

# Risks and Effects of the dispersion of PFAS on Aquatic, Terrestrial and Human populations in the vicinity of International Airports

Final report of the RE-PATH project 2009-2014



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## Summary

Due to the historical usage of aqueous film forming foams (AFFF) which contained per- and polyfluorinated alkylated substances (PFAS) at fire drills at Göteborg Landvetter Airport and Stockholm Arlanda Airport, elevated PFAS concentrations were found in surface waters and fish in the vicinity of these airports. As a result of these findings, Swedavia Swedish Airport and IVL Swedish Environmental Research Institute Ltd IVL initiated the co-financed project *Risks and Effects of the dispersion of PFAS on Aquatic, Terrestrial and Human Populations in the vicinity of international airports* (RE-PATH) to study the long-term consequences of the releases of PFAS from firefighting training sites.

Since the project start in 2009, about 700 samples have been collected and analysed for their PFAS content. Toxicity tests have been performed to investigate potential influence of PFAS on reproduction, immobility and hatching frequency. The sediment-water balance has been studied to elucidate whether the PFAS accumulated in sediments contributes substantially to the water concentrations. The bioaccumulation and depletion potential was studied in goldfish, zebrafish and in crayfish. Furthermore, the load of perfluorooctane sulfonate (PFOS) to Lake Mälaren was estimated and compared to releases via other sources. A mass balance model was developed to investigate the distribution and the rate of decline of PFOS-levels in the environment around Stockholm Arlanda Airport.

In this final report, the results from 2009 to 2014 are summarised. More details on methods and results are given in individual reports and publications (Woldegiorgis et al., 2010, Norström et al., 2011, Norström& Viktor, 2012, Norström et al., 2013, Ahrens et al., 2015).

The main conclusions from the project are:

- Firefighting training sites are significant point sources contributing to the environmental contamination of PFAS in Sweden.
- Individual firefighting training sites contribute only by a small fraction of the total load of PFAS to the Swedish environment, but can locally cause elevated concentrations of PFOS in fish and water by up to a factor of 100, in comparison to reference areas.
- The current contribution of PFOS and PFAS from Stockholm Arlanda Airport to Lake Mälaren is 1.3 kg and 2.4 kg/year respectively, and 0.9 and 1.3 kg/year respectively from Göteborg Landvetter Airport (reference year 2013).
- No risk for human health effects caused by intake of PFOS via water or fish consumption has been identified in the studied areas, using the current TDI level (150 ng/kg body weight).
- No ecotoxicological effects on local fish (perch) have been observed, at the prevailing conditions in the field.
- The concentrations of PFOS in the local environment surrounding Stockholm Arlanda Airport are expected to decline slowly. If no measures are taken, background levels will not be reached in the near future
- A risk for secondary poisoning of species higher up in the food chain such as fish eating species cannot be excluded

One of the major concerns of PFAS is their extreme persistence combined with their specific binding properties. This has resulted in contamination of groundwater in the vicinity of firefighting areas at several locations in Sweden. In some of these areas, where this groundwater is used as the main source for drinking water, elevated concentrations have been found in the human population, especially in young

children. In the case of Arlanda and Landvetter there are no large public drinking water sources in the close proximity to the contaminated areas, and the few private groundwater wells that are located near the two airports have been controlled, showing only background levels. The nearest public source of drinking water in the Arlanda area is Görvelns drinking water plant, located in north Mälaren. Although significant amounts of PFAS enter Lake Mälaren from multiple sources each year, the concentrations reported by Görvelns drinking water plant are similar to background levels for drinking water.

At Arlanda and Landvetter, consumption of local fish would be the main exposure for humans. The Swedish National Food Agency has estimated average daily intake (ADI) of PFOS for an adult person eating a balanced diet and drinking water to 0.6 ng PFOS/kg body weight, based on typical levels of PFOS in drinking water and food items. The current value for tolerable daily intake (TDI) for PFOS is 150 ng/kg body weight and day (EFSA 2008). Consumption of fish from Lake Halmsjön or Lake Västra Ingsjön once a week increases the ADI to 94 and 13 ng PFOS/ kg bw and day, respectively. In combination with contaminated drinking water, the TDI may be exceeded, as has been shown in highly contaminated areas. In these areas, exposure to drinking water alone may result in ADIs exceeding the TDI several times, as in the case of formula fed babies. Even though the fish from the contaminated areas in this study can be consumed with respect to PFAS, it is not recommended to eat freshwater fish too often due to high concentrations of mercury.

It has been shown in other studies that predators feeding almost exclusively on fish might be at risk since they feed on the whole fish which may contain several hundreds of micrograms of PFOS and due to biomagnification. Because of the high levels of PFOS in the fish in Lake Halmsjön, long-term effects on top predators cannot be excluded, although the current health status of the fish appears to be good. Other studies have indicated that a high body burden in fish in relation to the total body weight may have an effect on the physiological functions of the fish in the longer term.

The contribution from local sites to the large scale contamination of Swedish ecosystems by PFOS may be small, but the local impacts have been shown to be serious in several areas in Sweden. Because of this, mapping of contaminated areas as well as their surroundings is needed, and there is a need to establish site-specific limit values for soil and groundwater depending on local conditions. It is of utmost importance to ensure that existing contaminated areas do not result in damages to health and environment now, or in the future. A consistent regulatory practice, on all potential sources in the society is also important.

Even more important is to learn from experience. The main reason for the problems we see today is the extreme persistence of some of the compounds in the PFAS group, and the fact that most of the non-persistent PFAS degrade to form extremely stable compounds. This, combined with their tendency to be transferred to groundwater makes it extremely challenging to implement cost-effective remediation techniques. In many of the current AFFFs the fluorinated content comprises precursor compounds to perfluorinated alkylacids (PFAA), which are released to soil and groundwater, where they degrade to persistent PFAAs. Thus, just removing the persistent substances themselves from the products does not necessarily prevent environmental contamination of PFAAs from firefighting foams.

In October 2014, several PFAAs were added to the 'SIN list' ([www.chemsec.org](http://www.chemsec.org)), which is a concrete tool aimed to speed up the transition to a world free from hazardous chemicals. The substances included are identified by ChemSec as Substances of Very High Concern based on the criteria established by the EU chemicals regulation REACH. This illustrates that the problem with environmental contamination of PFAS is not limited to PFOS in AFFF, but concerns the entire chemical group, which has multiple application areas.

# Sammanfattning

RE-PATH (*Risks and Effects of the dispersion of PFAS on Aquatic, Terrestrial and Human populations in the vicinity of International Airports*) är ett samfinansierat projekt mellan SIVL och Swedavia. Projektet startade 2009 efter att förhöjda halter av PFAS konstaterats vid både Göteborg Landvetter Airport och Stockholm Arlanda Airport. Syftet med projektet har varit att undersöka effekter och risker med spridningen av PFAS från dessa flygplatser då dessa ämnen har läckt ut från brandövningsplatserna. PFOS har ingått i de filmbildande brandbekämpningsskum (AFFF) som har använts vid brandövningarna sedan början av 1980-talet fram till år 2003 men andra PFAS har funnits i de skum som använts efter 2003..

PFAS är ett samlingsnamn för en stor grupp högfluorerade kemikalier som har använts kommersiellt sedan 1950-talet. Strukturen hos PFAS ger dem ett brett användningsområde och förutom brandskum har dessa ämnen bland annat även använts i impregneringsmedel för textilier, rengöringsprodukter, elektronik, skidvalla m.m. Flera PFAS är extremt svårnedbrytbara och genom sin frekventa närvaro i miljön samt toxiska egenskaper är PFOS och PFOA de mest kända substanserna. PFOS är sedan 2008 förbjuden att använda inom EU, dock finns vissa undantagsområden.

Under åren 2009-2014 har ca 700 prover tagits i områdena kring Göteborg Landvetter Airport och Stockholm Arlanda Airport. Proverna har i huvudsak utgjorts av ytvatten, sediment, fisk samt olika fiskorgan. Även några enstaka fåglar och däggdjur har analyserats. De PFAS som har ingått i projektet är: PFBS, PFHxS, PFOS, PFDS, PFOSA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA och 6:2 FTS. Ett flertal toxicitetstester har också genomförts och faktorer som reproduktion, kläckning och immobilitet har studerats hos lokala arter från de närbelägna sjöarna. Bioackumulering och utsöndringshastigheter hos flera vattenlevande djur har också studerats. Försök med sediment-vatten balans har utförts för att studera sedimentets potential som källa till PFAS-koncentrationen i vattnet och en massbalans modell har utvecklats för att studera spridningen och minskningen av PFAS runt Arlanda flygplats. Arlandas bidrag av PFAS till Mälaren har jämförts med andra källor.

De huvudsakliga slutsatserna från RE-PATH är:

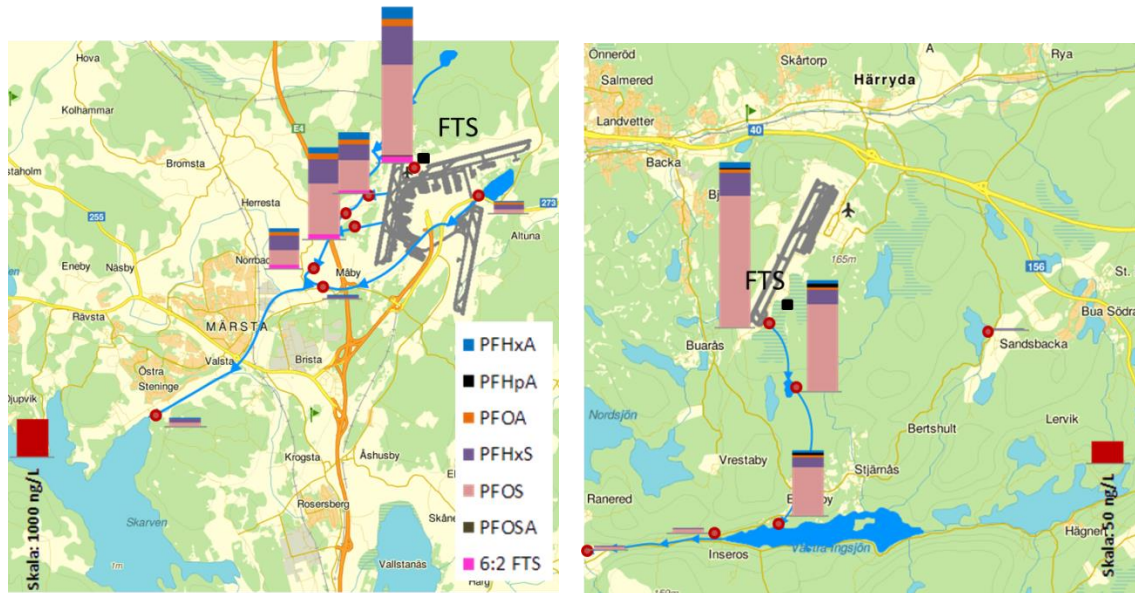
- Brandövningsplatser är viktiga punktkällor av PFAS till miljön i Sverige
- Enskilda brandövningsplatser bidrar till en liten del av den totala belastningen av PFAS till miljön men närbelägna lokala områden kan innehålla upp till 100 ggr högre halter i fisk och vatten jämfört med referensområden
- Stockholm Arlanda Airport bidrar idag med 1.3 kg PFOS (2.4 kg PFAS)/år till Mälaren
- Ingen påvisad risk för människa med avseende på intag av PFOS i närheten av flygplatserna, enligt nuvarande TDI (150 ng/kg kroppsvikt och dag)
- Inga akuta ekotoxikologiska effekter på lokala arter (främst abborre) har kunnat påvisas vid de koncentrationer PFAS som sjöarna utanför de båda flygplatserna innehåller.
- Det kommer att ta lång tid innan PFAS-koncentrationerna i närmiljön kring Stockholm Arlanda Airport har nått halter i samma nivå som referensområden
- Risk för sekundär förgiftning för fiskätande djur kan inte uteslutas

Delårsrapporter från år 2009, 2010, 2011 och 2012 har tidigare publicerats (Woldegiorgis et al., 2010, Norström et al. 2011, Norström och Viktor, 2012, Norström et al., 2013).



