Per- and polyfluoroalkyl Substances (PFAS) in atmosphere and deposition

Short review

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1 Introduction

Per- and polyfluoroalkyl substances (PFAS) are a class of thousands of synthetic compounds that have been widely used worldwide since the 1940s due to their ability to repel both grease and water, and to their stability towards chemical and microbial degradation (Glüge et al., 2022). The last property, which is due to the strong C-F bond, has also had the consequence that PFAS are very persistent when released into the environment.

The PFAS chemical group contains a long variety of - chemical compounds like perfluoroalkyl carboxylates (PFCAs), perfluoroalkyl sulfonates (PFSAs), fluorotelomer alcohols (FTOHs), perfluorooctane sulfonamides (FOSAs) and perfluorooctane sulfonamidoethanols (FOSEs) that have been investigated for the past 20 years. According to their functional groups, PFAS are defined as ionic and neutral. Ionic PFAS contain an acidic moiety that can dissociate in the environment, where these compounds are typically present in their anionic form. PFCAs and PFSAs belong to this group. Neutral PFAS containing for example hydroxyl and sulfonamide groups, such as FTOHs, FOSAs and FOSEs, do not dissociate in the environment.

Following the ban and/or restriction in use and production of several PFAS (e.g. PFOS and other long-chain PFAS), the manufacturing of PFAS has shifted to compounds with fewer fluorinated carbons, and perfluoroether-based substances. Although these novel PFAS are considered less bioaccumulative than the legacy PFAS (e.g. PFOS and PFOA), they are still persistent in the environment as their predecessors (Kjølholt et al., 2015).

Currently, 5000 identified PFASs have been registered in the global market (Glüge et al., 2022) and the known PFAS are considered to represent a small part (2-44%) of total anionic extractable fluorine (Yeung et al., 2013).

The aim of this short review is to summarize the sources and transport paths of PFAS in the atmosphere and their deposition. Data from the literature are mainly extracted from studies carried out in Europe and North America, where production and use of PFAS are similar to those in Denmark.

2 Sources and transport pathways for PFAS in the atmosphere

PFAS are introduced into the atmosphere from a large number of sources and via several mechanisms depending on their physical and chemical properties. They can be directly emitted from manufacturing facilities, landfills, wastewater treatment plants, use of firefighting foams, industrial applications, and PFAS-treated household and personal care products. Once in the atmosphere, PFAS undergo transport in both the gas phase and particle phase, resulting in PFAS deposition and contamination in remote regions far from the sources, as e.g. it has been observed in the Arctic regions for the past 20 years (Muir et al., 2019; Wong et al. 2021).

Neutral PFAS such as FTOHs, FOSA and FOSE are volatile and are present mainly in the gas phase. Once in the atmosphere the neutral precursors undergo oxidation resulting in formation of more polar ionic compounds (PFCAs and PFSAs). Atmospheric residence time for FTOH is between 50 and 70 days for photodegradation to the corresponding ionized PFAS (Ellis et al., 2004). Ionic PFAS are characterized by low volatility and higher water solubility than the neutral precursors and in the atmosphere are mostly bound to particle phase.

The long-range atmospheric transport of PFAS is an important process to affect their global distribution. Ionizable PFAS are water soluble and can therefore be transported along rivers and ocean currents but also bound to atmospheric particles as already mentioned. Enrichment of ionizable PFAS in sea spray aerosols has been recently considered as an additional long-range transport pathway (Sha et al., 2021). PFAS concentrations in the microlayer of the sea surface can be 4-5 orders of magnitude higher than in the bulk water (Johansson et al., 2019). PFAS can be transferred to the atmosphere via emission of sea spray and then transported over long distances.

PFAS in the atmosphere are delivered to aquatic and terrestrial ecosystems via wet and dry deposition, with wet deposition as the main process. In the absence of direct sources, atmospheric deposition is the dominant process for contamination of terrestrial and aquatic ecosystems. Wet deposition occurs with precipitation, while dry deposition is determined by surface resistance, quasi laminar resistance and aerodynamic resistance. Dust (particles larger than 10 μm) is removed by gravity whereas smaller particles are removed by the processes described above.

