Atlantic and Arctic Ocean. For example, a modeling study by Zhang et al. (2017) suggested that the import of PFAS into the Arctic Ocean with Atlantic water was greatest around 2000 and has since decreased, with a few tons PFOS being imported annually, though another study suggests that transport to the Arctic for PFAS remains consistent or is increasing.[10,22] One way to address these dynamic conditions is to rely on detection tools, such as passive sampling, that can be used for long deployments in these remote regions to derive representative concentrations and profiles.[9,20,23,24] In particular, a previously validated tube passive sampler design containing a microporous membrane filled with sorbent was deployed.[25−27] Hence, the aims of our study were to (i) test passive samplers as a sampling tool across different sites and water depths in the Fram Strait; (ii) compare PFAS profiles and concentrations to those of legacy POPs from the same sites; and (iii) infer import and export of PFAS from the Arctic Ocean and assess whether the underlying sources are changing.

■ METHODS AND MATERIALS

Chemicals and Reagents. Liquid chromatography-grade methanol (LC-MeOH) and water (LC-water) were purchased from Fisher Scientific (New Hampshire, USA) along with ammonium hydroxide (NH_4OH), ammonium acetate ($C_2H_7NO_2$), ACS-grade ethanol (EtOH), and ACS-grade methanol (MeOH). Analytical standards were used to create native compound standards from the Wellington PFAC-30PAR mix plus an additional four analytical compounds (Table S1). Mass labeled surrogate solutions were comprised of the Wellington Laboratories (Canada) MPFAC-24ES plus an additional three of their mass labeled compounds (Table S1).

Long-Term Deployments and Sampling Details. Single tube passive samplers that have been previously validated were deployed at three different monitoring locations of the LTER (Long-Term Ecological Research) observatory HAUSGART-EN in the Fram Strait at four depths each (Figure 1).[25,28] Passive samplers were deployed for just over one year (392−413 days), and discrete water samples were collected alongside the recovery of these passive samples during *RV Polarstern* Cruise PS131 in the summer of 2022. Water properties including average salinity, directional velocity, and temperature with depth were reported from long-term monitoring mooring sites as described in previous research.[16] Sites HGIV and F4 were deeper transects on the Eastern end of the Fram Strait (>1000 m), while site EGC the sampled shallower (<900 m) waters of the East Greenland Current. For more site details, see Table S2.

PFAS Sample Analysis. Discrete water samples of approximately 1000 mL were shaken, weighed, and spiked with 4 ng of isotopically labeled internal standard followed by offline weak anion exchange (WAX) solid phase extraction (SPE), in accordance with previously published methods.[25,29] A matrix spike experiment was performed to assess the recovery of a native PFAS spike in tap water (Table S3). Passive samplers
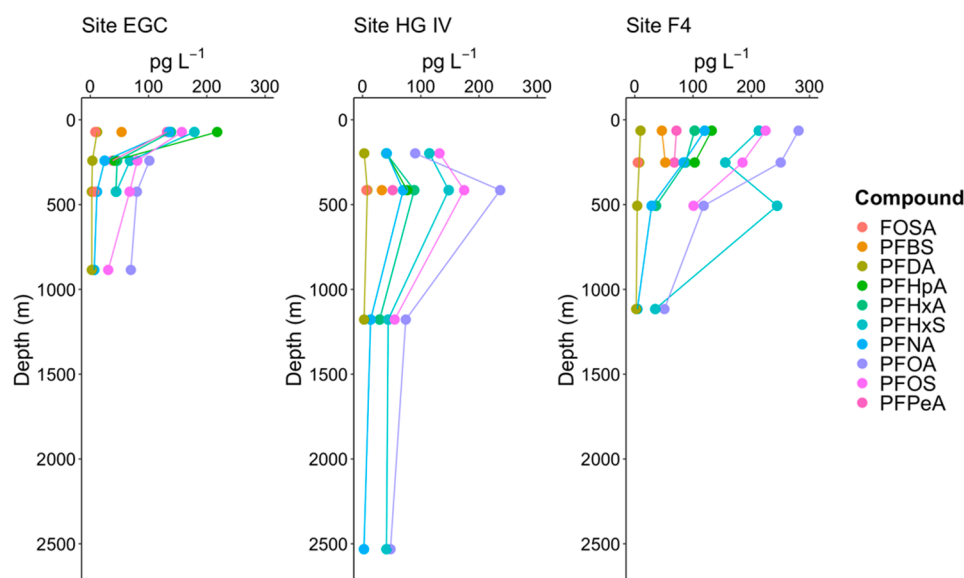
Figure 2. Time weighted average concentration profiles across depth in the Fram Strait.

