



# Stockholm Arlanda Airport as a source of per- and polyfluoroalkyl substances to water, sediment and fish



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## ABSTRACT

Fire training facilities are potential sources of per- and polyfluoroalkyl substances (PFASs) to the nearby environment due to the usage of PFAS-containing aqueous fire-fighting foams (AFFFs). The multimedia distribution of perfluoroalkyl carboxylates (PFCAs), perfluoroalkyl sulfonates (PFSAs), perfluorooctane-sulfonamide (PFOSA) and 6:2 fluorotelomer sulfonate (FTSA) was investigated near a fire training facility at Stockholm Arlanda Airport in Sweden. The whole body burden of PFASs in European perch (*Perca fluviatilis*) was  $334 \pm 80 \mu\text{g}$  absolute and was distributed as follows: Gonad > liver  $\approx$  muscle > blood > gill. The bioconcentration factor (BCF) and sediment/water partition coefficient ( $K_d$ ) increased by 0.6–1.7 and 0.2–0.5 log units, respectively, for each additional  $\text{CF}_2$  moiety for PFCAs and PFSAs. PFAS concentrations in water showed no significant decreasing trend between 2009 and 2013 ( $p > 0.05$ ), which indicates that Stockholm Arlanda Airport may be an important source for long-term contamination of the nearby environment with PFASs.

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## 1. Introduction

Aqueous fire-fighting foams (AFFFs) are concentrated liquids designed to extinguish hydrocarbon fuel fires and are used by commercial, public, and military fire-fighting organizations (Paul et al., 2009). Since the 1960s, per- and polyfluoroalkyl substances (PFASs) have been used in AFFFs due to their surface active characteristics to enable the formation of an aqueous film and to resist heat, oil, and water. PFASs have received increasing public attention due to their persistence, bioaccumulative potential, and possible adverse effects on humans and wildlife. As a consequence, perfluorooctane sulfonate (PFOS) and its precursors have been added to the persistent organic pollutants (POPs) list of the Stockholm Convention in May 2009, resulting in global restrictions on its use and production (Paul et al., 2009). After the voluntary phase-out or ban of the  $\text{C}_8$ -based PFASs, such as PFOS, the production shifted to PFAS precursors, due to their higher degradation potential, and to short-chain PFASs (perfluoroalkyl chain length of  $\text{C} < 8$ ), due to their lower bioaccumulation potential (Martin et al., 2003; Möller et al., 2010; Ahrens, 2011). However, PFAS precursors can be (bio)degraded under aerobic and anaerobic conditions to long- and short-chain perfluoroalkyl carboxylates (PFCAs) and perfluoroalkane sulfonates (PFSAs) which are the final

degradation products and extremely persistent in the environment (Rhoads et al., 2008; Wang et al., 2011).

Previous studies have linked the usage of AFFFs at fire training facilities to the contamination of the environment with PFASs (Moody et al., 2002, 2003; Awad et al., 2011; Kärrman et al., 2011; De Solla et al., 2012). PFOS and 6:2 fluorotelomer sulfonate (FTSA) were the dominant PFASs detected at AFFF contaminated sites with maximum concentrations in the  $\text{mg L}^{-1}$  and  $\mu\text{g g}^{-1}$  range for water and fish, respectively (Moody et al., 2003; Awad et al., 2011; Kärrman et al., 2011; De Solla et al., 2012). Once released into the environment, PFASs are subject to various partitioning, degradation, and transport processes depending on their physico-chemical properties and environmental conditions (Ahrens, 2011). The short-chain PFSAs and PFCAs are potentially more water soluble, whereas long-chain PFSAs and PFCAs seem to bind more strongly to particles and accumulate in the food chain (Higgins and Luthy, 2006; Ahrens et al., 2010). However, there is a lack of knowledge of the multimedia distribution of PFASs in the environment. AFFFs are a particularly problematic source of PFASs since high amounts are used in liquid form during a relatively short time period, which increases the potential for the PFASs to be released into the aqueous environment.

The aim of this study was to examine the transport and fate of PFASs in water, sediment and fish near a fire training facility at Stockholm Arlanda Airport. The specific objectives were to (i) determine the concentrations and composition profile of various

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PFASs in the multimedia environment (i.e., water, sediment and fish), (ii) to calculate fish bioconcentration factors (BCF) and the sediment/water partition coefficient ( $K_d$ ) of individual PFASs, and (iii) to investigate the spatial distribution and temporal trends of PFASs in a lake near the fire training facility.

## 2. Materials and methods

### 2.1. Sampling sites

The study was carried out in an area surrounding Stockholm Arlanda Airport, which is the largest airport in Sweden. Stockholm Arlanda Airport is an international airport located ~37 km north-northwest of Stockholm and is used by approximately 19 million people annually (2012). There is a fire training facility located in the north part of the airport area (Fig. 1) where PFOS-containing AFFFs (STHMEX-AFFF 3%, Dr. Richard Sthamer GmbH & Co. KG, Hamburg, Germany) were used frequently from the 1980s until 2003. From 2003, remaining stocks of STHMEX-AFFF 3% were used and then replaced by PFOS-free AFFFs (Presto AFFF and Moussol APS-P, Dr. Richard Sthamer GmbH & Co. KG, Hamburg, Germany); however, these PFOS-free AFFFs still contained other PFASs. According to the manufacturer, the PFOS-free AFFFs contain <10% of PFASs, but further details about the AFFF products are not available. In 2011, Stockholm Arlanda Airport stopped purchasing PFAS-containing AFFFs and a fluorine-free (FF)-AFFF (Moussol FF 3/6, Dr. Richard Sthamer GmbH & Co. KG, Hamburg, Germany) is used instead.