## 1.1 Vatten

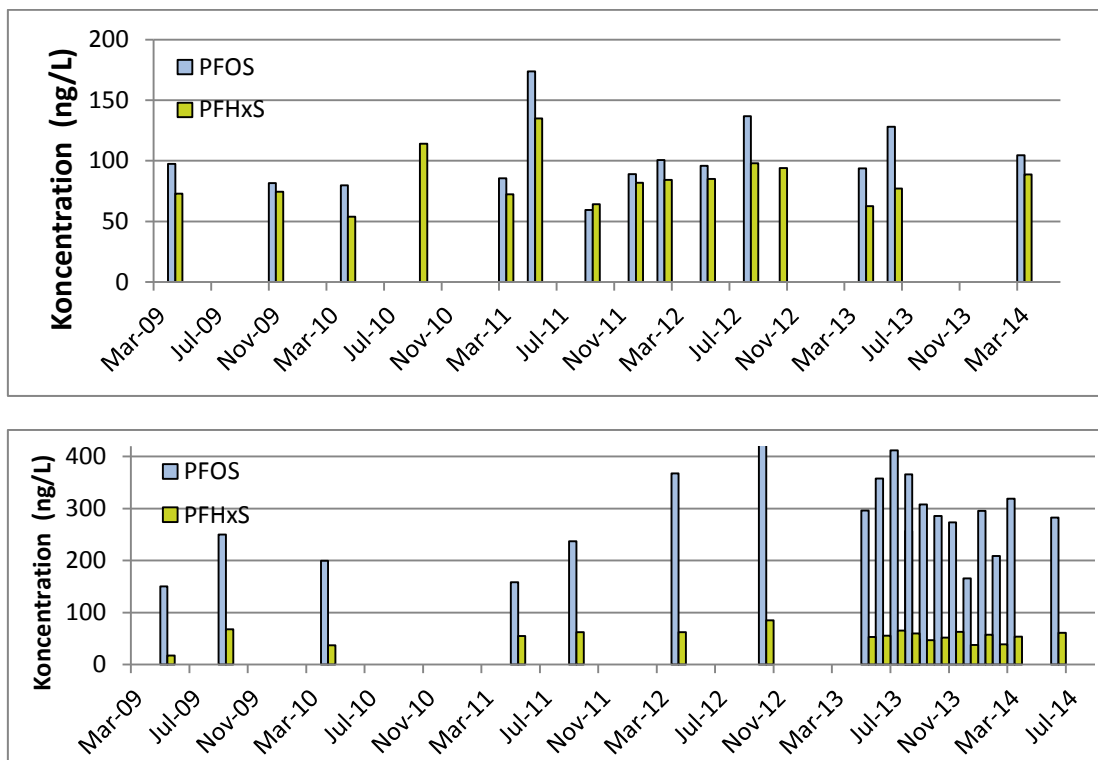
Figur 1.1 visar hur koncentrationerna av PFAS minskar med avståndet till brandövningsplatserna vid respektive flygplats. Koncentrationerna är högst närmast brandövningsplatserna och minskningen som sker med avståndet beror främst på utspädning. Trots minskningen innehåller vattnet som rinner ut i Mälaren ca 100 gånger högre halter jämfört med referenssjön. Vattnet som rinner ut i Västerhavet innehåller ca sex gånger högre halter än referenssjön.



Figur 1.1. Koncentration av PFAS (ng/l) i ytvatten med ökat avstånd från brandövningsplatserna vid Stockholm Arlanda Airport (vänster bild) och Göteborg Landvetter Airport (höger bild). Observera att staplarna har olika skalor.

Punkterna vid Halmsjöns utlopp vid Arlanda flygplats samt alla punkter vid Landvetter flygplats har provtagits två gånger per år sedan 2009. En regressionsanalys som gjorts på halterna från dessa fem år visar att halterna av PFOS inte minskar signifikant under denna period vid någon provtagningspunkt. Dock är fem år ett kort intervall för att studera tidstrender och uppföljande mätningar behövs. Figur 1.2 visar halterna mellan 2009 och 2014 vid Halmsjöns utlopp samt vid utloppet från Landvetters flygplats.

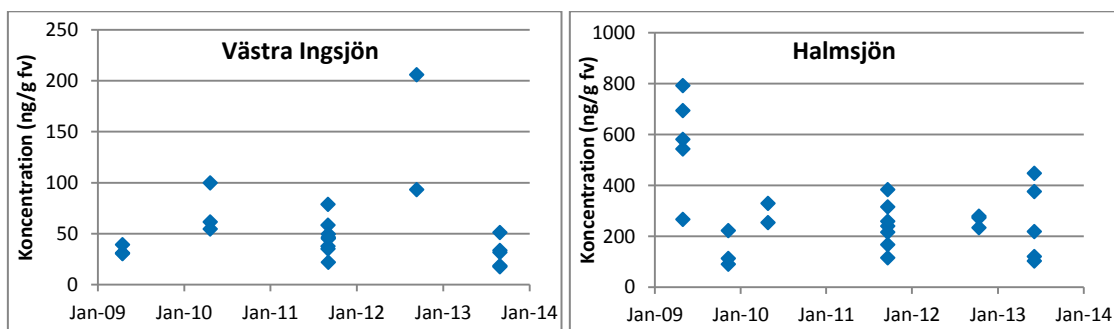




Figur 1.2. Tidstrend av PFOS och PFHxS i ytvatten vid Halmsjöns utlopp (överst) vid Arlanda flygplats och vid utloppet från Landvetters flygplats (nederst).

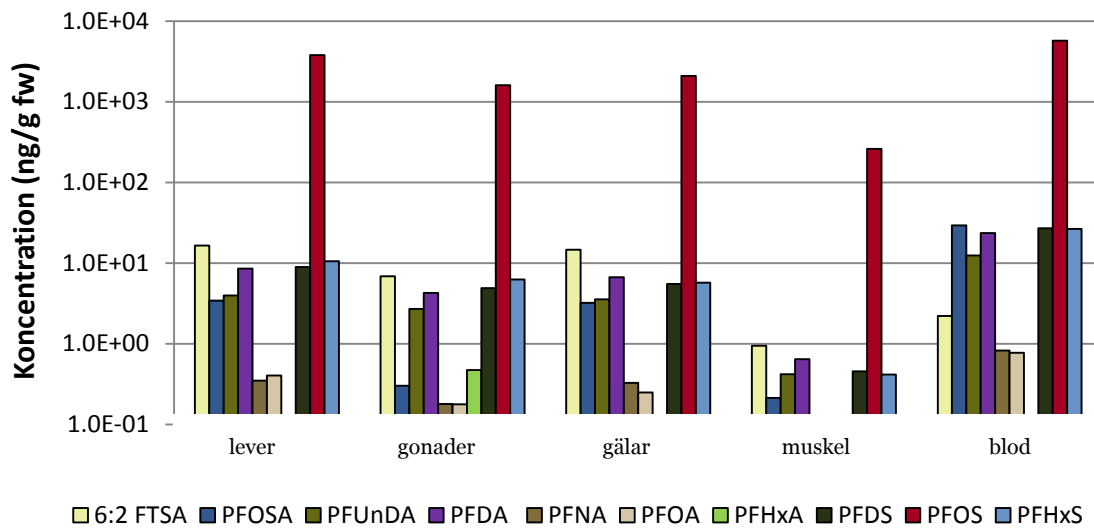
## 1.2 Fisk

Medelkoncentrationerna av PFOS i muskel från abborre från Västra Ingsjön söder om Landvetter flygplats var 54 ng/g färskvikt (17-205 ng/g färskvikt) och i abborre från Halmsjön vid Arlanda 305 ng/g färskvikt (90-792 ng/g färskvikt) under tidsperioden 2009-2013, vilket är 14 respektive 66 gånger högre än koncentrationerna i fisk från referenssjöarna Sandsjön (Landvetter) och Valloxen (Arlanda) där PFOS-halten var ca 4 ng/g färskvikt. Halterna i fisk har inte minskat signifikant ( $p > 0.05$ ) under denna tidsperiod, se figur 1.3.



Figur 1.3. Koncentrationen PFOS i abborre från Västra Ingsjön och Halmsjön under tidsperioden 2009-2013.

PFOS var den dominerande substansen och utgjorde 89-100% av den totala PFAS-koncentrationen i abborre från både Västra Ingsjön och Halmsjön, oberoende av vilken del av fisken som analyserades, se figur 1.4. PFAS binder till proteiner i blodet och de blodrikare vävnaderna uppvisade högre halter än muskeln.

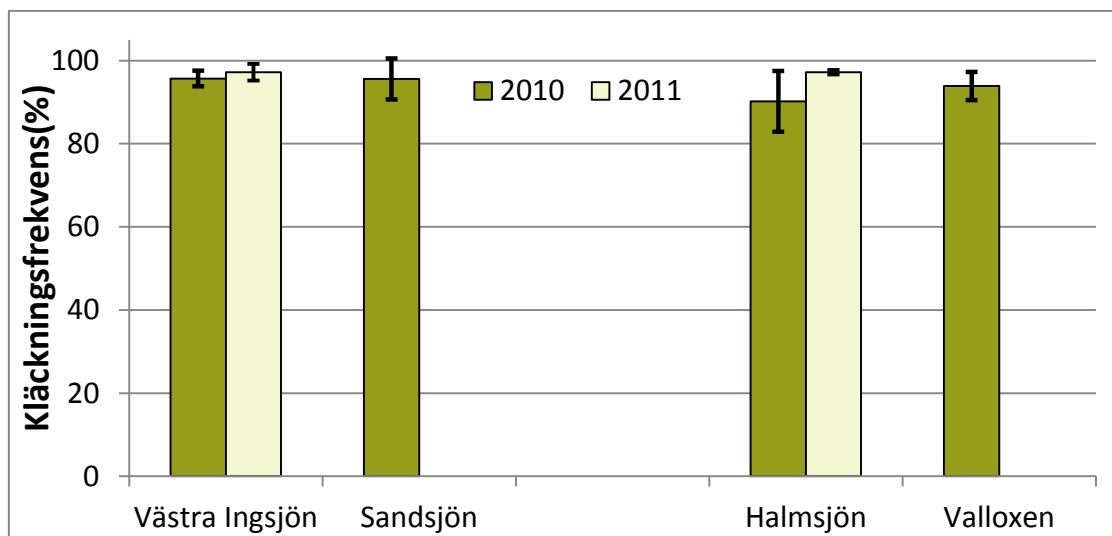


Figur 1.4. Koncentration av PFAS i olika vävnader från abborre. Observera logskalan på y-axeln.

Alla individer av fisk har ålders- och könsbestämts. Multivariat modellering i form av principalkomponentanalys (PCA) har utförts med ålder, kön och PFAS-halten i muskel som ingående parametrar. Den visade att varken ålder eller kön hade någon tydlig inverkan på PFAS-halten. Exponeringsförsök med sebrafisk på laboratoriet resulterade dock att hannar innehöll högre halter än honorna.

### 1.3 Ekotoxikologi

I projektet RE-PATH har under åren ett flertal toxicitetstester genomförts. Abborrom och grodyngel har samlats in från Västra Ingsjön och Halmsjön samt från referenssjöarna. Kläckningsfrekvens, immobilitetstest och morfologi har studerats på olika generationer. Inga ekotoxikologiska effekter har kunnat påvisas för dessa lokala arter vid de koncentrationer som råder i vattnet där de härstammar ifrån, se figur 1.5.



