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Polyfluorinated compounds in the atmosphere along a cruise pathway from the Japan Sea to the Arctic Ocean

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Neutral polyfluorinated alkyl substances (PFASs) were measured in high-volume air samples collected on board the research vessel *Snow Dragon* during the 4th Chinese National Arctic Expedition from the Japan Sea to the Arctic Ocean in 2010. Four volatile and semi-volatile...
PFASs (fluorotelomer alcohols (FTOHs), fluorotelomer acids (FTAs), perfluoroalkyl sulfonamides (FASAs), and sulfonamidoethanols (FASEs)) were analyzed respectively in the gas and particle phases. FTOHs were the dominant PFASs in the gas phase (61−358 pg/m³), followed by FTAs (5.2−47.9 pg/m³), FASEs (1.9−15.0 pg/m³), and FASAs (0.5−2.1 pg/m³). In the particle phase, the dominant PFAS class was FTOHs (1.0−9.9 pg/m³). The particle-associated fraction followed the general trend of FASEs > FASAs > FTOHs. Compared with other atmospheric PFAS measurements, the ranges of concentrations of $\Sigma$FTOH in this study were similar to those reported from Toronto, North America (urban), the northeast Atlantic Ocean, and northern Germany. Significant correlations between FASEs in the gas phase and ambient air temperature indicate that cold surfaces such as sea-ice, snowpack, and surface seawater influence atmospheric FASEs.

**Keywords:** Polyfluoroalkyl compounds; FTOHs; FTAs; FASE/FASAs; Arctic.

1. Introduction

In recent years, ionic per- or polyfluorinated alkyl substances (PFASs) have been detected globally (Giesy and Kannan, 2001). Among these ubiquitous compounds have been found perfluorooctane sulfonate (PFOS) and perfluorocarboxylates (PFCAs) such as perfluorooctanoate (PFOA), which are widespread and persistent environmental contaminants that have been detected in polar bears from the Canadian and European Arctic (Smithwick et al., 2005a, 2005b), dolphins in the Atlantic Ocean (Houde et al., 2005), seawater from the Pacific, Atlantic, and Arctic Oceans (Yamashita et al., 2005; Wei et al., 2007; Ahrens et al., 2009, 2010), and fur seal pups and penguin eggs in Antarctica (Schiavone et al., 2009).

The stability of PFOS and PFCAs precludes degradation or metabolism, which contributes to their global dispersion, persistence, and bioaccumulation. High water solubility and a low Henry’s law constant render PFOS and PFCAs susceptible to wet deposition, making it
unlikely that they are transported long distances atmospherically in the vapor phase (Loewen et al., 2008). It has been hypothesized that these chemicals are transported atmospherically as volatile precursors to remote areas where their degradation products are then deposited (Renner, 2001). Alternatively, long-range transport of PFCAs and PFOS to remote areas by ocean currents has also been hypothesized (Prevedouros et al., 2006).

Atmospheric transport with subsequent degradation of volatile and semi-volatile precursor compounds (e.g., fluorotelomer alcohols (FTOHs), perfluoroalkyl sulfonamides (FASAs), and perfluoroalkyl sulfonamidoethanols (FASEs)) to persistent PFCAs and PFOS has been hypothesized as a main transport mechanism (Ellis et al., 2004; Sulbaek Andersen et al., 2005; D'Eon et al., 2006; Martin et al., 2006). Because of their slow reaction with hydroxyl radicals, atmospheric lifetimes of more than 10–20 days for FTOHs and more than 20–50 days for FASAs have been estimated in smog chamber studies (Ellis et al., 2003; Martin et al., 2006). Field studies have indicated an atmospheric residence time of more than 50 days for FTOHs (Piekarz et al., 2007). These data indicate the possibility of regional and long-range atmospheric transport of FTOHs and FASAs. PFCAs and perfluoroalkyl sulfonates (PFSAs) have also been detected in snow samples from the high Arctic, where contamination is solely from atmospheric sources. This finding confirms the indirect precursor-based atmospheric transport and deposition of these compounds (Young et al., 2007).

