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Assessing temporal trends and source regions of per- and polyfluoroalkyl substances (PFASs) in air under the Arctic Monitoring and Assessment Programme (AMAP)

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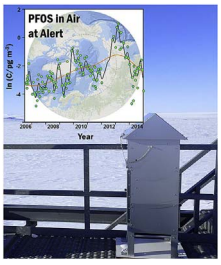
GRAPHICAL ABSTRACT



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ABSTRACT

Long-term Arctic air monitoring of per- and polyfluoroalkyl substances (PFASs) is essential in assessing their long-range transport and for evaluating the effectiveness of chemical control initiatives. We report for the first time temporal trends of neutral and ionic PFASs in air from three arctic stations: Alert (Canada, 2006–2014); Zeppelin (Svalbard, Norway, 2006–2014) and Andøya (Norway, 2010–2014). The most abundant PFASs were the perfluorooctanoic acid (PFOA), perfluorooctane sulfonic acid (PFOS), perfluorobutanoic acid (PFBA), and fluorotelomer alcohols (FTOHs). All of these chemicals exhibited increasing trends at Alert with doubling times (t_2) of 3.7 years (y) for PFOA, 2.9 y for PFOS, 2.5 y for PFBA, 5.0 y for 8:2 FTOH and 7.0 y for 10:2 FTOH. In contrast, declining or non-changing trends, were observed for PFOA and PFOS at Zeppelin (PFOA, half-life, $t_{1/2}$ = 7.2 y; PFOS $t_{1/2}$ = 67 y), and Andøya (PFOA $t_{1/2}$ = 1.9 y; PFOS $t_{1/2}$ = 11 y). The differences in air concentrations and in time trends between the three sites may reflect the differences in regional regulations and source regions. We investigate the source region for particle associated compounds using the Lagrangian particle dispersion model FLEXPART. Model results showed that PFOA and PFOS are impacted by air masses originating from the ocean or land. For instance, PFOA at Alert and PFOS at Zeppelin were dominated by oceanic air masses whereas, PFOS at Alert and PFOA at Zeppelin were influenced by air masses transported from land.

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1. Introduction

Per- and polyfluoroalkyl substances (PFASs) have been applied in a wide variety of consumer and industrial products such as oil and liquid repellents (Begley et al., 2005), firefighting foams (Ahrens et al., 2015), pesticides (Schnellmann, 1990), and food packaging materials (Gebbink et al., 2013). PFASs are ubiquitous in environmental media, such as air (Shoeib et al., 2006; Dreyer et al., 2009; Genualdi et al., 2010; Xie et al., 2013, 2015; Gawor et al., 2014; Bossi et al., 2016), snow (Kirchgeorg et al., 2013), and water (Ahrens and Bundschuh, 2014). They are also found in humans (Kato et al., 2011; Gebbink et al., 2015) and wildlife (Giesy and Kannan, 2001; Riget et al., 2010; Braune and Letcher, 2013). Among the entire chemical family of PFASs, perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) have received the most attention to date due to their environmental persistence, bioaccumulation and potential adverse effects on humans and wildlife.

PFOS, its salts and perfluorooctane sulfonyl fluoride (PFOSF), have been listed under Annex B of the Stockholm Convention on Persistent Organic Pollutants (POPs) (UNEP, 2017a) as well as in the amendment of the Aarhus protocol on POPs under the Convention on Long-range Transboundary Air Pollution (CLRTAP) (UNECE, 2015). In 2002, 3M, the main global manufacturer of PFOS at that time, voluntarily phased out the production of the chemical. PFOA, its salts and C_9 to C_{14}-based perfluoroalkyl carboxylic acids (PFCAs), are listed in the Candidate List of Substances of Very High Concern in Europe under Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) (ECHA, 2012, 2015, 2017). PFOA is being proposed for listing under the Stockholm Convention (UNEP, 2017b). In Canada and US, PFOA is restricted to specific use (Government of Canada, 2016a, b; USEPA, 2016).

PFASs are known to undergo long-range transport to remote environments, such as the Arctic, via oceanic or atmospheric transport, though the mechanism is not clear. Neutral PFASs, such as fluorotelomer alcohols (FTOHs), perfluorooctane sulfonamidoethanols (FOSEs) and perfluorooctane sulfonamides (FOSAs), are more volatile than perfluoroalkyl acids (PFAAs) and are susceptible to long-range atmospheric transport (LRAT) to the Arctic (Shoeib et al., 2006; Young et al., 2007; Schenker et al., 2008; Ahrens et al., 2011a). Neutral PFASs can be degraded in the atmosphere to form PFAAs and subsequently deposit via wet or dry atmospheric deposition (Ellis et al., 2004; Hurley et al., 2004; Martin et al., 2006; Schenker et al., 2008). PFAAs, also referred to as the ionic PFASs, are more water soluble and less volatile than neutral PFASs. Early modelling studies indicated that PFAAs are more likely to be transported via oceanic currents than the atmosphere (Armitage et al., 2006; Wania, 2007) but recent study by Yeung et al. (2017) suggested that atmospheric input accounts for the majority of measured PFOA in the Arctic Ocean.