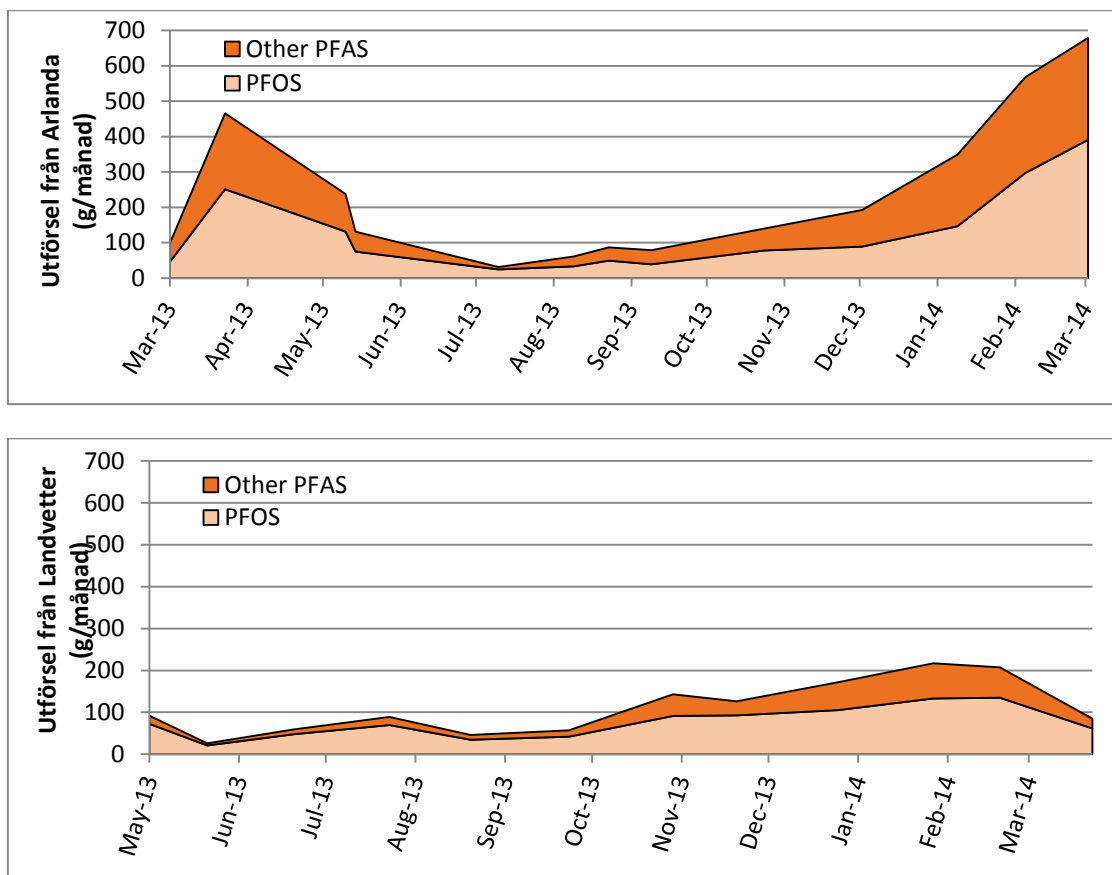
Figur 1.5. Kläckningsfrekvens (medelvärde och 95% konfidensintervall, n=3) av rom från abborre insamlad från Västra Ingsjön, Halmsjön samt referenssjöarna Sandsjön och Valloxen.

## 1.4 Utgående mängder från Stockholm Arlanda Airport och Göteborg Landvetter Airport

Från och med våren 2013 och ett år framåt togs prover en gång per månad vid provpunkten Broby F utanför Arlanda flygplats som är den punkt där flygplatsens avrinning från Kättstabäcken och Halmsjöbäcken går ihop och sedan rinner vidare ut i Mälaren. På samma sätt togs prover vid utloppet från Landvetters flygplats som så småningom når Västerhavet via Issjöbäcken och Västra Ingsjön. Vid dessa båda provtagningspunkter mäts vattenflödet kontinuerligt. Figur 1.6 visar den uppskattade mängden av PFOS och övriga detekterade PFAS som månadsvis rinner ut från respektive flygplats.  $\sum_{12}$ PFAS vid Arlanda varierade mellan 61-680 g per månad och vid Landvetter var motsvarande variation 24-220 g per månad. Detta motsvarar en årlig tillförsel av PFAS till Mälaren och Västra Ingsjön på 2.4 kg respektive 1.3 kg, där PFOS utgjorde 54% (1.3 kg) respektive 81% (0.9 kg). Halldata av PFAS för dessa beräkningar baseras på stickprov taget varje månad under ett år och inte på flödesproportionerliga samlingsprov.

På samma sätt har prover tagits i Uppsala, vid Fyrisåns utlopp till Mälaren. Den mängd  $\sum_{12}$ PFAS som årligen rinner ut där uppskattas till 4.6 kg (1.9 kg PFOS), d.v.s nästan dubbelt så mycket som utförseln från Arlanda. Tillförseln av PFOS till hela Mälarens yta via atmosfären är i samma storleksordning och uppskattas till ca 1.5 kg/år. Vid Landvetter flygplats är atmosfärens bidrag till Västra Ingsjön ca 0.0044 kg/år, vilket är betydligt mindre än bidraget från flygplatsen.

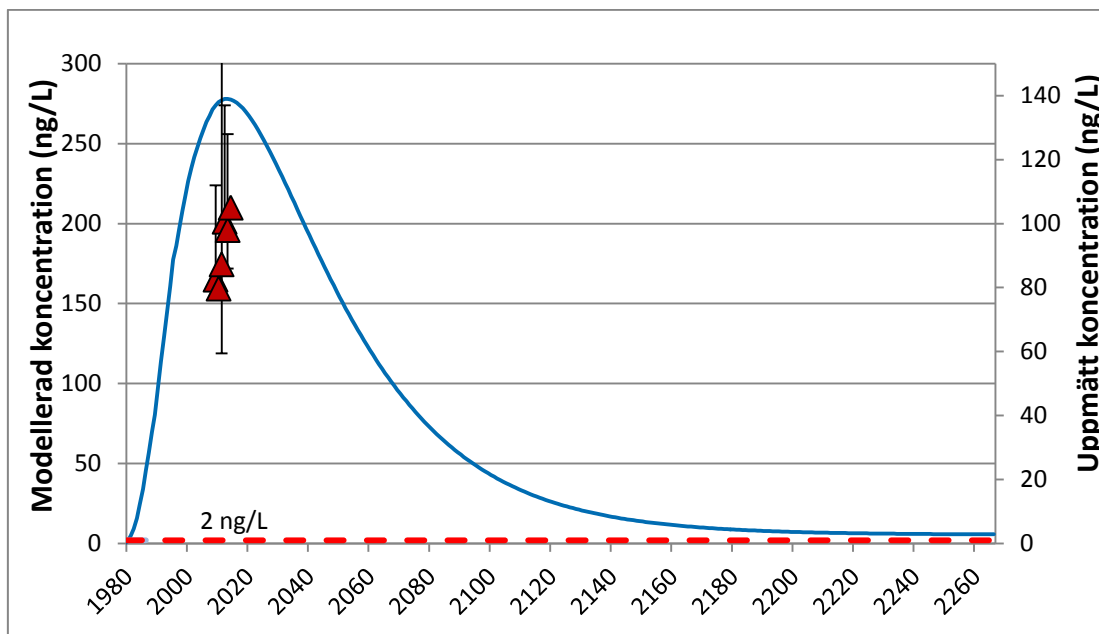
Vattnet i Mälaren som kommer från Uppsala och Märstaån passerar vattenintaget vid Görvelns vattenverk där Norrvatten har sin dricksvattenproduktion. Här är halterna av PFOS ca 6 ng/l, vilket är i samma nivå som "opåverkade" dricksvatten generellt i landet. Det tyder på att den PFOS som härstammar från Stockholm Arlanda Airport, Uppsala och övriga källor som t.ex. Rosersbergviken där det också skett brandövningar med PFAS-innehållande AFFF snabbt späds ut och inte nämnvärt påverkar dricksvattenkvaliteten.



Figur 1.6. Uppskattade utsläpp av  $\Sigma_{12}$ PFAS från Stockholm Arlanda Airport (överst) och Göteborg Landvetter Airport (nederst).

## 1.5 Långsam avklingning av halterna i närmiljön

En dynamisk massbalansmodell har använts för att beräkna hur lång tid det tar för mark och vatten på Arlandaområdet att nå bakgrunds nivåer av PFOS om inga saneringsåtgärder utförs. Arbetet har bland annat inneburit att uppskatta vattenflödena inom området och hur de är kopplade till varandra samt att beräkna den totala mängd PFOS som tillförts marken historiskt. Resultaten visar att minst 38 kg PFOS har använts sedan starten 1980. Enligt modellberäkningarna finns hälften av denna mängd fortfarande kvar i området och den största mängden PFOS ligger lagrad i marken och endast en liten del i grundvattnet (ca 0.3 kg). Grundvattnet utgör enligt dessa beräkningar en temporär reservoar dit PFOS transporteras för att sedan fortsätta ut via grundvattenströmningar som så småningom når ytan och bidrar till utflödet i Mälaren. Utflödet från mark (räknat på år 2012) domineras av transport till grundvatten (40 %) följt av transport till Kättstabäcken (36%), transport till Kolsta reningsverk (20%) medan transporten till Halmsjön utgör 4 % av utflödet från mark. I Halmsjön finns den huvudsakliga andelen PFOS i vattenmassan och endast en mindre del i sedimentet. Modellresultaten visar att med dagens minskningstakt kommer det ta ca 200 år för Halmsjön att nå bakgrunds nivåer i vattnet, se figur 1.7. Modellen visar också att den dominerande transportvägen för PFOS ut från flygplatsområdet som helhet är via Broby F och därmed går den största delen ut i Mälaren.



Figur 1.7. Modellerade halter av PFOS (ng/l) i Halmsjöns ytvatten över tid (blå linje) samt uppmätta halter (röda trekantar). Notera de olika skalorna på y-axlarna för uppmätta respektive modellerade koncentrationer.

## 1.6 Exponering för människa och djur

Det nuvarande TDI-värdet för PFOS är 150 ng/kg kroppsvikt och dag. Livsmedelsverket har uppskattat att medelintaget av PFOS för en vuxen som äter en balanserad kost är 0.6 ng PFOS/ kg kroppsvikt och dag, varav ca en tredjedel kommer från dricksvattnet. Om maten och/eller dricksvattnet är förorenat av PFOS ökar exponeringen och tabell 1 visar hur det dagliga intaget av PFOS ökar beroende på exponeringskälla. Tabellen visar att även om man äter fisk från Västra Ingsjön eller Halmsjön en gång per vecka så överskrider inte TDI.

På flera ställen i Sverige har PFAS från brandskumsanvändning förorenat grundvattnet vilket har lett till höga halter i dricksvattnet som t.ex. upptäcktes i Kallinge i Blekinge 2013. Tabell 1.1 visar att det dagliga intaget av PFOS ökar betydligt vid konsumtion av kraftigt förorenat dricksvatten och särskilt för små barn som får mjölkersättning, där TDI överskrider med mer än en faktor 3. När det gäller områdena kring Stockholm Arlanda Airport och Göteborg Landvetter Airport finns inga kommunala dricksvattentäkter i närheten. De privata grundvattenbrunnar som finns i närheten har kontrollerats av Swedavia och uppvisar bakgrundshalter.

Tabell 1.1. Uppskattad humanexponering vid konsumtion av mat och vatten med olika föroreningsgrad.

Individ	Konsumtion	PFOS koncentration	Dagligt intag (ng/kg kroppsvikt dag)
Medelsvensk	Bakgrundsexponering (mat och dricksvatten)		0.6
Fiskare, Arlanda	Bakgrundsexponering + fisk från Halmsjön 1 portion/vecka	300 ng/g f v	94
Fiskare, Landvetter	Bakgrundsexponering + fisk från Västra Ingsjön 1 portion/vecka	40 ng/g f v	13
Vuxen, Kallinge	Bakgrundsexponering + dricksvatten från Kallinge, 2 l per dag	4000 ng/l	114
Bebis, Kallinge	Mjölkersättning, dricksvatten Kallinge, 0.8 l per dag (bebis 4 månader, vikt 6.6 kg)	4000 ng/l	485

Ett flertal individer av abborre har under projektets gång analyserats med avseende på PFOS-innehållet i fler organ än muskel, d.v.s. lever, blod, gälar och gonader. Då PFAS binder till proteiner i blodet innehåller blodrika organ betydligt högre halter än muskeln. Från mätningarna kan det uppskattas att en enskild individ av abborre kan innehålla upp till 300 µg PFOS. Även om dessa halter inte leder till en negativ hälsopåverkan på människor i närområdet kan det inte uteslutas att fiskätande däggdjur och fåglar kan få en betydligt högre exponering, särskilt som dessa normalt äter hela fisken, och inte bara muskeln. Andra studier har visat att minskar fångade i Märstaområdet har de högsta halterna av PFOS som uppmätts i Sverige.