Several datasets are available for ambient air as well as for indoor air samples. 6:2 FTOH, 8:2 FTOH, 10:2 FTOH, N-ethyl fluoroctane sulfonamide (EtFOSA), N-methyl fluoroctane sulfonamidoethanol (MeFOSE), and N-ethyl fluoroctane sulfonamidoethanol (EtFOSE) were found to be widely distributed in the atmosphere of North America, Europe, and over an Atlantic transect (Martin et al., 2002; Shoeib et al., 2004; Stock et al., 2004b; Boulanger et al., 2005; Shoeib et al., 2005; Dreyer and Ebinghaus, 2009; Dreyer et al., 2009a, 2009b). PFASs are manufactured by two main synthesis routes. Generally, the production of FASAs/FASEs is
associated with the electrochemical fluorination (ECF) process, which has been in use since
the 1950s. FASAs/FASEs are predominantly used as paper protectors, as water- and dirt-
proofing in carpets/leathers/textiles, as performance chemicals (e.g., in aqueous film-forming
fire-fighting foams, AFFFs) (De Voogt and Saez, 2006), and as an insecticide (Sulfluramid).
The second manufacturing process, telomerization, has been in use since the 1970s to produce
exclusively linear compounds, including FTOHs. The production of FTOHs has continued to
increase, particularly for use as precursors in the production of fluorinated polymers used in
paper and carpet treatments, as well as in the production of paints, coatings, and adhesives
(Dinglasan et al., 2006).

The aim of this study was to investigate the inter-hemispherical gradient of neutral (volatile
and semi-volatile) precursors of PFOS and PFCAs in ambient air, starting from the Japan Sea,
adjacent to urbanized regions, and then on a transect towards the Arctic Ocean. Data presented
in this study yield new information on the spatial atmospheric distribution of PFASs on a
regional scale, as well as the identification of their source regions in the Northern Hemisphere.
The airborne PFAS data are compared with the results of previous studies. This study provides
the first evidence of the occurrence of airborne PFASs in ambient air samples from the Japan
Sea to the Arctic Ocean.

2. Materials and Methods

2.1. Chemicals and Standards

All solvents (Methanol, acetone, dichloromethane, and n-hexane) were residue-grade and
additionally distilled in a full glass unit prior to use. Details of the analytical standards of
FTOHs (FTOH 6:2, 8:2, 10:2, and 12:2), fluorotelomer acrylates (FTA 6:2, 8:2, and 10:2),
Me- and EtFOSA, Me- and EtFOSE, and mass-labeled internal standards are summarized in Table S2 (Supplementary Materials).

### 2.2 Sampling cruise

Air samples were taken onboard the Arctic expedition of the **R/V Snow Dragon** from the Japan Sea to the high Arctic (33.2–84.5 °N) during June–September 2010 (see Fig. 1). Air samples (~500 m³/sample, ~48 h) were taken using a high–volume air sampler placed in the front of the ship’s upper deck (20 m above sea level), with a glass fiber filter ([GFF], GF/F, pore size: 0.7 μm) to trap airborne particles, and a self–packed PUF/XAD–2 glass column (PUF: Ø 5.0 cm x 2.5 cm; 35 g XAD-2, particle size: 0.3-1.0 mm) for the gaseous phase. Both air column and filters were stored at –20 °C until analysis. Sampling parameters, including date, position, temperature, and wind speed, are listed in Table S1 in the Supplementary materials.

### 2.3 Sample preparation and instrumental analysis

Extraction and clean–up of the samples was done in a clean laboratory (class 10000). All solvents were residue–analysis grade and were distilled prior to use. Samples were spiked with 2500 pg of mass labeled internal standards (Wellington Laboratories) prior to extraction. Air columns were extracted in a modified Soxhlet apparatus for 16 h using dichloromethane/hexane (1/1), and air filter samples were Soxhlet extracted using dichloromethane for 16 h, respectively. Extracts were evaporated to 1–2 mL with rotary evaporator and then passed a glass column (i.d. 1 cm, length: 15 cm) packed with 3 g Na₂SO₄.
to remove residual water. The extracts were further concentrated down to 150 µL and spiked with 1000 pg $^{13}$C–HCB as injection standard.