### 2.2. Sampling

Surface water ( $n = 33$ ), sediment ( $n = 3$ ) and fish ( $n = 21$ , European perch (*Perca fluviatilis*)) samples were collected at Lake Halmsjön nearby Stockholm Arlanda Airport between 2009 and 2013 (Fig. 1, and Supplementary Tables S1, S2, S3). Perch tissue samples included muscle tissue ( $n = 18$ ), and blood, liver, muscle, gill and gonad tissues from three individual fish. In addition to the sampling at Lake Halmsjön (sampling site 13,  $n = 13$ , 2009–2013), surface water samples were collected at two reference lakes (sampling sites 1 and 2,  $n = 2$ , 2009), artificial ditches downstream of the fire training facility (sampling sites 5–10,  $n = 6$ , 2011), and various streams in the area surrounding Stockholm Arlanda Airport (sampling sites 3, 4, 11, 12, 14–18,  $n = 12$ , 2011) (for details see Supplementary information).

### 2.3. Chemicals

The target analytes included  $C_6$ – $C_{11}$  PFCAs ( $C_nF_{2n+1}COO^-$ , PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA),  $C_6$ ,  $C_8$ ,  $C_{10}$  PFSA ( $C_nF_{2n+1}SO_3^-$ ,

PFHxS, PFOS, PFDS), perfluorooctanesulfonamide (PFOSA,  $C_8F_{17}SO_2NH_2$ ) and 6:2 FTSA ( $C_6F_{13}CH_2CH_2SO_3^-$ ) plus [ $^{13}C_4$ ]-PFOS and [ $^{13}C_4$ ]-PFOA as mass-labeled internal standards (IS).

### 2.4. Analysis

The water samples were extracted by solid phase extraction (SPE) according to the ISO/DIS 25101:2009 method using Oasis WAX cartridges (Waters, 150 mg, 6 mL, 30  $\mu m$ ) (ISO/DIS25101, 2008). The fish tissue and unsieved sediment samples were extracted based on the solid-liquid extraction method described by Powley et al. (2005). The separation and detection of PFASs were performed by high-performance liquid chromatography (HPLC, UFLC Shimadzu, model CBN-20A, Japan) coupled to a mass spectrometer interfaced with an electrospray ionization source in a negative-ion mode (ESI-MS/MS, API 4000, AB Sciex, Foster City, CA, USA). The isotope dilution method was used for quantification. As standard procedure, laboratory blanks, method detection limits (MDLs), and recoveries were examined. Blank concentrations were <1% of the concentrations measured in the samples. MDLs were in the range of tens to hundreds of picograms per liter and grams, respectively. The recoveries in the sample were typically in the range of 70–110% (for details see Supplementary information).

## 3. Results and discussion

### 3.1. PFAS composition profiles in water, sediment and fish

All 11 target PFASs were detected and quantified in water, sediment and perch tissues (i.e. blood, liver, muscle, gill, gonad). The composition profiles PFASs varied between compartments in Lake Halmsjön (Supplementary Fig. S1). In water, PFOS (~36%), PFHxS (~29%), and PFOA (~20%) were the dominant PFASs, followed by PFHxA (~8%). PFOS was even more predominant in sediment and perch tissues with a contribution of ~86% and ~99%, respectively. This indicates a compound-specific distribution of PFASs in the multi-compartment environment (Martin et al., 2003; Higgins and Luthy, 2006; Ahrens, 2011).

### 3.2. Tissue distribution of PFASs in perch

PFASs were determined in blood, liver, muscle, gill and gonad tissues in perch from Lake Halmsjön. No significant decreasing trend was observed for the PFAS concentrations in muscle tissues between 2009 and 2012 ( $p > 0.05$ , ANOVA), and therefore they are discussed as average values in the following. PFOS was the predominant PFAS in all tissue samples. The highest average  $\Sigma$ PFAS concentrations were detected in blood with  $5900 \pm 900$  ng g $^{-1}$  ww ( $n = 3$ ), followed by liver ( $3900 \pm 500$  ng g $^{-1}$  ww,  $n = 3$ ), gill

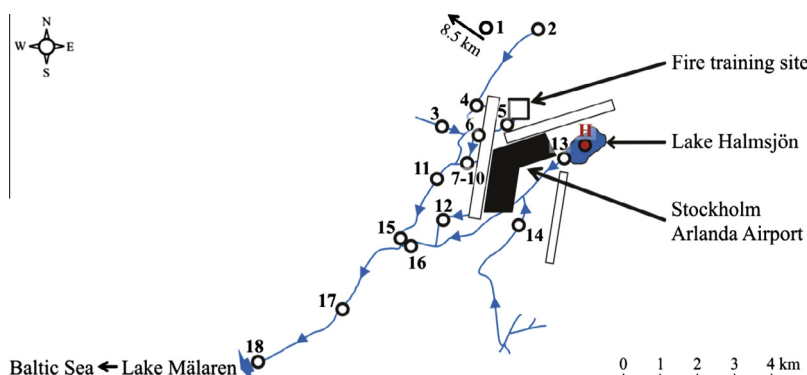


Fig. 1. Sampling sites for water sampling (1–18) and sampling of water, sediment and European perch (*Perca fluviatilis*) (H) at Stockholm Arlanda Airport, Sweden.

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