## 1.7 Slutsatser

PFOS har under alla år som projektet pågått kunnat detekteras i samtliga prover som insamlats runt flygplatserna och även i alla prover tagna i referenssjöar, det vill säga från områden som inte har påverkats av flygplatser, samhällen eller industri. I t.ex. Halmsjön är halterna i ytvatten och fisk 100 gånger högre än i referenssjöarna. Trots de relativt höga halter som uppmätts i fisk och vatten bedöms exponeringen för människor vara liten då ingen kommunal dricksvattentäkt är påverkad samtidigt som fisken kan anses säker för konsumtion 1-2 gånger per vecka (enligt nu gällande TDI), vilket är betydligt oftare än normal konsumtion av egenfångad fisk, och fiskeförbud dessutom har upprättats. Läckaget från brandövningsplatserna får dock en betydelse som lokal utsläppskälla då det inte kan uteslutas att fiskätande däggdjur skulle kunna påverkas då de konsumerar hela fisken.

Då PFOS tillhör en grupp av ämnen där det inte finns några naturliga nedbrytningsvägar kommer dessa ämnen stanna kvar i naturen under långtid framöver. Det finns idag metoder som renar vatten från PFOS men effektiviteten och kostnaden styrs av hur rent vattnet är från andra ämnen som t.ex. humus och metaller. En multikriterieanalys (riskvärdering), d.v.s. en värdering av möjliga åtgärder, deras kostnader och eventuella bieffekter i relation till de önskade effekterna skulle behöva göras innan ett beslut om omfattande renings- eller saneringsåtgärder tas.

Den största oron gällande PFAS idag är förknippad med förorening av dricksvatten via grundvattnet. Det är därför viktigt att redan förorenade områden varken i dag eller i framtiden ger upphov till läckage som når vårt dricksvatten. Kartläggningar över sådana områden behövs samt platsspecifika riktvärden för mark och grundvatten.

Det är också viktigt att lära sig från erfarenheterna kring PFOS-problematiken. Ett flertal AFFF som används idag innehåller PFAS som kan brytas ner till extremt stabila PFAA i naturen. Sedan oktober 2014 är PFDS, PFDA, PFNA och PFHxS upptagna på "SIN-listan" ([www.chemsec.org](http://www.chemsec.org)), som är till för att verka för en värld fri från substanser som är identifierade som "Substances of Very High Concern", enligt kriterier från EU:s kemikalielagstiftning REACH. Detta visar att problematiken med förorening av vår miljö av PFAS inte begränsar sig till enbart PFOS i AFFF, utan hela gruppen PFAS med dess stora användningsområde.

***Abbreviations used within this report***

AFFF – Aqueous film forming foam

PFAS - per- and polyfluorinated alkyl substances)

PFAA – perfluorinated alkyl acids

PFSA – perfluorinated sulfone acids

PFCA – perfluorinated carboxylic acids

PFBS – perfluorobutane sulfonic acid

PFHxS – perfluorohexane sulfonic acid

PFOS – perfluorooctane sulfonic acid

PFDS – perfluorodecane sulfonic acid

PFHxA – perfluorohexanoic acid

PFHpA – perfluoroheptanoic acid

PFOA – perfluorooctanoic acid

PFNA – perfluorononanoic acid

PFDA – perfluorodecanoic acid

PFUnDA – perfluoroundecanoic acid

PFOSA – perfluorooctane sulfonamide

6:2 FTS – 6:2 fluorotelomer sulfonate



## 2 Introduction

### 2.1 Background

In the year 2006, IVL Swedish Environmental Research Institute Ltd conducted an environmental screening study of per- and polyfluoroalkyl substances (PFAS) on commission by the Swedish Environmental Protection Agency (Woldegiorgis et al., 2006). Enhanced levels of perfluorooctane sulfonate (PFOS) were found in surface water south of Göteborg Landvetter Airport, which led to further studies confirming that also the fish contained enhanced PFOS levels (Woldegiorgis and Viktor, 2008). The usage of PFAS-containing Aqueous Foam Forming Films (AFFF) during fire extinguishing was later shown to be the main source to the elevated levels (Vägverket Konsult AB 2007, Woldegiorgis och Viktor, 2008). In a follow-up study in 2007 high levels were also found in the fish in Lake Halmsjön, just outside Stockholm Arlanda Airport, which led to a ban on fishing in the lake.

As a result of these findings, Swedavia Swedish Airports (previously Luftfartsverket) and IVL initiated the co-financed project *Risks and Effects of the dispersion of PFAS on Aquatic, Terrestrial and Human Populations in the vicinity of international airports* (RE-PATH) in 2009. The aim was to study the long-term consequences of the use of PFAS-containing AFFF at Göteborg Landvetter Airport and at Stockholm Arlanda Airport.

Since the project start in 2009, about 700 samples have been collected and analysed for their PFAS content. Several toxicity tests have been performed to investigate potential influence of PFAS on reproduction, immobility and hatching frequency. The sediment-water balance has been studied to elucidate whether the sediment contributes substantially to the water concentration. The bioaccumulation and depletion potential was studied in goldfish, zebrafish and in crayfish. Furthermore, the estimated PFOS load to Lake Mälaren was compared to releases via other sources. A mass balance model was developed to investigate the distribution and the rate of decline of PFOS-levels from the environment around Stockholm Arlanda Airport.

This report summarizes the results from 2009 to 2014. More details are given in individual reports and publications (Woldegiorgis et al., 2010, Norström et al., 2011, Norström& Viktor, 2012, Norström et al., 2013, Ahrens et al., 2015).

### 2.2 Per- and polyfluoroalkyl substances (PFAS)

#### 2.2.1 Properties and use

PFAS is the joint term for a large group of substances which have been used for commercial purposes since the 1950's (Kissa, 2001). PFAS include substances with different functional groups and with carbon chain lengths spanning from C1 to C20 or even higher, see Appendix for structures of the PFAS included in this study. They all have an aliphatic carbon chain of varying length where the hydrogen atoms attached to at least one carbon atom have been replaced by fluorine atoms, represented by the formula  $CF_3[CF_2]_n[CH_2]_m-$  (Buck et al., 2011). This moiety gives the PFAS oleophobic properties and the strength of the C-F binding causes them to be extremely stable. Their technical properties are unique and have led to an extensive and increasing use in technical applications and articles since the start of the production in the 50's. Main application areas have been in impregnation agents for clothes and textiles, in cleaning agents, ski waxes, as insect repellent and in fire-fighting foams, but also in the surface treatment industry including the development of fluoropolymers used in water-repelling clothes and frying pans (Vestergren, 2011).

Many of the PFAS substances have negligible vapour pressures and will thus not be present in air, however a large number of precursor compounds, such as the fluorotelomer alcohols and the perfluoroalkane

sulfonamidoethanols do. Generally, the organic carbon-water partition coefficient  $K_{oc}$  is used to describe the partitioning between water and solid matrices, but the mechanisms for sorption differ from those of “traditional” persistent pollutants, since PFAS tend to bind to membranes and proteins rather than fatty tissues and octanol-like matrices.

The substances of main public interest are the perfluorooctane sulfonate (PFOS) and the perfluorooctanoate (PFOA) due to their extreme persistence, their frequent occurrence in the environment and their toxic properties as well as the extensive historical uses together with the fact that many precursor compounds eventually break down to form e.g. PFOS and PFOA (Prevedouros et al., 2006). In 2001, the production of PFOS, PFOA and related compounds ceased in the western world (Europe and North America), and was replaced by other, equally persistent but less bioaccumulative PFAS, in particular those that degrade to the four-carbon alternative PFBS. However, production of the long-chain alternatives still occurs in some countries, including e.g. China.

No official data on use of individual PFAS in Sweden is available (SPIN register) but the Swedish Chemicals Agency has reported a total use of 24 tonnes of perfluoroalkane sulfonates (PFASs) (Buck et al., 2011) and perfluoroalkyl carboxylic acids (PFCAs) in the year 2004 (KEMI, 2006).

## 2.2.2 Environmental impact of PFAS

The extensive use of PFAS in a large number of different consumer products has resulted in environmental releases and, as a consequence, exposure to humans and wildlife. Initially regarded as biologically inactive, it was gradually discovered that PFAS could cause adverse toxic effects in animals, and the observation of (PFOS) and (PFOA) in human samples throughout the world led to deep concerns about their potential impact on human and animal welfare. Due to their unusual binding mechanisms in biological tissues, resulting from their oleophobic nature (they bind to proteins rather than adipose tissue), they have extremely long elimination half-lives in humans and animals (3-5 years), thus have long time for concentrations to build-up in humans and wildlife, which may lead to adverse negative effects. (Vestergren, 2011).

## 2.2.3 Regulation of PFAS

PFOS is the only substance in the PFAS group regulated today, and has been the subject of a gradual increase in successive regulation, as listed below:

- 2008: The PFOS content in products must not exceed 0.001 % by mass, and this rule applies also to substances that can degrade to form PFOS (Directive 2006/122/EG). There are some exceptions to this rule, e.g. hydraulic fluids, certain medical devices and metal plating.
- 2008: EFSA (European Food Safety Authority) establishes a TDI-value (tolerable daily intake) for PFOS of 150 ng/kg bw/day
- 2009: PFOS is included on the Stockholm Convention for POPs<sup>1</sup> due to its extremely persistent, toxic and bioaccumulative properties.
- 2011 (27 June): Ban of use of PFOS-containing firefighting foams
- 2013: PFOS is included in the EU Water Framework Directive (Directive 2013/39/EU), with Annual Average Environmental quality standards
  - 0.65 ng/L (fresh surface water)
  - 0.13 ng/L (marine surface water)
  - 9.1 ng/g f.w. (biota)

The National Food Agency in Sweden has recommended a limit for the sum of seven perfluoroalkyl acids PFAA (PFAA<sub>7</sub>); 90 ng/L (Livsmedelsverket 2015).

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<sup>1</sup> <http://chm.pops.int/default.aspx>

## 2.3 PFAS and Aqueous Film Forming Foams (AFFF)

AFFF is used for fires classified as B – liquid fires, which includes e.g. petroleum and various solvent fires. The exact compositions of AFFF mixtures on the market are normally not disclosed to the public for proprietary reasons but typical ingredients are solvents, fluorocarbon surfactants and hydrocarbon-based surfactants (Moody & Field 2000). The foam forms a surface on the top of the burning liquid which prevents evaporation and radiation of heat. The main function of the fluorinated components is to act as main fire extinguishing agents and to produce an sealing sheet over the burning liquid which prevents re-ignition. The new generation of firefighting foams that are used today no longer contain PFOS, due to the recent ban in the EU and North America, but have been replaced by other fluorinated substances.

The products currently available on the Swedish market can contain different additives but the information on the active ingredients is often confidential. In many types of foams the active ingredients are still per- and polyfluorinated substances such as e.g. telomer alcohols (FTOH), 6:2 FTS (Herzke et al., 2012, KEMI 2014, Dagostiono & Mabury, 2014). When these polyfluorinated compounds degrade they form extremely stable final products (e.g. 6:2 FTS forms PFPeA, PFHxA, Wang et al., 2011a) which are just as persistent as PFOS. Lately, also fluorine-free film forming alternatives have been developed, such as Moussol-FF (Dr Sthamer) and Ecoguard (by Chemguard). Siloxane-based surfactants also seem to be new alternatives to the fluorine-based surfactants (Hetzer et al., 2014).

In Sweden there are several producers of firefighting foams and distributing companies on the Swedish market (KEMI 2014).

### 2.3.1 Historical usage of AFFF and its implications

#### 2.3.1.1 Usage of AFFF at Stockholm Arlanda Airport and Göteborg Landvetter Airport

At Stockholm Arlanda Airport there is an old and a new fire training facility. The old training facility was used during the 70s and the beginning of the 80s. AFFF was introduced when the new training facility was constructed in the middle of the 80s. Until 1996, fire drills were performed approximately once per month and about 300 L of foam (AFFF) was consumed each time. AFFF was also used at other occasions, such as special fire drills and for demonstration purposes. The fire drills were performed by filling a pond made of concrete with kerosene that was put on fire. In 1996 the training facility was reconstructed and the concrete was removed. Simulators were used instead and a less amount of AFFF was consumed at the fire drills.