3 Sampling of PFAS in air and precipitation

The most common method used to collect both gas and particulate phase in the atmosphere is active sampling at high or medium flow rate using quartz fiber filters with a combination of polyurethane foam plugs (PUF) and resins (XAD). Typically, the sampling volume is in the range 300-2500 m^3 for outdoor samples. Low volume sampling using solid phase extraction (SPE) cartridges has also been employed for collection of airborne PFAS in areas with high PFAS concentrations.

Passive samplers consisting of polyurethane foam (PUF) disks and PUF disks impregnated with resins (SIP-PUF) are used for long-term monitoring of airborne PFAS at sites where active sampling is not possible. Passive samplers are deployed for at least three months, and the derived concentrations are time-integrated over the entire period. To derive atmospheric concentrations, passive samplers must be carefully calibrated to determine the uptake rate but the derived concentrations are in general connected with larger uncertainty than the more common method using active sampling (Ahrens et al., 2013).

Sampling of PFAS in precipitation is usually carried out with wet-only samplers that automatically open only during precipitation events. Wet only samplers can be operated if electrical power is available. Automatic samplers are also available for separate collection of wet and dry deposition. Bulk samplers consist of open buckets that collects both wet and dry deposition and can be operated without electrical power.

To detect airborne PFAS at ppq (pg/m^3) range in the atmosphere and at ng/L levels in precipitation, extensive quality assurance measures must be undertaken during sampling and shipping of samples to the laboratory since contamination of PFAS may occur during the different operations.

4 PFAS in the atmosphere

Both ionic and neutral PFAS are present in the atmosphere. Concentrations of neutral PFAS are usually much higher than those of ionic PFAS. Neutral PFAS include FTOHs, FOSA and FOSE, while ionic PFAS include PFCAs (e.g. PFOA), PFSAs (e.g. PFOS), and the novel PFAS (e.g, HFPO-DA with trade name Gen-X). Neutral PFAS are characterized by different vapor pressures and thus they are partitioned between gas and particulate phase depending on the temperature. In general, field measurements of PFAS confirm that partitioning from the gas phase to the particulate phase is more favorable with increasing carbon atom chain length and decreasing temperature. Ionic PFAS are expected to be present in their non-volatile anionic form in particle phase.

Ionic and neutral PFAS concentrations in the atmosphere found in selected studies are summarized in Table 4.1. Most of the studies on airborne PFAS investigate separately ionic and neutral PFAS. The analysis of the two groups of compounds requires the use of gas chromatography (GC) for the neutral PFAS and liquid chromatography (LC) for the ionic PFAS and in both cases with mass spectrometry (MS) detection. FOSA and FOSE can be analyzed with both GC and LC and their concentrations are sometimes reported together with the ionic PFAS. Wang et al., (2022a) applied a common extraction method followed by fractionation to GC and LC analysis for passive samplers (SIP-PUF). Müller et al. (2012) and Dreyer et al. (2009b) reported the concentrations of both neutral and ionic PFAS using a high-volume sampler. They analyzed a fraction of the filter for ionic PFAS, and the remaining filter and PUF/XAD for the neutral PFAS.

Among the neutral PFAS, 8:2 FTOH is the most abundant compound found in the atmosphere followed by 6:2 FTOH and then FTOH 10:2. PFCAs and PFOS are the most common compounds found in particulate phase (Faust, 2023). However, short-chain PFAS were found to be dominant in India, Japan and South Korea in a recent study carried out in Asiatic countries (Lin et al., 2020). Emerging PFAS such as HFPO-DA, which is a common PFOA replacement, are beginning to be detected at significant concentrations, as e.g. in Japan, accounting for 27% of all quantified PFAS (Lin et al., 2020).

Generally, airborne PFAS exhibit a decreasing gradient from cities with high population density towards less industrialized sites (Wang et al., 2022a). Muller et al. (2012) found that background concentrations of PFAS were a factor of five lower than urban concentrations in Zurich.

Dreyer et al. (2009b), showed that neutral PFAS are moderately correlated with temperature and that higher concentrations were observed in summer, based on an annual atmospheric concentration study. Wang et al. (2022b) found that ionic PFAS exhibited significant seasonal variations, which were higher in summer. 8:2 FTOH and 10:2 FTOH concentrations were positively correlated with PFOA and PFDoA concentrations, suggesting that PFOA and PFDoA were probably the photodegradation products of FTOHs confirming the results from laboratory experiments.