were spiked with internal standards and extracted with MeOH following previously published methods with slight modifications, including the addition of a third 24-h extraction sequence, to yield a final extract volume of 18 mL that was then concentrated to 0.5 mL under a gentle stream of nitrogen at 40 °C. The passive sampler assembly has been reported earlier during the sampler's validation.[25] For the calculation of time weighted averages from passive sampler profiles, see SI section S2 and Tables S9−S10.

Quality Control and Instrumental Analysis. Method detection limits (MDLs) were calculated from laboratory and travel blanks collected for passive sampler (10−3300 pg sampler^{-1}) and solid phase (10−3900 pg L^{-1}) extraction procedures. The median and three times standard deviation of the blank concentrations were summed to determine MDLs (see Table S4). Method recoveries were evaluated using mass labeled surrogates purchased from Wellington, including their MPFAC24 mixture and three additional single compounds (Tables S5−S6). Recoveries of mass labeled standards ranging from 60 to 140% were accepted, with an exception made for perfluoroundecanoic acid (PFUnDA), which had an average recovery of 55% due to losses during the extraction procedure (for details, see SI). The instrumental analysis was performed by using a SCIEX Exion LC AC UHPLC system coupled to a SCIEX X500R quadrupole time-of-flight tandem mass spectrometer (QTOF MSMS). For additional details on instrumentation, see SI section S1.

Estimation of PFAS Mass Flows. Estimation of PFAS mass flows (Mg per year) across the Fram Strait was calculated as described previously, with water masses assigned based on location and depth to Polar Water (PW), Atlantic Water (AW), and Recirculating Atlantic Water (RAW).[16] This approach was employed at the same three sites and depth profiles characterized in Zhang et al. (2023).[16] Detected PFAS profiles were then integrated along the depth transect of the water mass to derive a mean concentration for each compound using the castr package in R from depths of 0 to 400 m for PW, and 0 to 840 m for RAW and AW, as described previously.[18] For further details on the calculation and method, see SI section S3.

■ **RESULTS**

Passive Sampler Uptake. A total of 10 PFAS were detected at least once in passive samplers above MDLs, at all three sites. These compounds displayed a general decrease in accumulated mass with depth, ranging from 0.01 to 1.9 ng sampler^{-1} for each individual compound (Figure S1). PFOA and PFOS were present in the highest amounts across all sites (0.14−1.9 ng sampler^{-1}), but all compounds were generally detected at the same frequency across depth and space (Table S7). No HFPO-DA ("Gen X") was detected in the passive samplers or water grabs from this study, despite previous detection.[12] Passive sampler percent of equilibrium (i.e., progress for PFAS to reach thermodynamic equilibrium between the passive sampler and the surrounding seawater) ranged from 1.0 to 54% across all 10 compounds, decreasing with chain length (Table S9). Other compounds aside from the 10 reported had no peaks detected or were below MDLs. When one of these 10 PFAS was found to be <MDLs, a replacement value of the MDL/$\sqrt{2}$ was used instead for calculating sum PFAS or integrated concentration with depth. As discussed in a previous study, it is unlikely that biofouling in the shallower sites impacted uptake.[25]

Seawater Grab Results. Most PFAS in the grab samples were below the MDL. The only compound detected above its MDL greater than 75% of the time was perfluorooctane sulfonamide (FOSA), for which concentrations ranged from 18 to 140 pg L^{-1} (Table S8). The next most prevalent compound was the long chain nonanoic carboxylate perfluorononanoic acid (PFNA), which was detected in 42% of samples above MDLs and ranged from 13−17 pg L^{-1}. Overall, calculated MDLs ranged from 10 to 3900 pg L^{-1}, displaying how low PFAS concentrations are in these remote waters (Table S4).