At Göteborg Landvetter Airport the firefighting area has been situated at the same location since the airport opened up in 1977. Exercises are done on a 900 m<sup>2</sup> dense concrete plate provided with drainage. The use of AFFF has been used in similar way as at Arlanda Airport.

From the beginning of 1980 until 2003 a PFOS-containing AFFF (STHMEX-AFFF 3%) was used. In 2003 the product was replaced by PFOS-free AFFFs (Presto AFFF and Moussol APS-P), but the remaining stocks of the old AFFF were used up. The new PFOS-free AFFFs still contained other PFASs (<10% according to the manufacturer), but further details about the products are not available.

Since 2008 AFFF is no longer used at fire drills at the Swedavia airports and in 2011 Swedavia started to use a fluorine-free alcohol-resistant foam (Moussol FF 3/6). All fire engines were also cleaned for residues of PFOS.

### **2.3.1.2 Dispersion of PFAS from fire training facilities in Sweden**

PFAS-containing AFFFs have been used at a large number of firefighting training facilities in Sweden, at the Swedavia airports as well as at the fire training sites run by the Swedish Armed Forces. In addition, the civil Rescue Service (Räddningstjänsten) is also responsible for a number of training sites where PFAS-containing foams have been used. Studies have shown enhanced levels of PFAS at several areas which are connected to firefighting training areas and where fish, surface water, groundwater and also drinking water have been contaminated. Examples of active or former military sites where a PFAS contamination problem has been observed are located in e.g. Uppsala, Ronneby, Halmstad and Tullinge in Botkyrka (Defoort et al., 2012, Filipovic et al., 2015). Several of the Swedavia airports have elevated levels of PFAS in the groundwater (Svensson 2014). Further investigations are on-going.

The dispersion of PFAS from firefighting training areas is also an international problem, which has been shown in several countries where high levels of PFAS have been found in ground- and surface water and in fish in lakes close to the airports. There are examples from Canada (Awad et al., 2011, Moody et al., 2002, de Solla et al., 2012), USA (Moody et al., 2003), and Norway (SFT 2008). Some of the studies show that the levels are enhanced even though the use of PFOS-containing AFFF has since long stopped.

### **2.3.1.3 PFAS in drinking water**

In 2011 high concentrations of PFAS were discovered in the drinking water in Tullinge, Botkyrka community (Marko Filipovic, personal comm.). Since then, PFAS have been detected in enhanced concentrations at several places where drinking water is taken from groundwater, such as Uppsala, Ronneby, Båstad and Halmstad. With the exception of Båstad they all have in common that the origin of the contamination is a firefighting training area where the Swedish Armed Forces have practiced fire drills. The concentrations in Ronneby were particularly high (PFAA<sub>7</sub> 6000 ng/L) leading to enhanced blood levels in the local human population.

If PFAA<sub>7</sub> in drinking water exceeds 90 ng/L, the National Food Agency recommends that you should make measurements to minimize the PFAA level as far as possible and examine the origin of the contamination. If PFAA<sub>7</sub>>900 ng/L it is recommended that pregnant women, women that are trying to get pregnant and breast feeding women do not drink the water. (Livsmedelsverket 2015).

### 3 Methods

#### 3.1 Study sites

In Figure 3.1 the study sites are shown and the black dots represent where surface water has been sampled. At Göteborg Landvetter Airport the sites are marked Lref-L6 and at Stockholm Arlanda Airport Aref-A7).

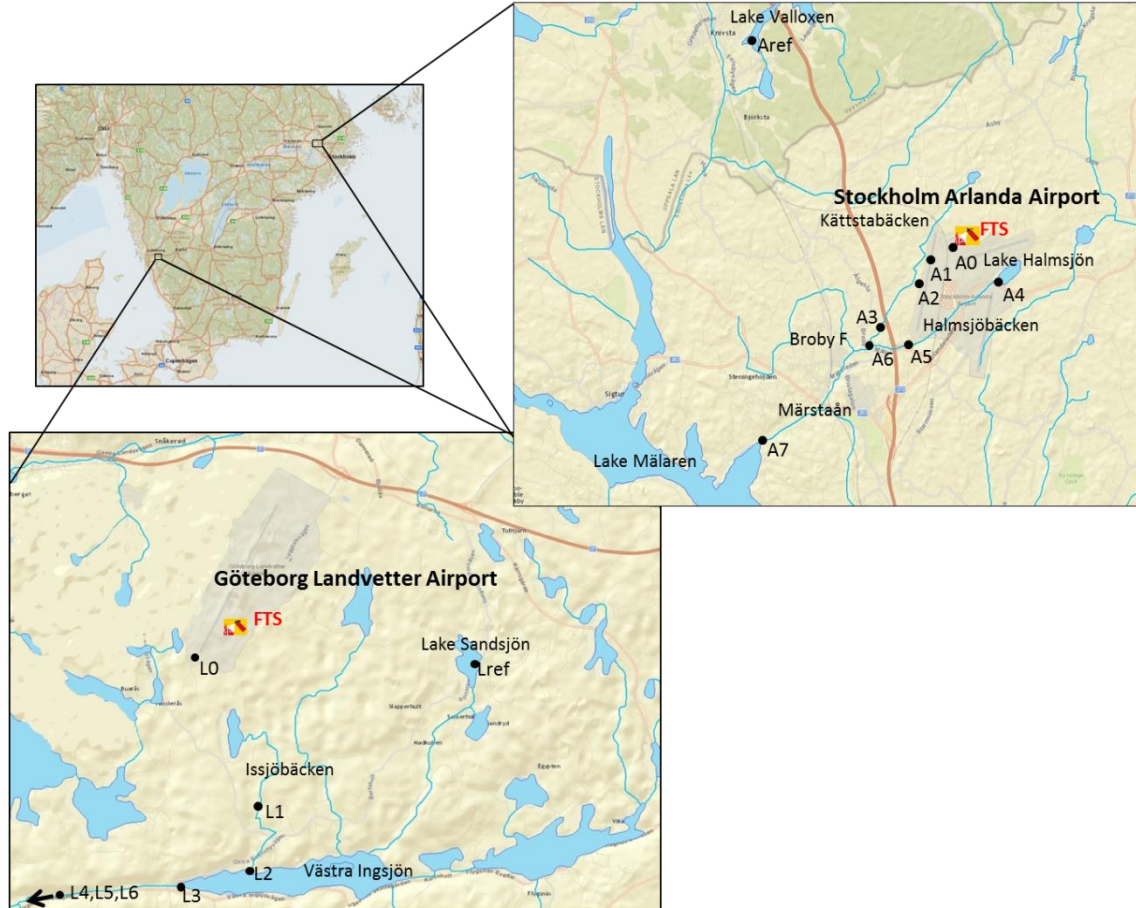


Figure 3.1. Map over the catchment area of Göteborg Landvetter Airport and Stockholm Arlanda Airport. Black dots show sampling sites for water. In the Landvetter area fishing has been performed in Lake Västra Ingsjön, Lake Lilla Issjön and Lake Sandsjön. In the Arlanda area fishing has been performed in Lake Halmsjön and Lake Valloxén. FTS= fire training site.

##### 3.1.1 Göteborg Landvetter Airport

Göteborg Landvetter Airport is an international airport serving the Gothenburg region in Sweden which opened in 1977. With 5.2 million passengers in 2014 it is Sweden's second largest airport.

The area around the firefighting training area slopes to the west and storm water and precipitation are collected in water pipes and is led to the storm water ponds. The water used for firefighting is collected and treated as waste. When the water from the airport area and from the firefighting area has passed all storm water ponds, it runs out in the stream Issjöbäcken, which is the main stream for the catchment area of the airport. Issjöbäcken runs through the Lake Lilla Issjön, and then further down to the Lake Västra Ingsjön, situated south of the airport, see Figure 3.1. In 2011 the contaminated area beneath the firefighting training

area was delimited and the water from the area is collected in the small pond and purified from PFAS in an active carbon filter before it reaches the storm water ponds.

Lake Sandsjön, located east of the airport, has been used as the reference lake. This lake is only contaminated by atmospheric deposition.

### 3.1.2 Stockholm Arlanda Airport

Stockholm Arlanda Airport is an international airport located ~37 km north-northwest of Stockholm and is used by approximately 22.4 million people annually (2014). It is the largest airport in Sweden. This study was carried out in the area surrounding the airport.

A new fire training facility is located in the northern part of the airport area (Figure 3.1). Since the reconstruction in 1996, the water used at the fire drills is transported to Kolsta sewage treatment plant. Today the ground water from the fire fighting training area is pumped through a carbon filter. This filter is not dimensioned for the whole water volume but was originally set up with the aim to study the effectiveness of cleaning PFOS from water.

The hydrology in the area divides the water flow in two directions. The major part of water from the firefighting training area flows to the west into Kättstabäcken, see Figure 3.1, while some of the ground water flows to the east and is connected to Lake Halmsjön, which has its out flow in the stream Halmsjöbäcken. The out flow of the airports catchment area (Kättstabäcken and Halmsjöbäcken) joins at Broby F, which then goes out in Lake Mälaren.

Lake Valloxen, located North West of the airport, has during the project been used as the reference lake. The lake is only contaminated by atmospheric deposition.

## 3.2 Sampling and analytical methods

The matrices included in the measurements are surface water, sediment and fish.

Surface water was sampled each year between 2009 and 2013 at Göteborg Landvetter Airport (sites Lref-L6) and Stockholm Arlanda Airport (sites Aref-A7). The sampling sites at respective airport are shown in Figure 3.1. During 2013-2014 also sampling sites in the Uppsala region were included.

During the whole time period of the project sampling of fish, mainly perch, was performed once or twice per year in Lake Halmsjön and Lake Västra Ingsjön. In the reference lakes, Valloxen and Sandsjön, fishing was performed in 2009 and 2010.

Sediment samples were taken during 2009 and 2010 and analysed for its PFAS content. The sediment used for the water-sediment balance study was sampled in 2010. The sampling of the sediment cores and the dating using the isotope  $^{137}\text{Cs}$  is described in Woldegiorgis et al. 2010.

All samples have been extracted with well-known methods and the extracts have been analysed using HPLC-MS/MS. For further details, see Appendix.

## 3.3 Data analysis

Regression analysis was applied to determine if there are any significant time trends for the content of PFAS in the water and fish samples. To further investigate the variation pattern in the fish Principal Component Analysis (PCA) was performed. Further information is given in Appendix.



### 3.4 Modelling

An important aspect of the contamination problem of PFOS at Stockholm Arlanda Airport is the time required for the system to respond to various control measures, alternatively the recovery time of the system if no measures are taken. To investigate these issues, a dynamic mass balance model was developed and parameterized using the existing knowledge on the dimensions, properties and water flows into an eight-box model (Figure 3.2): one soil segment (1) representing the contaminated area down to a depth of 1 m, one sediment segment representing the upper 0.2 m of the bottom of Lake Halmsjön (4) and six different water segments representing the ground water aquifer (2), Lake Halmsjön (3), the KDA<sup>2</sup> storm water treatment pond (5), the stream Kättstabäcken (6) and the upper and lower parts of the stream Halmsjöbäcken, representing those parts of Halmsjöbäcken located upstream versus downstream of the Benstockenbäcken (7-8). The burial of PFOS in the streams as well as in the storm water pond have been assumed to be insignificant, thus sediment was not included in these parts of the system. Every segment of the model was characterised according to dimensions, particle content as well as organic carbon content, using existing literature. All model parameters and their references are included in Appendix.