Table 4.1. Summary of literature for PFAS concentrations in the atmosphere.

Location (number of sites) and type	Sampling	Neutral PFAS pg/m^3 (min-max)	Ionic PFAS pg/m^3 (min-max)	Sampling period	Reference
China (23)- urban	SIP-PUF	4.8-1400	3.7-330	July 2019-May 2020	Wang (2022a)
Norway (2)- coastal	HVS	n.a.	0.004-3.4	2018-2020	Sha (2022)
Czech Republic (1)- rural	HVS	n.a.	0.03-2.08*	2012- 2014	Paragot (2020)
Germany and Austria (2) -Alpine	LVS	3.6-109	n.a.	2007-2010	Xu (2017)
Büsum, Germany (1)-coastal	HVS	8.6-155	n.a.	2011-2012	Wang (2014)
Zurich, Switzerland (2) – urban/remote	HVS	0.6-1200	0.14-7.7	August 2010	Müller (2012)
Hamburg, Germany (2) - semirural	HVS	40-244	0.8-11	October-Novem. 2007	Dreyer (2009a)
German Bight (1)- marine (ship)	HVS	4.4-224	3.2-6.4	October-Novem. 2007	Dreyer (2009a)
Hamburg, Germany (2)- semirural	HVS	3.3-143	0.5-3.6	2007-2008	Dreyer (2009b)
Greenland (1)- Arctic remote	HVS	0.06-22.4	n.a.	2008-2015	Skov (2017)

HVS= high volume sampling with filter, PUF and XAD
LVS = low volume sampling with solid phase cartridge (SPE)
n.a. = not analyzed
* FOSAs and FOSEs included

5 PFAS in precipitation

PFAS have been detected in precipitation from urban, rural, and remote sites around the world. Most studies have focused on the "legacy PFAS", i.e., PFOS and PFOA, and those compounds with carbon chain equal to or larger than eight. With the phasing out and/or restrictions of PFOS-and PFOA-based production, the focus has shifted on the shorter-chain compounds and PFAS of other functional classes such as HFPO-DA.

Most of the data published in the literature are obtained by sampling wet deposition and very few using bulk deposition. A few studies have investigated the contribution of wet and dry deposition, respectively, to the total deposition of PFAS. The main conclusion from these studies is that wet deposition removes PFAS from the atmosphere an order of magnitude more effectively than dry deposition (Shimizu et al., 2021). PFAS concentrations found in precipitation from selected studies are summarized in Table 5.1.

Generally, PFCAs with carbon chain from 4 to 10 are the compounds found with highest detection frequency. PFOS is the most frequently detected perfluorosulfonic acid (PFSA) with frequency greatly exceeding the second most detected PFSA, namely PFHxS. Perfluorotelomer sulfonates (FTSA) have been sporadically found and their presence in precipitation has been associated to specific sources such as the use of certain firefighting foams, and industrial processes such as chrome plating (Pfotenhauer et al., 2022). HFPO-DA, an emerging PFAS, has also been detected in precipitation in a recent study in USA (Pike et al., 2021) at concentrations between 0.8 and 1.0 ng/L.

Short chain PFAS with carbon number below eight are recently found to be dominating the total PFCAs concentrations in North America (Pfotenhauer et al., 2022; Pike et al., 2021; Gewurtz et al., 2019) with PFBA and PFHxA as the dominating compounds, as a probable result of the US EPA's 2006 Stewardship Program on reduction of emission of PFOA and related chemicals (US EPA, 2006).

Total PFAS concentrations as reported by e.g., Gewurtz et al. (2019), Pfotenhauer et al., 2022, and Pike et al., (2021) (Table 5.1) fall into the ranges reported for rural areas away from any direct sources such as manufacturers or major users of PFAS. As observed by Gewurtz et al. (2019) in the Canadian Great Lakes region, PFOS and PFOA concentrations in precipitation were higher at urbanized sites compared to more remote sites, indicating the effect of local anthropogenic sources. On the other side, concentrations of the shorter chain PFAS (e.g. PFBA) were comparable among the different sites, indicating that these compounds are evenly distributed in the global atmosphere. Although production of PFOA and PFOS has largely been replaced by the production of shorter chain compounds or other sub-classes of PFAS compounds, the frequent presence of legacy PFAS in precipitation is due to either precursors transformation or continuing emission from historic reservoirs or fugitive emissions from industrialized sites close to urban environments.