Time Weighted Average Concentration and Profiles. Derived time weighted average concentrations ranged from 2.4 to 360 pg L^{-1}, with an average propagated uncertainty of ±39% (for details see SI section S2). Detection was dominated by C5−C8 perfluorocarboxylic acids (PFCAs) and C4−C8 perfluorosulfonic acids (PFSAs) including PFOA and PFOS (Figure 2). There was a general decrease in concentration with depth, aside from a few exceptions, similar to prior observations for the same compounds previously studied in Arctic waters.[12] Detection of

PFAS decreased in particular for passive samplers deployed below 250 m, though PFOS and PFOA remained detectable (Figure 2). For reference, the MDL values for detection in passive samplers equate to limits near 20 pg L^{-1} when converted to time weighted average concentrations.

Site HG-IV, in the eastern Fram Strait, displayed a unique depth profile of PFAS concentrations: higher PFAS concentrations were present at a depth of 415 m than were found at the shallowest (198 m) depth, likely due to the sinking of warmer Arctic waters already enriched in PFAS, when they encounter colder Polar water (Figure 2).[16] Another exception to the decrease with depth was the observation of PFHxS at site F4, which reached a maximum concentration at a depth of 498 m and was potentially liberated from settling particles. A similar profile has been previously observed at this site for polycyclic aromatic hydrocarbons (PAHs).[16] The PFAS observed in this study suggest that sources of PFAS to the Fram Strait and Arctic are increasingly influenced by replacement compounds including shorter-chain FTOHs and PFBS. While legacy compounds such as PFOA, PFNA, PFHxS, and PFOS were all still present at high concentrations relative to other studies in the region, PFBS (a short chain PFAS) as well as PFPeA, PFHxA, and PFHpA (short chain PFCA) were present at similar concentrations (Table S11).[12,22,23,30,31] Principal component analysis was used to examine if water parameters such as depth, temperature, or salinity influenced PFAS profiles (Figure S2). Only depth strongly impacted PFAS profile composition, with deep (>800 m) samples clustering away from the 64−500 m samples (Figure S2). Results from passive and discrete grab samples are discussed in SI section S4. In addition, the PFHpA/PFOA ratios derived and whether it can indicate atmospheric deposition can be found in SI section S5.

Comparison to Legacy POP Profiles. Passive sampler-derived PFAS concentrations and depth profiles were generally similar to those reported for PAHs at the same study sites, despite different physiochemical properties and reactivities between the two groups.[16] Unlike PAHs, PFAS do not undergo the same depth-depletion via microbial degradation that has been suggested previously.[16,32] PFAS profiles, like those reported for PAHs, differ from depth profiles for polychlorinated biphenyls (PCBs) and organochlorine pesticides, which have displayed higher concentrations within the intermediate and deep waters of the Fram Strait, likely due to their earlier ban/phase outs and possible settling on particles (Figure S3).[33] For further discussion, please see SI section S6.