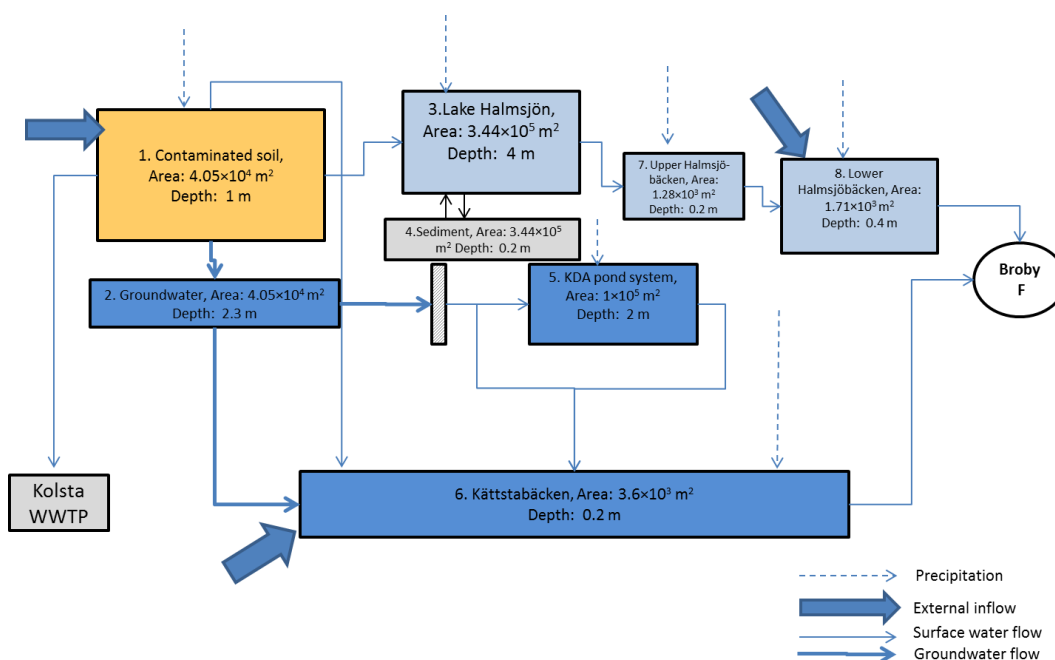


Figure 3.2. Structure of the Stockholm Arlanda Airport eight-box mass balance model. The arrows show the different transport routes of PFOS in the system.

### 3.5 Ecotoxicity tests

The aim of this part of the project was to evaluate the toxicity and chronic effects to ecological relevant species. Laboratory ecotoxicity tests were performed using perch (*Perca fluviatilis*) roe from Lake Västra Ingsjön and Lake Halmsjön, as well as their corresponding reference lakes, Lake Sandsjön and Lake Valloxen. Endpoints as hatching frequency and morphology were studied in water with the same PFOS concentration as the original water from respectively lake.

Different aquatic organisms were also used for determination of the toxicity by pure PFAS (PFOS, PFOA, and 6:2 FTS) and several kinds of firefighting foam. For PFOS-containing AFFF, STHMEX-AFFF 3% was used and for PFOS free AFFF, Presto was used and Moussol FF was used as the fluorine free foam. For the toxicity to embryo/larval stage of zebrafish (*Danio rerio*) a standard test protocol were used according to

<sup>2</sup> KDA = Kättstabäcken DagvattenAnläggning = here used to denote the Kättstabacken stormwater treatment facility



OECD 210. Moorfrog (*Rana arvalis*) and perch (*Perca fluviatilis*) were chosen to represent reptiles and fish living in waterbodies receiving water from the two airports. Toxicity tests were performed with eggs/early larvae stage of both species representing the most sensitive stages in their life-cycle. The exposure time was at least 14 days and we followed the protocol used for determination of toxicity to larvae of fathead minnow (U.S Fish and wildlife service 1996).

The methods are in detailed described in Norström et al. 2011 and Norström and Viktor, 2012.

## 4 Results and discussion

### 4.1 PFAS in surface water near airports

Figure 4.1 shows measured concentrations of different PFAS in surface water outside the two airports in a gradient with increasing distance from the fire training sites (denoted by FTS in the figure). The figure shows concentrations measured in 2012 but both the concentrations and the pattern are similar for all years sampled. As illustrated, the sampling points located in the close proximity to the fire training sites display highest concentrations, which then rapidly decline with increasing distance from the training site, as a result of dilution. However, the levels at the stream outlets which enter Lake Mälaren and the Western Sea are still a factor of 100 and 6, respectively, higher than the levels at the local reference sites Lake Valloxen ( $A_{ref}$ ) and Lake Sandsjön ( $L_{ref}$ ).

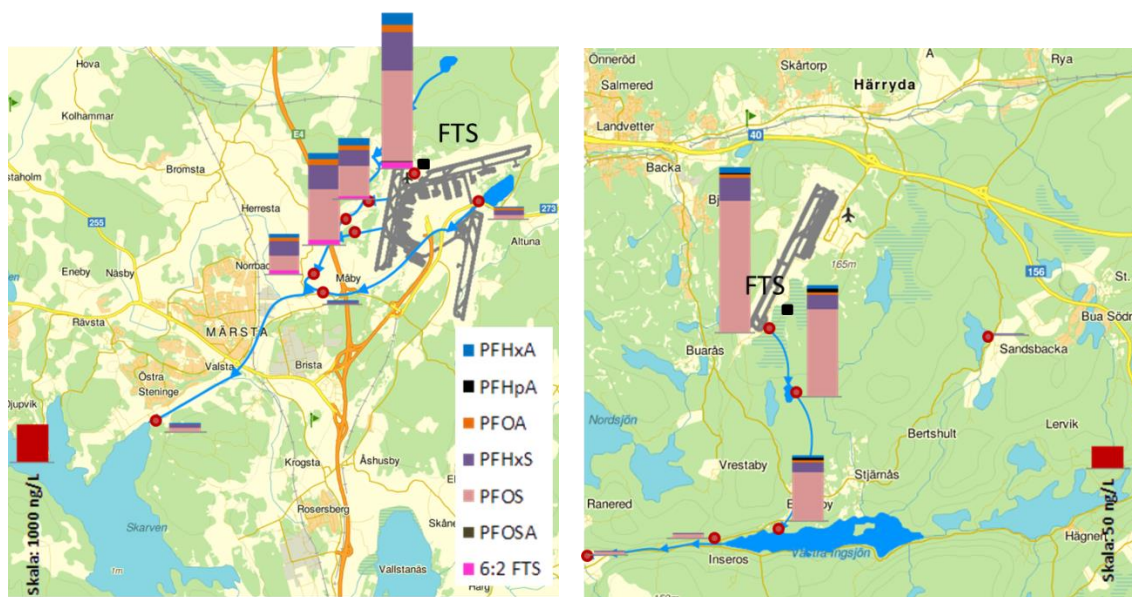


Figure 4.1. Concentrations of PFAS in surface water with increasing distance from the fire training sites (FTS) at Stockholm Arlanda Airport (left) and Göteborg Landvetter Airport (right). Note the different scales of the bars.

#### 4.1.1 PFAS patterns

As evident in Figure 4.1, PFOS account for the main part of the detected PFAS in the water samples near the two airports. This is shown in more detail in Figure 4.2, where the average distribution pattern between different PFAS in surface water at increasing distance from the fire training sites is shown. The figure clearly illustrates that the relative contribution of PFOS decreases with increasing distance from the source, from about 70 % of the total concentration close to the source to 60 % in the outlet, at Göteborg Landvetter airport compared to 40 % in the reference lake Sandsjön ( $L_{ref}$ ). At Stockholm Arlanda the corresponding contribution of PFOS decreases from 60 % near the fire training site ( $A_0$ ) to 46 % at the river outlet to lake Mälaren ( $A_7$ ), which can be compared to about 30 % contribution at the reference lake Valloxen ( $A_{ref}$ ). Overall, PFOS was the most abundant substance in the water samples, followed by PFHxS, with the exception of the two reference lakes, where PFOA was the second most dominant substance in Sandsjön ( $L_{ref}$ ), whereas PFDA and PFBS dominated the samples in the reference lake Valloxen outside Stockholm Arlanda ( $A_{ref}$ ). In a recent screening study of PFAS in river water (Ahrens et al., 2014), where samples from 41 rivers in Sweden were analysed, the most dominant substances were generally PFBS and PFHxS, whereas PFOS accounted for a smaller part of the total concentrations, possibly reflecting a more diffuse origin of PFAS in the rivers.

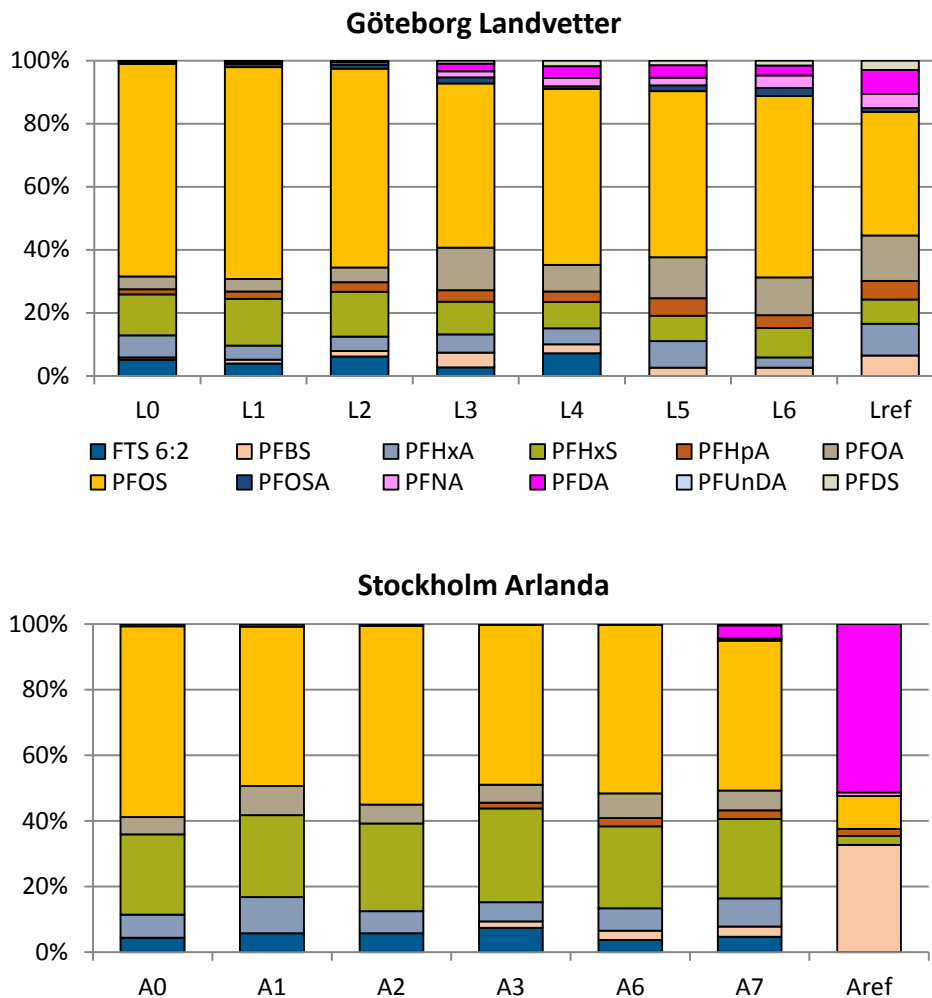


Figure 4.2. Average PFAS patterns in surface water with increasing distance from the source areas at Göteborg Landvetter airport (upper, L0-Lref, see Figure 3.1) and Stockholm Arlanda Airport (lower, A0-Aref, see Figure 3.1), where Lref and Aref are the reference lakes.

#### 4.1.2 Concentration changes of PFAS over time

Figure 4.3 - Figure 4.5 show how the concentrations of four PFAS (PFOS, PFHxS, PFOA and PFHxA) have varied during the time period 2009-2014 at lake Halmsjön (sampling point A4), Arlanda (Figure 4.3), at the outlet of Göteborg Landvetter airport (sampling point L0, Figure 4.4) and at the outlet of lake Västra Ingsjön downstream from Landvetter (sampling point L3, Figure 4.5). Over these five years, only one statistically significant trend ( $p < 0.05$ ) could be observed, namely a slight decreasing trend of PFOA in surface water at the outlet of Lake Västra Ingsjön, downstream of Göteborg Landvetter Airport. For the remaining sites and substances, no clear trend was observed. In 2011, a carbon filter was installed at Göteborg Landvetter to capture the amounts of PFAS in water from the fire training site. This does not seem to have affected the outgoing concentrations to any significant extent. There even seems to be a slight increase of PFOS and PFHxA (Figure 4.4) in the water leaving the airport, although this was not confirmed statistically. A more detailed assessment of long term trends is presented in chapter 4.6 on modelling at Stockholm Arlanda Airport.