Data for PFAS in precipitation in Denmark are available from Strand et al. (2007). The data are generated during a multi-compartment screening for PFAS in the Danish environment carried out in 2004. The samples analyzed were taken at eight background stations for the National Monitoring

Programme for the aquatic and terrestrial environment (part of NOVANA). Of the seven PFAS analyzed, four (PFHxS, PFOS, PFDA and PFUnA) were found at concentrations above the detection limit with ΣPFAS in the range 1.6-38.3 ng/L. The compounds found with the highest concentrations were PFDA and PFUnA. These concentrations are in the same range found at other rural and semi-rural sites, as e.g. in Northern Germany (Dreyer et al., 2010) and the Great Lakes Region (Gewurtz et al., 2019). However, these data are only indicative as a part of a preliminary screening performed with samples already collected for other purposes and not following a strict protocol for specific sampling of PFAS. Moreover, the total PFAS concentrations at the Danish sites might be underestimated if compared to more recent studies, as more compounds are included in the analytical program of the cited studies.

Table 5.1. Summary of literature for PFAS concentrations in precipitation.

p	Sampling	Number of PFAS analyzed (most frequently found)	Total PFAS min-max conc. in ng/L	Sampling period	Reference
Wisconsin, USA (8) - rural	Wet only	34 (22)	0.7-6.1	April-November 2020	Pfotenhauer (2022)
Chongqing, China (1)- urban	Wet only	17 (13)	0.9-191.3	July 2020-April 2021	Wang (2022)
Ohio-Indiana, USA (6)- rural/urban	Wet only	15* (10)	50-850**	Summer 2019	Pike (2021)
Great Lakes, Canada (3)- urban/rural/remote	Wet only	12 (10)	0.26-14.0	2006-2018	Gewurtz (2019)
Råö, Sweden (1)- remote	Bulk	16 (8)	2.18-5.88	2015	Johansson (2018)
Stockholm, Sweden (1)- urban	Bulk	16 (8)	0.95-6.39	2015-2016	Johansson (2018)
Krycklan, Sweden (1)- remote	Bulk	10 (9)	0.90-3.62	May-November 2011	Filipovic (2015)
Barsbüttel, Germany (1) – semi-rural	Bulk with filter	37 (17)	1.6-48.6	2007-2008	Dreyer (2010)
Denmark (8)- rural	Bulk	7 (4)	1.6-38.3	October 2004	Strand (2007)

*Trifluoroacetic acid (TFA) included in the analysis
** TFA contribution ~ 90% of total PFAS

Differences in concentration and concentration profile of PFAS at different sites may provide indication of local sources versus the general background concentrations. Wang et al. (2022b) used positive matrix factorization (PMF) to apportion the sources of PFAS in precipitation at an urban site in southwest China and found that, on a yearly average, the usage of aqueous film forming foams (AFFF) and paper packaging production were two major sources of PFAS to wet and dry deposition. Pike et al., (2021) applied ANOVA modeling and correlation matrices to a dataset of PFAS concentrations from six rural and urban sites in Wyoming, USA. They identified local point sources as an important contributor for the PFAS profiles at the investigated locations, suggesting that mitigation of local sources could lower the deposition fluxes of PFAS.

6 Concentrations of PFAS in the atmosphere in Northern Greenland

Neutral PFAS have been measured in the atmosphere at Villum Research Station at Station Nord in North Greenland (Villum) since 2008 for the Arctic Monitoring and Assessment Program (AMAP).

Atmospheric concentrations (sum of gas phase and particulate phase) of the analysed PFASs in the period 2008-2015 are summarized in Table 6.1.

Table 6.1. Average, median and concentration ranges (plus minus 1 std.dev. of weekly measurements, pg m^{-3}) of neutral PFASs (pg m^{-3}) in years 2008-2015 (sum of gaseous and particulate phase). Samples below the detection limit were assigned half of the detection limit value (0.05-0.25 pg/m^3). The values are given only for those compounds detected in more than 30 % of the samples.