PFAS and Water Mass Age. Prior work established mean estimated water mass ages across the Fram Strait, which ranged from modern (last 50 years) for the top 500 m to around 50−100 years at 1000 m to about 250 years for waters at 2,000 m depth.[18] The detection of several PFAS at depth clearly shows that even the anionic PFAS, which have been produced for several decades, are not perfect water mass tracers but are also transported to depth via settling particles.[31]

PFAS Mass Flows through the Fram Strait. In the Fram Strait, the highest flow of $_{10}\Sigma$ PFAS was observed in the Atlantic Water (AW) with a mass flow of 112 Mg year^{-1} (±82) northwards into the Arctic Ocean (Figure 3). The Recirculating Atlantic Water and Arctic Atlantic Water (RAW/AAW) and Polar Water (PW) both had southward PFAS mass flows to the North Atlantic Ocean of −66 (±36) Mg year^{-1} and −33 (±19) Mg year^{-1}, respectively (Figure 3). These values are larger than mass flows reported earlier for sum PAHs, sum organophosphate esters (OPEs), and sum polybrominated diphenyl
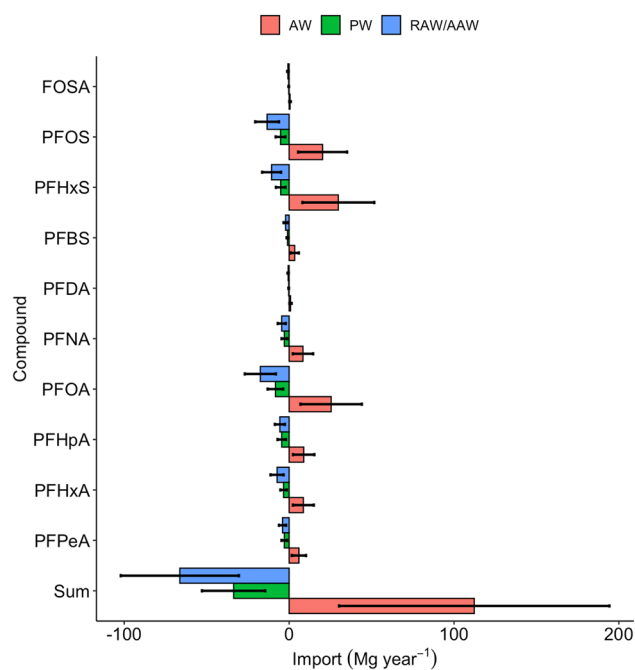
Figure 3. Estimated mass export of individual PFAS compounds in Atlantic Water (AW), Polar Water (PW), and Recirculating Atlantic Water/Arctic Atlantic Water (RAW/AAW).

ethers (PBDEs) in the Fram Strait.[15,16,33] While no PCB mass flows have been reported for these same sites in the literature, their concentrations were, at times, multiple orders of magnitude below the PFAS′ reported in this study, suggesting PFAS may constitute the largest organic contaminant mass flows across the Fram Strait that is currently known.[17] There are likely seasonal fluctuations in PFAS concentration and water mass volumes in the Fram Strait that are difficult to quantify. We hence relied on annual mean concentrations and literature estimates of water mass fluxes.

Σ_{10} PFAS mass flows across the Fram Strait were very similar in both directions, with +112 and −100 Mg year^{-1} estimated in this study. While there was good agreement for sum PFAS between Arctic imports and exports (within 20%), the difference ranged from 1 to 74% between inputs and exports for most individual PFAS (Table S12). This balance between inputs and outputs was also noted in other studies for both PFAS and PAHs in the Fram Strait.[12,16] Once again, this overall balance between inputs and outputs was also observed when using a traditional mass flux approach, albeit with more discrepancies between individual compounds' North and South fluxes, as these results are uncorrected for total water volume (Table S13). Only FOSA displayed a wider discrepancy (74% difference), with approximately twice as much being exported from the Arctic Ocean via the Fram Strait as was entering with Atlantic Water (0.56 Mg year^{-1} versus −1.0 Mg year^{-1}). This likely represents the preferential transport, deposition, and remobilization of the volatile FOSA in the Arctic Ocean. For the anionic PFAS, in contrast, very similar mass flows were estimated (Figure 3). The estimated PFAS mass flows were similar to prior estimates in the Fram Strait derived from grab samples.[12] The slight increase in export/input estimated in this study relative to the results of Joerss et al. (2020) could be due to the time weighted average nature of the data collected by these passive samplers.[12] Furthermore, these mass flow estimates rely on yearly average

water concentrations derived from passive samplers, with their 39% uncertainty, as well as the uncertainty associated with the transport volumes of each water mass reported in the literature, which ranged from 54 to 73%.[18]