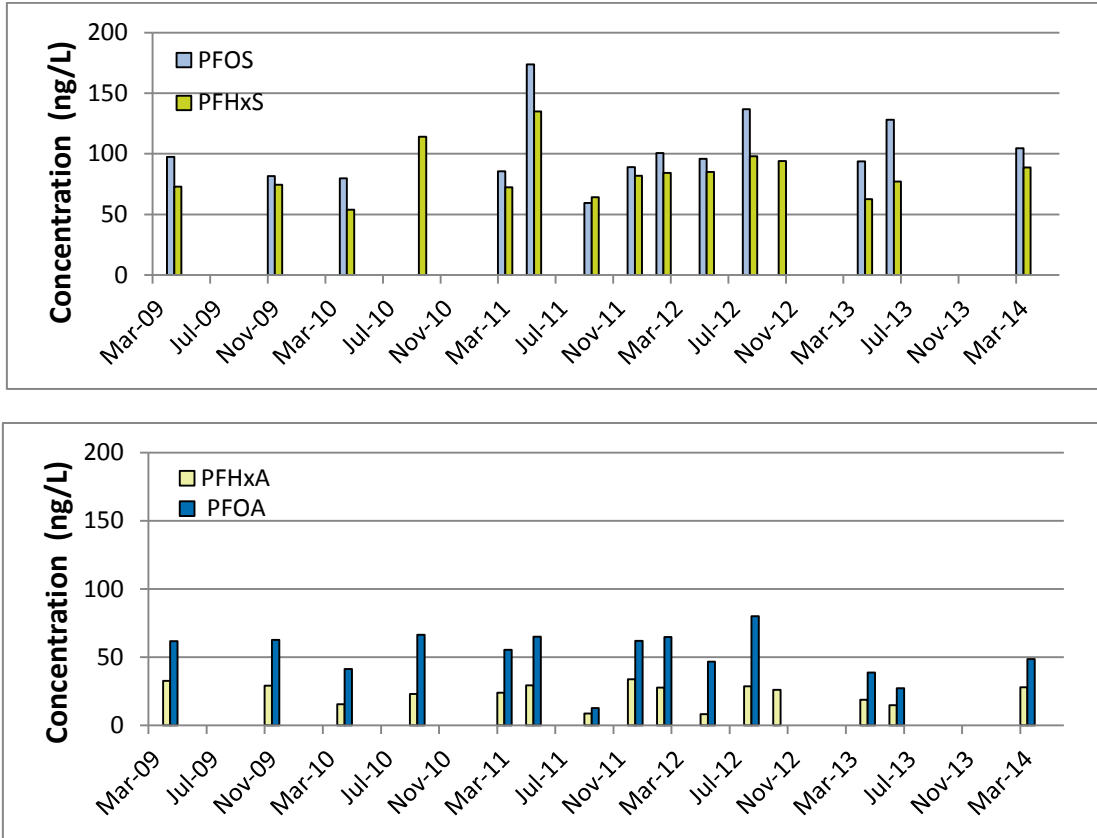


Figure 4.3. Concentrations of four PFAS in surface water of Lake Halmsjön (A4), near Stockholm Arlanda Airport during the time period 2009-2014.

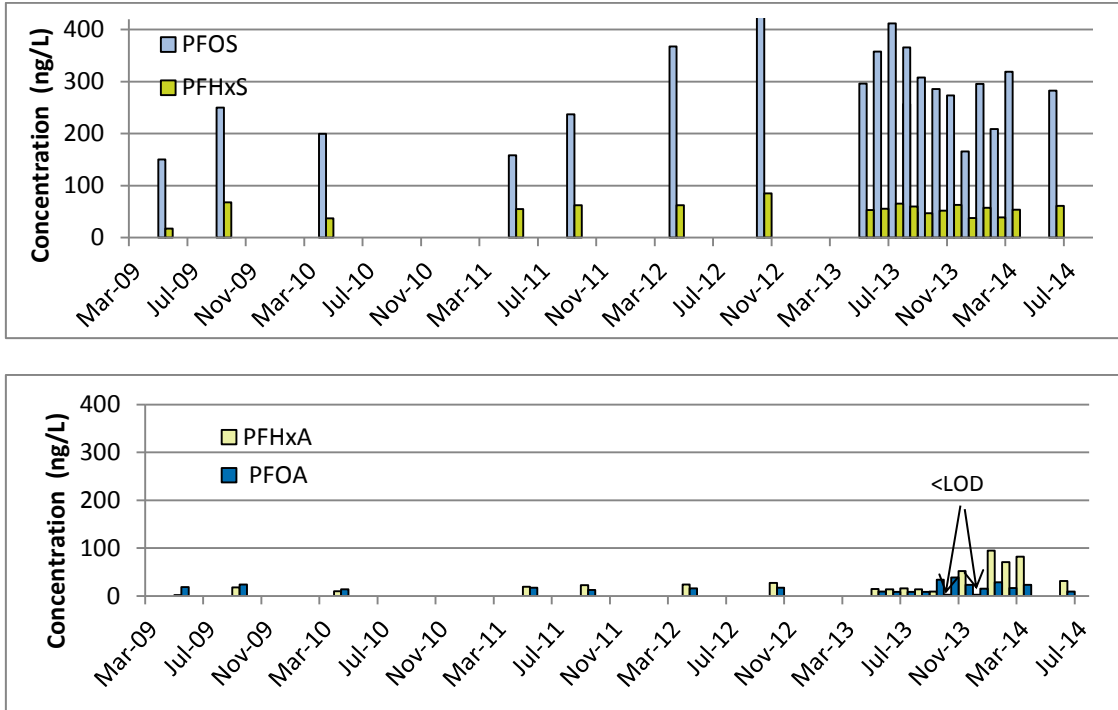


Figure 4.4. Concentrations of four PFAS in surface water at the airport outlet (L0), at Göteborg Landvetter Airport during the time period 2009-2014. Two values below the detection limit have been assigned half the value of LOD.

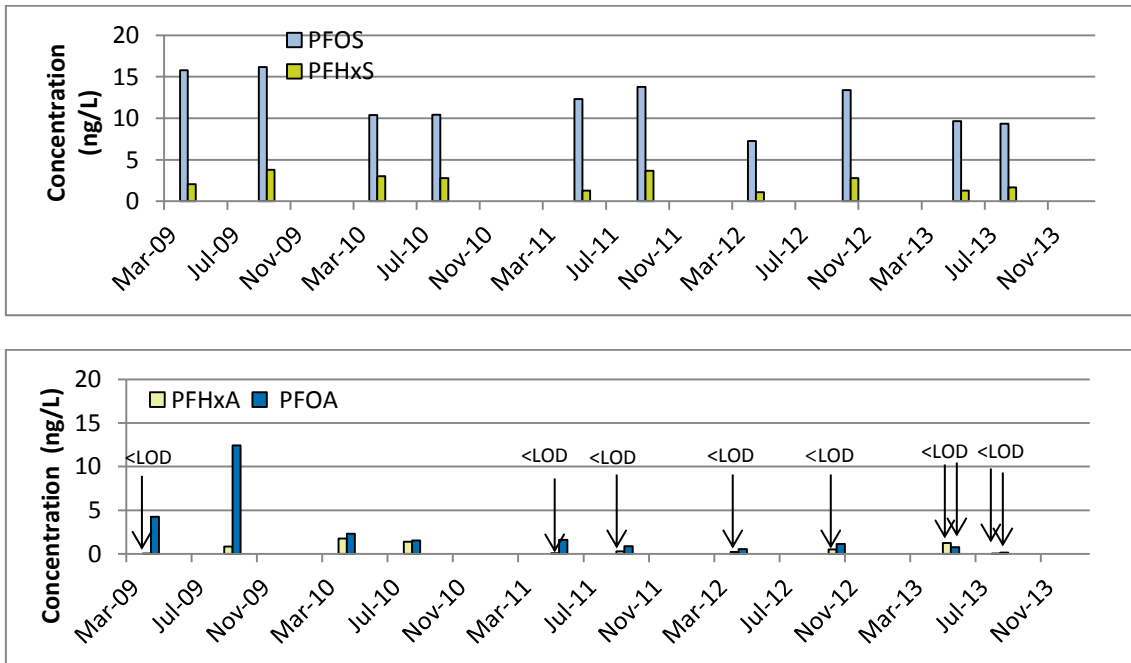


Figure 4.5. Concentrations of four PFAS in the surface water of Lake Västra Ingsjön (L3), downstream from Göteborg Landvetter Airport during the time period 2009-2014. Values below the detection limit have been assigned half the value of LOD, and are denoted with an arrow.

## 4.2 The role of sediments as a secondary source to PFOS in water

The sediment core studies conducted at four different locations in eastern Lake Mälaren indicate an occurrence of PFOS in the area since 30-40 years, although the levels were highest in the most recently deposited sediments, see Figure 4.6. This may be interpreted as a reflection of the increased usage of PFOS since the production start in the early 1950's, possibly in combination with enhanced removal due to bioturbation in the older sediment layers. The PFOS concentration in the top layers are of similar order of magnitude or slightly lower than the levels measured in the top layers of Lake Halmsjön (11-44 ng/g dw) and Lake Västra Ingsjön near Landvetter (7.8 ng/g dw) (Woldegiorgis et al., 2010), which are substantially higher than the concentration in sediments from the reference lake Valloxen of 0.17 ng/g dw (sampled in 2009). This brings out the question – do the high levels in top layers of sediment contribute to the concentrations in the water?

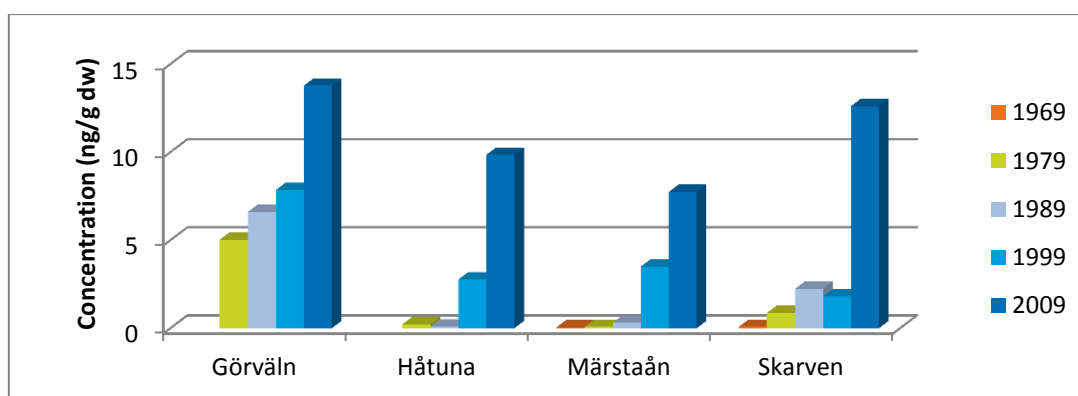


Figure 4.6. PFOS concentrations in sediment cores in eastern Lake Mälaren.

To answer this question, fugacity ratios were calculated for water and sediment in Lake Västra Ingsjön and Lake Halmsjön. A fugacity ratio of 1 implies a system in equilibrium, i.e. no net transport from one phase to another. The ratios were calculated according to:

$$\frac{f_w}{f_{sed}} = \frac{C_w \div Z_w}{C_{sed} \div Z_{sed}}$$

where  $C_w$  and  $C_{sed}$  are the concentrations ( $\text{mol}/\text{m}^3$ ) and  $Z_w$  and  $Z_{sed}$  ( $\text{mol}/\text{m}^3 \text{Pa}$ ) are the fugacity capacities for water and sediment, respectively.  $Z_w$  and  $Z_{sed}$  were calculated to  $4.0 \times 10^5$  and  $2.1 \times 10^7$ , respectively (see Appendix for details). Applying median concentrations in sediment (data from Woldegiorgis et al., 2010 and Axelström, 2010) and water for Lake Halmsjön and Västra Ingsjön (data from Re-path annual reports), with the molecular weight of PFOS 499 g/mol resulted in fugacity ratios ranging between 0.5-1.6 (see Table 4.1).