Detailed analysis methods have been described elsewhere (Dreyer et al., 2008). In brief, analysis was performed using gas chromatography–mass spectrometry (GC–MS) using an Agilent 6890 gas chromatograph and an Agilent 5973 mass spectrometer, Agilent Technologies) in selective ion monitoring (SIM) mode with positive chemical ionization (PCI). The ion masses are summarized in Table S2 in the Supplementary Materials. The response factors were derived from the calibration curves (6-points) made for response ratio between targets compounds (0–250 ng/mL) and mass labeled internal standards (12.5 ng/mL).”

2.4 Quality assurance and control

All air columns were pre-cleaned with organic solvents (methanol, acetone, and hexane/acetone in turn for 96 h) and GFFs were heated at 450 °C for 12 h prior to use. Na$_2$SO$_4$ was cleaned with dichloromethane for 12 h and heated at 450 °C for 12 h prior to use. To avoid degradation of the target compounds, air columns were protected against UV–sunlight during sampling using aluminum foil. Breakthrough of the target analytes using these sampling methods has been checked previously on board the German icebreaker *R/V Polarstern*. Three field blanks were run for each sample type, with blanks showing very low values ranging from 0.01 to 0.2 pg/m$^3$ (12:2 FTOH). Method detection limits (MDLs) were derived from mean blank values plus three times the standard deviation ($\sigma$) (for compounds
showing no blanks, a peak area of 100 was adopted as background response), which ranged from 0.1 to 0.3 pg/m³ for both gas and particle phases.

2.5 Air Mass Back-Trajectories

Air mass origins of the individual air samples collected during the cruise segments were calculated using NOAA’s HYSPLIT model. Air mass back-trajectories (BTs) were calculated in 6-h steps tracing back the air masses for 5 days using the sampling height as arrival height.

3. Results and discussion

3.1. Concentrations of Airborne PFASs

Total air concentrations of target compounds at various points sampled along the cruise route are presented in Figure 1 and summarized in Table 1 (for details, see Table S3, Supplementary Materials). PFASs were detected in all air samples, with FTOHs/FTAs and FASAs/FASEs ubiquitous at the sites sampled, except for MeFOSA, which was not detected at sample site A6.

3.1.1. FTOHs, FTAs, FASEs, and FASAs in the gas phase

The PFAS concentrations in the gas phase ranged from 77.9 to 416 pg/m³, and the highest gas-phase total concentrations of PFASs (416 pg/m³) were measured in sample A2 in the Okhotsk Sea, followed by sample A1 (346 pg/m³) in the Sea of Japan. The lowest total PFAS concentrations were found in samples A4 (78 pg/m³) in the Bering Sea and A9 (118 pg/m³) in the Beaufort Sea.

Generally, the FTOH concentrations were about 10 times higher than those of FTAs and about 24 times those of FASAs/FASEs. The dominant FTOH was 8:2 FTOH (44.6–286 pg/m³),
followed by 10:2 FTOH (6.1–47 pg/m³), 12:2 FTOH (4.2–16 pg/m³), and 6:2 FTOH (0.8–12 pg/m³). This distribution pattern was the same at almost all sampling sites. Interestingly, 12:2 FTOH was detected in north Germany in a previous study, with the concentration ranging from 1.3 to 8.0 pg/m³ (Dreyer and Ebinghaus, 2009).