Outlook. This study demonstrated the ability of passive samplers to overcome the detection limits of PFAS in remote, low-concentration marine environments. The time weighted average concentration derived in this study shows that PFOA and PFOS were still present at high concentrations in Arctic water masses, but shorter chain compounds (PFPeA, PFHxA, PFBS) were also routinely present across the Fram Strait.

The detection of shorter chain PFCAs in the Arctic water masses may reflect industry's switch to shorter chain replacements (i.e., PFBS replacing PFOS or 6:2 FTOH replacing 8:2 FTOH).[5,8,21,34] A comparison of PFAS to legacy POP profiles indicated similar trends across the Fram Strait with depth and across sites, implying that similar oceanographic processes are mostly responsible. Given the persistence of perfluorinated acids, this would also imply that biodegradation is of limited importance as a removal mechanism for those legacy POPs and PAHs.

Overall, our results demonstrate that PFAS are circulating in the Arctic Ocean and North Atlantic water masses in a feedback loop, as was previously suggested for PAHs.[16] This feedback loop consisted of the continued release of volatile compounds at mid-latitudes that are transported to high latitudes, where they are buoyed by smaller, local secondary sources and are then re-exported back to the North Atlantic Ocean.[16] In the case of PFAS, this transport is more complicated, as the volatile polyfluoroalkyl compounds may degrade into stable anionic perfluoroalkyl acids in the high northern latitudes and are then exported back to the Atlantic Ocean.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.3c00835.

> Additional analytical details, methods, standards, quality control and assurance, LC/MS conditions, details on field deployment sites and conditions, and calculation of time weighted averages. (PDF)

> Tables containing information on partition coefficients for passive samplers and further details on calculations of mass flows, time weighted average concentrations, and discrete results. (XLSX)

AUTHOR INFORMATION

Corresponding Author

Rainer Lohmann — *University of Rhode Island, Narragansett 02882, United States;* ● orcid.org/0000-0001-8796-3229; Phone: (401) 874-6612; Email: rlohmann@uri.edu

Authors

Matthew Dunn — *University of Rhode Island, Narragansett 02882, United States;* ● orcid.org/0000-0002-8902-8434

Simon Vojta — *University of Rhode Island, Narragansett 02882, United States*

Thomas Soltwedel — *Alfred Wegener Institute Helmholtz Center for Polar and Marine Research, 27570 Bremerhaven, Germany*

Wilken-Jon von Appen — *Alfred Wegener Institute Helmholtz Center for Polar and Marine Research, 27570 Bremerhaven, Germany*

Complete contact information is available at:
https://pubs.acs.org/10.1021/acs.estlett.3c00835

Notes

The authors declare no competing financial interest.

REFERENCES

(1) Stockholm Convention on Persistent Organic Pollutants (POPs). In *Encyclopedia of Corporate Social Responsibility*; Idowu, S. O., Capaldi, N., Zu, L., Gupta, A. D., Eds.; Springer Berlin Heidelberg: Berlin, Heidelberg, 2013; pp 2336−2336. DOI: 10.1007/978-3-642-28036-8_101506.

(2) Ojo, A. F.; Peng, C.; Ng, J. C. Assessing the Human Health Risks of Per- and Polyfluoroalkyl Substances: A Need for Greater Focus on Their Interactions as Mixtures. *J. Hazard. Mater.* **2021**, *407* (August 2020), 124863.

(3) Sunderland, E. M.; Hu, X. C.; Dassuncao, C.; Tokranov, A. K.; Wagner, C. C.; Allen, J. G. A Review of the Pathways of Human Exposure to Poly- and Perfluoroalkyl Substances (PFASs) and Present Understanding of Health Effects. *J. Expo. Sci. Environ. Epidemiol.* **2019**, *29* (2), 131−147.