The atmosphere is the medium with the fastest response to changes in emissions, hence the temporal variation of contaminants in remote Arctic air may indicate the effectiveness of chemical control regulations and changes in usage pattern. Here, we provide the first overview of measured concentrations and time trends of PFASs in Arctic air. We analyzed long-term atmospheric monitoring data of PFASs at three Arctic stations, namely Alert (Canada) from 2006 to 2014, Zeppelin (Svalbard, Norway) from 2006 to 2014, and Andøya (Norway) from 2010 to 2014. The occurrences, relative abundances, and distribution of the different groups of PFASs (neutral PFASs and PFAAs), their seasonal and long-term variations, source regions, and temperature dependence are discussed. This work is a joint effort by Canada's Northern Contaminants Programme (NCP) and the national Norwegian Pollutant Monitoring Program (SFT) in collaboration under the Arctic Council's Arctic Monitoring and Assessment Programme (AMAP). Data presented here are produced by two national long-term air monitoring programs for organic pollutants initiated in the 1990s. Each program follows its own established protocol for sample collection, chemical analysis, data management, quality assurance and control (QA/QC).

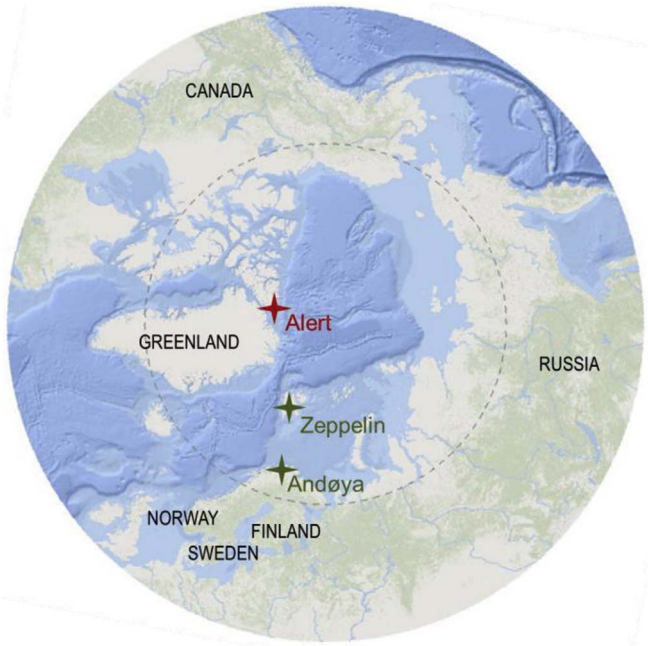

Fig. 1. Map of sampling sites.

2. Material and methods

2.1. Sampling sites and method

Air sampling was carried out at the Canadian High Arctic station of Alert, Nunavut, Canada (82° 30′ N 62° 20′ W) and the Norwegian-operated stations of Zeppelin, Svalbard, Norway (78° 54′ N 11° 53′ E) and Andøya, Norway (62° 16′ N 16° 0′ E). Fig. 1 shows the locations of the three sampling sites. Samples from the Canadian site, Alert, were analyzed by Environment and Climate Change Canada (ECCC) and those from the Norwegian sites were analyzed by the Norwegian Institute for Air Research (NILU). Data for the Norwegian sites are extracted from the European Monitoring and Evaluation Programme (EMEP) Website (http://ebas.nilu.no/).

At Alert, air samples (n = 169) were collected from August 2006 to February 2015 using a Tisch PS-1 high-volume active air sampler (HV-AAS) (Tisch Environmental, Cleves, OH) made of stainless steel (volatile methyl siloxane (VMS)-free sealing in the HV-AAS sampling head). Each sample represents ~2000 m^3 of air sampled over 7 days. Sampling occurred once per month from October to February and once every other week between March and September. Each sample comprised of a glass fiber filter (GFF, TE-G653, TISCH Environmental Inc.), and a cartridge containing two polyurethane foams (PUF, 2 × 2.5 inch diameter and 1 × 2.5 inch diameter, Tisch Environmental Inc.), and 15 g of XAD-2 (Supelpak™-2, 20–60 mesh, pre-cleaned, Sigma Aldrich). Hence, both gas- and particle phase PFASs were collected.

At Zeppelin, air samples (n = 383) were collected from September 2006 to December 2014, while at Andøya, air samples (n = 249) were collected from November 2009 to December 2014. The Norwegian air samples were taken with a Digitel (DH77) HV-AAS using only a GFF. This results in sampling of solely the particle-phase PFASs. Sampling at the Norwegian sites occurred on a weekly basis, with one sample over 48 h collecting an air volume of ~1 200 m^3. Exceptions to these sampling procedures were that at Zeppelin in 2007, air samples were collected every second week, and at Andøya the sampling duration was 24 h instead of 48 h from November 2009 to September 2010. Details of the sampling protocol for the Norwegian sites are described elsewhere (Nizzetto and Aas, 2016).

The targeted PFASs for air samples from Alert included i) neutral PFASs: 6:2, 8:2, 10:2 FTOHs, 6:2, 8:2, 10:2 fluorotelomer acrylates

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