The chemical pattern of FTOHs in the present study was, in order, 8:2 FTOH > 10:2 FTOH > 6:2 FTOH from the Japan Sea to the Arctic Ocean, a pattern similar to that reported for the Arctic (Shoeib et al., 2006), northeast Atlantic Ocean (Shoeib et al., 2010), and the western USA and Japan (Piekarz et al., 2007; Oono et al., 2008a, 2008b). The chemical pattern of FTOHs was consistent with the estimated atmospheric residence times of FTOHs, with 80 d, 70 d, and 50 d for 8:2 FTOH, 10:2 FTOH, and 6:2 FTOH, respectively (Piekarz et al., 2007).

For the FTAs, 8:2 FTA was dominant (3.3–28.7 pg/m³), followed by 10:2 FTA (1.8–19.0 pg/m³) and 6:2 FTA (0.1–0.2 pg/m³). FTAs were also detected in north Germany, with values of 1.7–15 pg/m³ for 8:2 FTA, 0.8–6.6 pg/m³ for 10:2 FTA, and 4.2–5.7 pg/m³ for 6:2 FTA (Dreyer and Ebinghaus, 2009), closely similar to the chemical pattern of FTAs measured in the present study. FTAs were not included in previous studies from Northwest Europe (Barber et al., 2007), North America (Stock et al., 2004b), Western Canada (Loewen et al., 2008), the Canadian Arctic (Stock et al., 2007), the northeast Atlantic Ocean (Shoeib et al., 2010), and the Arctic atmosphere (Shoeib et al., 2006).

For the FASAs/FASEs, MeFBSE was dominant (1.7–15 pg/m³), followed by MeFBSA (0.2–1.3 pg/m³), EtFOSE (0.1–0.7 pg/m³), EtFOSA (0.1–0.9 pg/m³), MeFOSE (0.1–0.6 pg/m³), and MeFOSA (0–0.3 pg/m³). The concentrations of FASEs were much higher than those of FASAs. The atmospheric lifetimes of FASEs were shorter than those of FASAs (~2 d in contrast to >20 d) (D’Eon et al., 2006; Martin et al., 2006), whilst MeFBSE has a lifetime of 2 days, much shorter than the 20–50 day lifetime of its parent compound MeFBSA. This result suggests that FOSEs probably originated from nearby pollution sources or were transported by
mist aerosols, which arrived in the study regions without long-range atmospheric transportation, especially for MeFBSE. Surprisingly, the concentration of MeFBSE was about one order of magnitude higher than that of MeFBSA, even in the Arctic region, from sampling sites A8 to A12. It is thought that the PFASs enriched in the sea surface microlayer may be transported into the atmosphere as organic-rich particles via wave–wind interactions (Cincinelli et al., 2001). Prevedouros et al. (2006) also indicated that PFASs concentrated in the surface waters of oceans and rivers could be transported into the air as marine aerosols, and that they could partition onto particle surfaces when spray droplets evaporated, and thus be transported long distances in the atmosphere. The MeFOSE measured in this study probably reached the Arctic region through marine aerosol transportation, suggesting that this transmission mode is possible for other FASEs.

3.1.2. FTOHs, FTAs, FASEs, and FASAs in the particle phase

All FTOHs, FTAs, FOSAs, and FOSEs were quantified in the particle phase of ambient air from the Japan Sea to the Arctic Ocean. The \( \Sigma \)FTOH/FTA/FASA/FASE concentrations in the particle phase ranged between 2.2 and 13.1 pg/m³ and were hence about 35 times lower than in the gas phase. The highest \( \Sigma \)FTOH/FTA/FASA/FASE concentrations were found in the Bering Sea and Beaufort Sea (13.1 and 11.3 pg/m³, respectively); the lowest concentrations were found in the Arctic Ocean and East China Sea (2.2 and 2.4 pg/m³, respectively). The dominant PFAS classes were FTOHs (1.0–9.9 pg/m³) with 8:2 FTOH as the dominant compound (0.2–8.2 pg/m³), followed by FASEs (0.44–4.9 pg/m³) with MeFBSE (0.1–4.5 pg/m³). The concentrations of individual FTA and FASA compounds were below 0.3 pg/m³, while the \( \Sigma \)FTOH concentrations were about 14 and 23 times higher than the \( \Sigma \)FTA and \( \Sigma \)FASA concentrations, respectively, and the \( \Sigma \)FASE concentrations were about 7 and 11 times higher than those of \( \Sigma \)FTAs and \( \Sigma \)FASAs, respectively.
3.1.3. Gas/particle partitioning