(4) Wang, Z.; Dewitt, J. C.; Higgins, C. P.; Cousins, I. T. A Never-Ending Story of Per- and Polyfluoroalkyl Substances (PFASs)? *Environ. Sci. Technol.* **2017**, *51* (5), 2508−2518.

(5) Muir, D. C. G.; De Wit, C. A. Trends of Legacy and New Persistent Organic Pollutants in the Circumpolar Arctic: Overview, Conclusions, and Recommendations. *Sci. Total Environ.* **2010**, *408* (15), 3044−3051.

(6) Smithwick, M.; Norstrom, R. J.; Mabury, S. A.; Solomon, K.; Evans, T. J.; Stirling, I.; Taylor, M. K.; Muir, D. C. G. Temporal Trends of Perfluoroalkyl Contaminants in Polar Bears (*Ursus Maritimus*) from Two Locations in the North American Arctic, 1972−2002. *Environ. Sci. Technol.* **2006**, *40* (4), 1139−1143.

(7) Lescord, G. L.; Kidd, K. A.; De Silva, A. O.; Williamson, M.; Spencer, C.; Wang, X.; Muir, D. C. G. Perfluorinated and Polyfluorinated Compounds in Lake Food Webs from the Canadian High Arctic. *Environ. Sci. Technol.* **2015**, *49* (5), 2694−2702.

(8) Skaar, J. S.; Ræder, E. M.; Lyche, J. L.; Ahrens, L.; Kallenborn, R. Elucidation of Contamination Sources for Poly- and Perfluoroalkyl Substances (PFASs) on Svalbard (Norwegian Arctic). *Environ. Sci. Pollut. Res.* **2019**, *26* (8), 7356−7363.

(9) Yeung, L. W. Y.; De Silva, A. O.; Loi, E. I. H.; Marvin, C. H.; Taniyasu, S.; Yamashita, N.; Mabury, S. A.; Muir, D. C. G.; Lam, P. K. S. Perfluoroalkyl Substances and Extractable Organic Fluorine in Surface Sediments and Cores from Lake Ontario. *Environ. Int.* **2013**, *59* (2013), 389−397.

(10) Muir, D.; Miaz, L. T. Spatial and Temporal Trends of Perfluoroalkyl Substances in Global Ocean and Coastal Waters. *Environ. Sci. Technol.* **2021**, *55* (14), 9527−9537.

(11) MacInnis, J. J.; Lehnherr, I.; Muir, D. C. G.; Quinlan, R.; De Silva, A. O. Characterization of Perfluoroalkyl Substances in Sediment Cores

from High and Low Arctic Lakes in Canada. *Sci. Total Environ.* **2019**, *666*, 414−422.

(12) Joerss, H.; Xie, Z.; Wagner, C. C.; Von Appen, W.-J.; Sunderland, E. M.; Ebinghaus, R. Transport of Legacy Perfluoroalkyl Substances and the Replacement Compound HFPO-DA through the Atlantic Gateway to the Arctic Ocean—Is the Arctic a Sink or a Source? *Environ. Sci. Technol.* **2020**, *54* (16), 9958−9967.

(13) Hartz, W. F.; Björnsdotter, M. K.; Yeung, L. W. Y.; Hodson, A.; Thomas, E. R.; Humby, J. D.; Day, C.; Jogsten, I. E.; Kärrman, A.; Kallenborn, R. Levels and Distribution Profiles of Per- and Polyfluoroalkyl Substances (PFAS) in a High Arctic Svalbard Ice Core. *Sci. Total Environ.* **2023**, *871*, 161830.

(14) Garcia-Barrios, J.; Drysdale, M.; Ratelle, M.; Gaudreau, É.; LeBlanc, A.; Gamberg, M.; Laird, B. D. Biomarkers of Poly- and Perfluoroalkyl Substances (PFAS) in Sub-Arctic and Arctic Communities in Canada. *Int. J. Hyg. Environ. Health* **2021**, *235*, 113754.