Compound	Average	Median	Range
6:2 FTOH	2.73	1.93	0.23-16.5
8:2 FTOH	4.48	3.37	0.23-22.4
10:2 FTOH	1.32	0.67	0.10-9.68
N-Me-FOSA	0.34	0.22	0.10-3.41
N-Et-FOSA	0.30	0.22	0.11-1.93
N-Me-FOSE	0.48	0.28	0.07-7.46
N-Et-FOSE	0.57	0.22	0.06-5.96

The average yearly sum of the seven measured neutral PFAS ($\sum_7$PFAS) in the atmosphere at Villum ranged from 1.82 to 32.1 pg m^{-3}. The most abundant compound was 8:2 FTOH (44% of $\sum_7$PFAS), followed by 6:2 FTOH (25% of $\sum_7$PFAS) and 10:2 FTOH (14% of $\sum_7$PFAS). Among the FOSA/FOSE, the dominant compound was N-Me-FOSE (average: 0.61 pg m^{-3}) followed by N-Et-FOSE (average: 0.50 pg m^{-3}), N-Me-FOSA (average: 0.44 pg m^{-3}) and N-Et-FOSA (0.33 pg m^{-3}). The concentrations of 6:2 FTOH measured in this study were in the same range as those reported by other Arctic studies at Alert and at Ny Ålesund.

Sources of PFAS in the atmosphere in Denmark

In Denmark there is no large-scale manufacturing of perfluorochemicals that can represent a considerable emission of PFAS to the atmosphere. Medium- and long-range transport is expected to be the main source for PFAS in the atmosphere, as e.g. observed by Dreyer et al. (2009a and 2009b) at the Northern coast of Germany (German Bight) and Hamburg. However, there are several industrial activities that can contribute to atmospheric emission of PFAS (diffusive local sources) as reported by Nicolajsen and Tsitonaki (2016).

Diffusive local sources can be represented by wastewater treatment plants (WWTP) and landfills. Water to air transport of PFAS from WTTP tanks is enhanced by the turbulence caused by the aeration processes. Emissions from landfills are likely associated with waste containing surfactants material with small percentages of unbound residual FTOHs, FOSAs and FOSEs (Ahrens et al. 2009).

Incineration of solid wastes might be a potential source of atmospheric PFAS. Several studies performed at controlled conditions have demonstrated that temperatures of 1000 ^{o}C and higher does not release the most studied PFAS such as PFOA or PFOS (Stoiber et al., 2020). However, it is not yet clear how

the different classes of PFAS (precursors included) are degraded under different thermal conditions Liu et al., 2021).

Marine aerosols enriched with PFAS from sea spray could also be a source of transport of atmospheric PFAS back to land, particularly in the case of the west coast where the dominant wind direction is from the sea (West) towards the land.

7 Conclusions

Generally, the presence of PFAS in atmosphere and wet/dry deposition is associated with proximity to urban or industrial locations, where emissions occur from diffuse sources and industrial processes. Meteorological parameters are generally not correlated with PFAS concentrations in the atmosphere. The negative correlation with temperature observed in several studies supports the hypothesis that PFAS levels in the atmosphere are mostly determined by advection and long-range atmospheric transport.

Taking into consideration the published literature on PFAS in rainwater, the concentration of the four PFAS included in the recently proposed guidelines for drinking water (PFOA, PFOS, PFHxS and PFNA) often exceed these levels (Cousins et al., 2022). The Σ_4-PFAS limit of 2 ng/L recently established by the Danish EPA (Miljøministeriet, 2021) has been exceeded in most of the wet deposition levels collected at various worldwide locations since 2010 (Cousins et al., 2022). The global distribution of PFAS observed in the environment (soil and aquatic ecosystems) is a result of the ubiquitous presence of PFAS in atmospheric deposition.

Although production of PFOS and PFOA has largely been replaced by the production of shorter chain compounds or other sub-classes of PFAS containing ether bonds (as e.g., HFPO-DA = Gen X), these legacy compounds are still found in precipitation and atmosphere. Their occurrence might be due to transformation of precursor compounds and/or re-emission from historic reservoirs.

Long-term studies of PFAS in precipitation (Gewurtz et al., 2019) have shown that legacy PFAS concentrations have decreased over time, likely in response to phase-out and regulatory action from the North American and European authorities. Shorter chain PFAS such as PFBA and PFHxA appear to be increasing in the most recent years, since these compounds are being used as replacement chemicals for PFOS, PFOA, and the longer chain PFCAs.

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