The distribution ratio of the particle and gas phases of a chemical will influence its fate and transport in the atmosphere. While the FTOHs, FTAs, FASAs, and FASEs were dominantly distributed in the gas phase, individual compounds showed a different partitioning profile (see Fig. 2). The particle-associated fraction ($\varphi = c_{\text{particulate}}/(c_{\text{particulate}} + c_{\text{gaseous}})$) represents the fraction of particles ($c_{\text{particulate}}$) in relation to the gas phase ($c_{\text{gaseous}}$) in air. In the present study, a significant proportion of FASEs/FASAs were in the particulate phase in the samples, with an average of 53% for EtFOSE, 48% for MeFOSE, 21% for MeFBSE, 21% for MeFOSA, 17% for EtFOSA, and 2% for MeFBSA. The functional group has the greatest influence on the portioning of PFCs; the highest particle-associated fraction was observed for FASEs (~26%), followed by FASAs (12%), FTOHs (2.7%), and FTAs (1.8%). A similar pattern was observed in previous studies from other regions, such as the Atlantic Ocean (Shoeib et al., 2010), North Atlantic and Canadian Archipelago, the Arctic (Shoeib et al., 2006), Toronto, Canada (Shoeib et al., 2006), Germany (Jahnke et al., 2007a), the UK (Barber et al., 2007), and northwest Europe (Barber et al., 2007). Interestingly, the particle-associated fraction of ethyl FOSE and FOSA (53% and 17%, respectively) was comparable to the methyl FOSE and FOSA (48% and 21%, respectively), suggesting that the length of alkyl carbon chain had little influence on the gas/particle partition of FASEs/FASAs. Furthermore, the particle-associated fraction for FTOHs was, in order, 12:2 FTOH > 6:2 FTOH > 10:2 FTOH > 8:2 FTOH, which was not consistent with the thermodynamic law that the vapor pressure of the longer-chain FTOHs is lower than in the shorter-chain FTOHs (e.g., 144 Pa for 10:2 FTOH; 254 Pa for 8:2 FTOH; 713 Pa for 6:2 FTOH; Stock et al., 2004a). The reasons for this are not clear, but may be related to differences in particulate matter, a potential contribution of sea spray, and/or mist aerosols (Prevedouros et al., 2006) in the cruise samples that may absorb to the GFF and
contain FTOHs, or differences in atmospheric residence times.

3.1.4. Correlation analyses

Of the sum concentrations of all substance classes in the cruise samples, $\sum$FTOH and $\sum$FTA showed a highly significant positive correlation (Spearman correlation: $p < 0.01$, $r_s = 0.729$). FTOHs were used as precursors to produce FTAs for fluorotelomer-based polymers (Prevedouros et al., 2006), which suggests the possible FTOHs and FTAs contamination originated from the similar sources. $\sum$FASAs and $\sum$FASEs were positively correlated ($p < 0.05$, $r_s = 0.575$), indicating that the sources of these compounds are related. For individual PFASs (Table S4, Supplementary Materials), the concentrations of FTOHs were significantly correlated with each other ($p < 0.05$), except 6:2 FTOH. The concentration of 8:2 FTA correlated significantly with that of 10:2 FTA ($p < 0.01$, $r_s = 0.973$). Significantly positive correlations were also observed between MeFBSA and MeFBSE ($p < 0.01$, $r_s = 0.725$), and the concentrations of the two compounds were higher than that of other FASAs/FASEs. This result suggests the sources of MeFBSA and MeFBSE are related, and probably were derived from sources different than those of other FASAs/FASEs. Furthermore, the concentrations of MeFOSE and EtFOSE were significantly correlated ($p < 0.01$, $r_s = 0.918$). Interestingly, MeFOSA and EtFOSA were not significantly positively correlated with other PFAS ($p > 0.05$), which points to a variability in the source and/or differences in atmospheric persistence among these chemicals.