(15) McDonough, C. A.; De Silva, A. O.; Sun, C.; Cabrerizo, A.; Adelman, D.; Soltwedel, T.; Bauerfeind, E.; Muir, D. C. G.; Lohmann, R. Dissolved Organophosphate Esters and Polybrominated Diphenyl Ethers in Remote Marine Environments: Arctic Surface Water Distributions and Net Transport through Fram Strait. *Environ. Sci. Technol.* **2018**, *52* (11), 6208−6216.

(16) Zhang, L.; Ma, Y.; Vojta, S.; Morales McDevitt, M.; Hoppmann, M.; Soltwedel, T.; Kirk, J.; De Silva, A.; Muir, D.; Lohmann, R. Presence, Sources and Transport of Polycyclic Aromatic Hydrocarbons in the Arctic Ocean. *Geophys. Res. Lett.* **2023**, *50* (1), e2022GL101496.

(17) Sun, C.; Soltwedel, T.; Bauerfeind, E.; Adelman, D. A.; Lohmann, R. Depth Profiles of Persistent Organic Pollutants in the North and Tropical Atlantic Ocean. *Environ. Sci. Technol.* **2016**, *50* (12), 6172−6179.

(18) Stöven, T.; Tanhua, T.; Hoppema, M.; Von Appen, W.-J. Transient Tracer Distributions in the Fram Strait in 2012 and Inferred Anthropogenic Carbon Content and Transport. *Ocean Sci.* **2016**, *12* (1), 319−333.

(19) Kwok, K. Y.; Yamazaki, E.; Yamashita, N.; Taniyasu, S.; Murphy, M. B.; Horii, Y.; Petrick, G.; Kallerborn, R.; Kannan, K.; Murano, K.; Lam, P. K. S. Transport of Perfluoroalkyl Substances (PFAS) from an Arctic Glacier to Downstream Locations: Implications for Sources. *Sci. Total Environ.* **2013**, *447*, 46−55.

(20) Young, C. J.; Furdui, V. I.; Franklin, J.; Koerner, R. M.; Muir, D. C. G.; Mabury, S. A. Perfluorinated Acids in Arctic Snow: New Evidence for Atmospheric Formation. *Environ. Sci. Technol.* **2007**, *41* (10), 3455−3461.

(21) Simcik, M. F.; Dorweiler, K. J. Ratio of Perfluorochemical Concentrations as a Tracer of Atmospheric Deposition to Surface Waters. *Environ. Sci. Technol.* **2005**, *39* (22), 8678−8683.

(22) Zhang, X.; Zhang, Y.; Dassuncao, C.; Lohmann, R.; Sunderland, E. M. North Atlantic Deep Water Formation Inhibits High Arctic Contamination by Continental Perfluorooctane Sulfonate Discharges. *Glob. Biogeochem. Cycles* **2017**, *31* (8), 1332−1343.

(23) Benskin, J. P.; Muir, D. C. G.; Scott, B. F.; Spencer, C.; De Silva, A. O.; Kylin, H.; Martin, J. W.; Morris, A.; Lohmann, R.; Tomy, G.; Rosenberg, B.; Taniyasu, S.; Yamashita, N. Perfluoroalkyl Acids in the Atlantic and Canadian Arctic Oceans. *Environ. Sci. Technol.* **2012**, *46* (11), 5815−5823.

(24) Savvidou, E. K.; Sha, B.; Salter, M. E.; Cousins, I. T.; Johansson, J. H. Horizontal and Vertical Distribution of Perfluoroalkyl Acids (PFAAs) in the Water Column of the Atlantic Ocean. *Environ. Sci. Technol. Lett.* **2023**, *10* (5), 418−424.