3.2. Comparison with other reported airborne PFASs

Figure 3 provides an overview of $\sum$FTOH and $\sum$FASA/FASE concentrations determined in this study and reported by other research groups. However, it should be noted that analyzed substances in the previous studies differed to some extent (Stock et al., 2004b; Shoeib et al.,
In general, the concentrations of \( \sum \text{FTOHs} \) in this study were in the same ranges as seen in studies of Toronto, North America (urban), the Northeast Atlantic Ocean, and Northern Germany, higher than those of the North Atlantic and Canadian Archipelago, Arctic (Stock et al., 2004b; Dreyer and Ebinghaus, 2009; Shoeib et al., 2006, 2010), but lower than those of urban and traffic-influenced areas in England, Germany, Northwest Europe, and Japan (Barber et al., 2007; Jahnke et al., 2007a; Oono et al., 2008a, 2008b). This result indicates that the industrialized regions are potential sources of \( \sum \text{FTOHs} \).

Compared with data published for other regions, \( \sum \text{FTOH} \) levels were significantly higher in Japan, probably suggesting point sources, which is consistent with the higher \( \sum \text{FTOHs} \) at sampling sites A1 (Sea of Japan), A2 (Bering Sea), and A13 (East China Sea) in this study. As for \( \sum \text{FASAs}/\text{FASEs} \), the concentrations were lower than in all regions studied previously, except the Southern Hemisphere (Jahnke et al., 2007b). Similarly, the higher concentrations of \( \sum \text{FASAs}/\text{FASEs} \) were also correlated with industrialized regions, and \( \sum \text{FASA}/\text{FASE} \) levels were significantly higher in North America, suggesting possible point sources.

The \( \sum \text{FTOH}/\text{FASA}/\text{FASE} \) concentrations (2.0–13 pg/m\(^3\)) in the particle phase were similar to those of Toronto, Canada (Shoeib et al., 2006), Waldhof (rural area), Germany (Jahnke et al., 2007a), Hazelrigg, UK (Barber et al., 2007), and Kjeller, Norway (Barber et al., 2007), and were about 2–3 times lower than those of the North Atlantic and Canadian Archipelago, Arctic (Shoeib et al., 2006), Hambury (urban area), Germany (Jahnke et al., 2007a), Northern Germany (Dreyer and Ebinghaus, 2009), and Manchester, UK (Barber et al., 2007). In addition, the pattern of the PFAS classes in the particle phase followed the trend \( \text{FASEs} > \text{FASAs} > \text{FTOHs} \), which was in accordance with the general trends observed in other areas, such as Northern Germany; Hazelrigg and Manchester, UK; Kjeller, Norway; and Hamburg and
3.3. Identification of air mass sources and influence of ambient temperatures on FASEs

gas/particle partition

Air mass back-trajectories were used to determine the source routes of PFASs, and all 13 sites were primarily oceanic in origin. The air mass back-trajectories (6-h steps) and altitudinal profiles of the air mass parcels of five selected stations (A1 (120 h), A5 (120 h), A11 (96 h), A13 (120 h), and A15 (120 h)) are shown in Fig. 4. As the analysis of air mass back-trajectories shows, the air masses were mainly from oceanic source routes, even for the samples taken in the Japan Sea. We therefore investigated the influence of ambient temperature on the airborne concentration of volatile and semi-volatile PFASs, and their distribution between the gaseous and particulate phases, to identify the probable influence of sea/air exchange. FTOHs, FTAs, and FASAs showed no significant correlation with ambient temperature in the gas/particle phase, which indicates that their presence is not associated with a temperature effect. However, we found a significant correlation of FASE concentrations (sum of gas and particulate phases) with ambient temperature (p < 0.05, $R^2 = 0.699$), and a significant correlation was also found between FASE concentrations in the gas phase and ambient temperature (p < 0.05, $R^2 = 0.740$) (see Fig. 5). Similar results were also observed in Germany, and the North Atlantic and Canadian Archipelago (Shoeib et al., 2006; Jahnke et al., 2007a). This phenomenon could be related partly to the significant revolatilization of particle-bound compounds from the filter at higher temperatures (Gundel et al., 1995) or associated with the process of sea/air exchange. Further investigations are needed, especially studies of the concentrations of FASEs in the sea surface microlayer or sub-layer in these areas. In the present study, although the influence from the continent is not that high, much higher contaminations would be expected if the air had come directly from Asian countries. The data
with higher FTA concentrations (5.2–48 pg/m$^3$) presented in this study agree with the higher concentrations reported in Japan, which confirmed an Asian origin (Oono et al., 2008a, 2008b); fewer data have been reported from Europe and North America.

4. Conclusions

Generally, the concentrations of PFASs were higher in urban regions, and FTOHs were the most dominant compounds of PFASs, followed by FTAs, FASEs, and FASAs. The relatively high concentrations of FTOHs at sample sites A1 (Sea of Japan), A2 (Bering Sea), and A13 (East China Sea) were probably caused by the countries of East Asia, where $\Sigma$FTOH and $\Sigma$FTA levels are much higher than in Europe and North America. The particle-associated fractions of PFASs followed the general trend FASEs > FASAs > FTOHs. Given the importance of particle-gas partitioning for determining the fate and transport pathways of semi-volatile chemicals, further empirical studies are required. Ambient air temperature had a significant influence on the partitioning behavior of FASEs, indicating that the atmospheric behavior of these compounds was influenced by temperature-driven surface exchanges, possibly including extensive areas of sea-ice, snow pack, and surface seawater.

In summary, this study documents the first measurements of FTOHs/FTAs and FASAs/FASEs in ambient air sampled on a cruise transect from the Japan Sea to the Arctic Ocean. Our study shows the wide distribution of FTOHs/FTAs and FASAs/FASEs in the ambient atmosphere, not only in low-latitude urban regions but also in remote Arctic regions. The findings confirm the hypothesis of long-range atmospheric transport of neutral, volatile precursors of PFCAs and PFOS to remote regions.

Acknowledgments

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Appendix A: Supplementary materials

Supplementary data associated with this article can be found in Appendix A: Supplementary materials.

References:


Schiavone, A., Corsolini, S., Kannan, K., Tao, L., Trivelpiece, W., Torres Jr., D., Focardi, S., 2009. Perfluorinated contaminants in fur seal pups and penguin eggs from South Shetland,


**Figure Captions**

**Figure 1.** Total concentrations of PFASs in the atmosphere along the cruise track from the Japan Sea to Arctic Ocean.

**Figure 2.** Composition profile of PFASs in the gaseous phase (left) and particle phase (right) along the sampling transect.

**Figure 3.** Concentrations (minimum, maximum) of FTOHs and FASAs/FASEs in ambient air of different areas ((a) Shoeib et al., 2006, (b) Loewen et al., 2008, (c) Stock et al., 2004b, (d) Shoeib et al., 2010, (e) Barber et al., 2007, (f) Dreyer and Ebinghaus, 2009, (g) Jahnke et al., 2007a, (h) Oono et al., 2008b.)

**Figure 4.** Air mass back trajectories (6 h steps) and altitudinal profiles of the air mass parcels for the selected air samples A1 (120 h), A5 (120 h), A11 (96 h), A13 (120 h) and A15 (120 h). The black line indicates the cruise leg.

**Figure 5.** Correlations between FASEs concentrations and ambient temperatures in atmosphere of sampling stations.
<table>
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<tr>
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<th>Phase</th>
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<th>Arithmetic mean</th>
<th>Standard deviation</th>
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Figure 1.
Figure 2.
This study

Figure 3.
Figure 5.