(25) Dunn, M.; Becanova, J.; Snook, J.; Ruyle, B.; Lohmann, R. Calibration of Perfluorinated Alkyl Acid Uptake Rates by a Tube Passive Sampler in Water. *ACS EST Water* **2023**, *3* (2), 332−341, DOI: 10.1021/acsestwater.2c00384.

(26) Kaserzon, S. L.; Vijayasarathy, S.; Bräunig, J.; Mueller, L.; Hawker, D. W.; Thomas, K. V.; Mueller, J. F. Calibration and Validation of a Novel Passive Sampling Device for the Time Integrative Monitoring of Per- and Polyfluoroalkyl Substances (PFASs) and Precursors in Contaminated Groundwater. *J. Hazard. Mater.* **2019**, *366* (September 2018), 423−431.

(27) Gardiner, C.; Robuck, A.; Becanova, J.; Cantwell, M.; Kaserzon, S.; Katz, D.; Mueller, J.; Lohmann, R. Field Validation of a Novel Passive Sampler for Dissolved PFAS in Surface Waters. *Environ. Toxicol. Chem.* **2022**, *41* (10), 2375−2385.

(28) Soltwedel, T.; Bauerfeind, E.; Bergmann, M.; Bracher, A.; Budaeva, N.; Busch, K.; Cherkasheva, A.; Fahl, K.; Grzelak, K.; Hasemann, C.; Jacob, M.; Kraft, A.; Lalande, C.; Metfies, K.; Nöthig, E.-M.; Meyer, K.; Quéric, N.-V.; Schewe, I.; Włodarska-Kowalczuk, M.; Klages, M. Natural Variability or Anthropogenically-Induced Variation? Insights from 15 Years of Multidisciplinary Observations at the Arctic Marine LTER Site HAUSGARTEN. *Ecol. Indic.* **2016**, *65*, 89−102.

(29) Morales-McDevitt, M. E.; Dunn, M.; Habib, A.; Vojta, S.; Becanova, J.; Lohmann, R. Poly- and Perfluorinated Alkyl Substances in Air and Water from Dhaka, Bangladesh. *Environ. Toxicol. Chem.* **2022**, *41* (2), 334−342.

(30) Ahrens, L.; Barber, J. L.; Xie, Z.; Ebinghaus, R. Longitudinal and Latitudinal Distribution of Perfluoroalkyl Compounds in the Surface Water of the Atlantic Ocean. *Environ. Sci. Technol.* **2009**, *43* (9), 3122−3127.

(31) Yamashita, N.; Taniyasu, S.; Petrick, G.; Wei, S.; Gamo, T.; Lam, P. K. S.; Kannan, K. Perfluorinated Acids as Novel Chemical Tracers of Global Circulation of Ocean Waters. *Chemosphere* **2008**, *70* (7), 1247−1255.

(32) González-Gaya, B.; Martínez-Varela, A.; Vila-Costa, M.; Casal, P.; Cerro-Gálvez, E.; Berrojalbiz, N.; Lundin, D.; Vidal, M.; Mompeán, C.; Bode, A.; Jiménez, B.; Dachs, J. Biodegradation as an Important Sink of Aromatic Hydrocarbons in the Oceans. *Nat. Geosci.* **2019**, *12* (2), 119−125.

(33) Ma, D.; Zhong, H.; Lv, J.; Wang, Y.; Jiang, G. Levels, Distributions, and Sources of Legacy and Novel per- and Perfluoroalkyl Substances (PFAS) in the Topsoil of Tianjin, China. *J. Environ. Sci.* **2022**, *112*, 71−81.

(34) Nilsson, H.; Kärrman, A.; Rotander, A.; van Bavel, B.; Lindström, G.; Westberg, H. Biotransformation of Fluorotelomer Compound to Perfluorocarboxylates in Humans. *Environ. Int.* **2013**, *51* (2013), 8−12.

■ NOTE ADDED AFTER ASAP PUBLICATION

The Abstract graphic and Figure 3 were corrected on January 10, 2024.