Table 4.1. Concentrations, fugacity capacities and fugacity ratios between water and sediment of PFOS in lakes Halmsjön and Västra Ingsjön.

Lake	Year	$C_w$ (ng/L)	$C_w$ ( $\text{mol}/\text{m}^3$ )	$Z_w$ ( $\text{mol}/\text{m}^3 \text{Pa}$ )	$f_w$ (Pa)	$C_{sed}$ (ng/g dw)	$C_{sed}$ ( $\text{mol}/\text{m}^3$ )	$Z_{sed}$ ( $\text{mol}/\text{m}^3 \text{Pa}$ )	$f_{sed}$	$f_w/f_{sed}$
Halmsjön	2009	82	$1.6 \times 10^{-7}$		$4.1 \times 10^{-13}$	14	$6.0 \times 10^{-6}$		$2.8 \times 10^{-13}$	<b>1.4</b>
	2010	158	$3.2 \times 10^{-7}$		$7.9 \times 10^{-13}$	49	$2.1 \times 10^{-5}$		$1.0 \times 10^{-12}$	<b>0.8</b>
	2013	98	$2.0 \times 10^{-7}$	$4.0 \times 10^5$	$4.9 \times 10^{-13}$	45	$1.9 \times 10^{-5}$	$2.1 \times 10^7$	$9.1 \times 10^{-13}$	<b>0.5</b>
Västra Ingsjön	2009	16	$3.2 \times 10^{-8}$		$8.0 \times 10^{-14}$	7.8	$3.4 \times 10^{-6}$		$1.6 \times 10^{-13}$	<b>0.5</b>

This implies that currently, the sediment-water systems in Lake Hamsjön and Lake Västra Ingsjön are close to equilibrium, i.e. the net transport of PFOS between water and sediment is limited. The main transport pathways of PFOS to the two lakes are instead inflowing water from the stream Issjöbacken to Lake Västra Ingsjön at Landvetter and via diffuse transport from soil runoff to Lake Halmsjön at Arlanda. In addition small amounts of PFOS are also added via atmospheric deposition to both lake systems. If the inflows from these upstream sources decrease or cease, the importance of sediments as secondary sources of PFOS will increase. The importance of sediments as secondary sources of PFOS has been shown in other studies (Arias et al., 2015).

## 4.3 PFAS in biota

### 4.3.1 Fish

The concentrations of PFOS in perch muscle in Västra Ingsjön, Landvetter and Halmsjön, Arlanda averaged at 54 (17-205) and 305 (90-792) ng/g fw, respectively during the time period 2009-2014 (Figure 4.7), which is 14 and 66 times higher than the observed levels in the reference lakes Sandsjön (average 3.9 ng/g fw) and Valloxen (average 4.6 ng/g f.w.) respectively. No particular trend was observed ( $p > 0.05$ ), although the concentrations observed in the first year (2009) in Lake Halmsjön were significantly higher compared to the concentrations observed in the subsequent years. PFOS accounted for 89 – 100 % of the PFAS concentrations measured in perch in these two lakes, followed by PFOSA and only minor contribution from other PFAS.

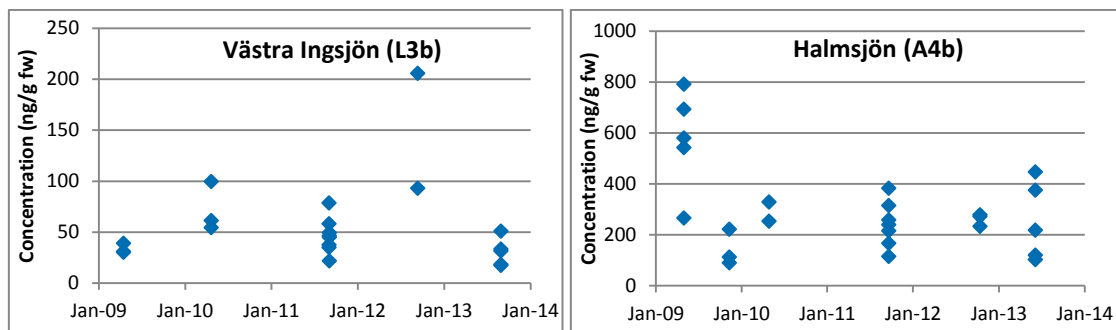


Figure 4.7. Concentrations of PFOS in perch from Lake Västra Ingsjön (left) and Lake Halmsjön (right) 2009-2013.

The concentrations at Lake Västra Ingsjön and Lake Halmsjön, a factor of 5 and 30 higher than median concentrations of PFOS in perch muscle in Lake Vättern (11.3 ng/g fw) and about 30 and 150 times higher than median concentrations measured in perch muscle from the Baltic Sea (2.13 ng/g fw) (Berger et al., 2009).

The results from PCA using concentrations of the sum of PFOS and PFOSA, are shown in Figure 4.8. As evident from the plot on the left, the samples collected in Lake Lilla Issjön (L1), clearly deviate from the remaining sites in the Landvetter region, indicating higher concentrations compared to Lake Västra Ingsjön and the reference lake Sandsjön, which is explained by the proximity of sampling point L1 to the source area. For Lake Halmsjön (plot on the right), the samples collected in 2009 display higher levels, as observed also in Figure 4.7. Other than that, no striking patterns can be distinguished from the plots, i.e. there are no clear differences in concentrations between young and old or between female and male fish which here is shown for Lake Halmsjön (right plot).



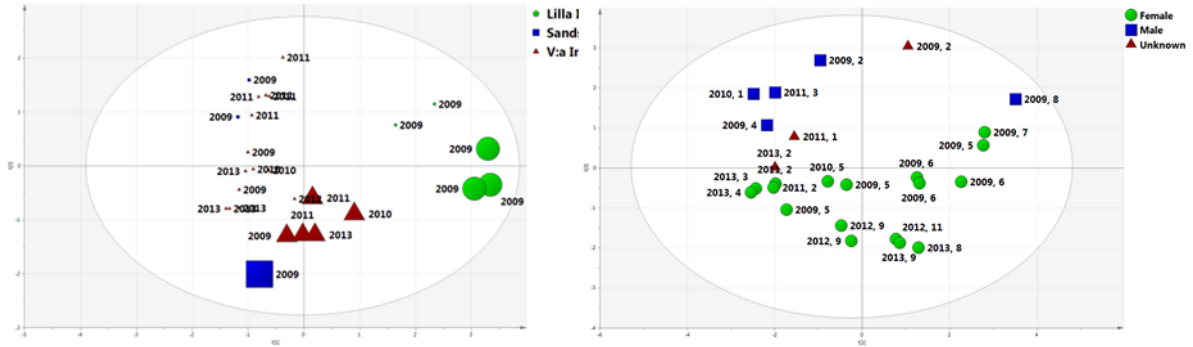


Figure 4.8. PCA score plot based on PFOS+PFOSA concentrations in perch muscle from Lake Västra Ingsjön, Lilla Issjön, Sandsjön (left) and Lake Halmsjön (right) during the time period 2009-2013. The labels represent the year of sampling and in the right plot also the age of the individual I indicated. In the left plot the larger symbols represent females.

#### 4.3.1.1 Tissue distribution and whole body burden of PFAS in perch

The concentrations of PFOS presented above are based on levels in perch muscle, which is the part of the fish normally consumed by humans. Because of their oleophobic nature, PFAS partition predominantly to blood-rich organs (see chapter 2.2.1), thus the muscle levels do not reflect the overall occurrence in fish. The distribution of PFAS between different parts of the fish was investigated by analysing the concentrations in different tissues of three female perch sampled in Lake Halmsjön (A4b) in 2012 (Ahrens et al., 2014). By multiplying those with the individual tissue weight, the distribution between tissues was obtained. The mean whole body burden in perch of all detected PFASs was estimated to be  $334 \pm 80 \mu\text{g}$  absolute ( $n = 3$ ). PFOS was the predominant compound in all tissue samples with a contribution of  $\sim 99\%$  ( $330 \pm 80 \mu\text{g}$  absolute). PFAS whole body burden distribution for the individual tissues decreased in the following order: gonad (54%) > liver (19%)  $\approx$  muscle (17%) > blood (7%) > gill (2%) (Figure 4.9, left). From a concentration perspective, the blood-rich tissues encompassed the highest levels, and were highest in blood and liver (Figure 4.9, right).

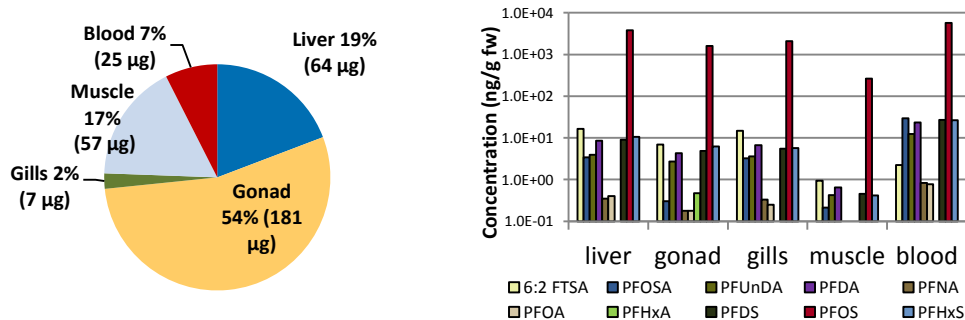


Figure 4.9. PFAS whole body burden distribution in percent and  $\mu\text{g}$  per tissue in brackets (left) and concentrations in liver, gonad, gills, muscle and blood (right, note the log-scale on the y-axis) in perch from Lake Halmsjön, Sweden.

For individual PFAS, the distribution between tissues varied depending on the functional group and fluorocarbon chain length, see Figure 4.10. Most PFASs were mainly distributed in blood (PFOSA, PFDS and  $\text{C}_8\text{-C}_{11}$  PFCAs with a contribution of between  $\sim 46\%$  and  $77\%$ ), whereas 6:2 FTS, PFOS, and PFHxS were predominantly found in gonad tissue (between  $\sim 36\%$  and  $76\%$ ). The compound-specific distribution of PFAS in perch can be influenced by differences in accumulation and elimination of individual compounds (Martin, et al., 2003).

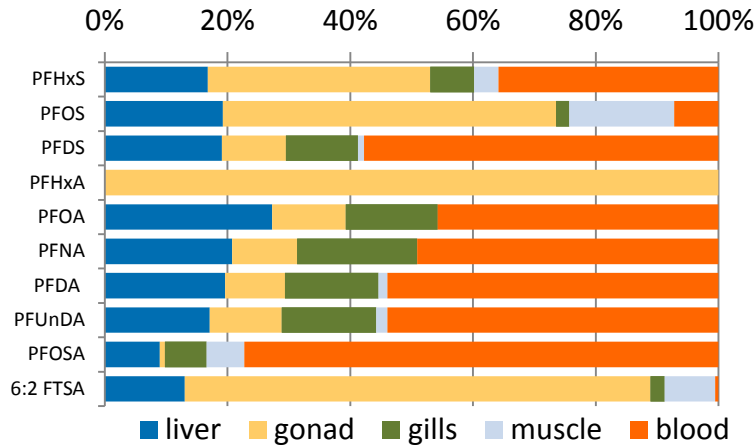


Figure 4.10. Tissue distribution of PFAS burdens in perch from Lake Halmsjön.

The tissue distribution was also studied in perch samples from Lake Sandsjön, Lake Västra Ingsjön and Lake Valloxen (Norström et al., 2011). Even though the concentrations in the individuals from these lakes were lower compared to the perch from Lake Halmsjön, the distribution between the tissues were similar. This pattern has also been seen in other fish species as rainbow trout (*Oncorhynchus mykiss*) (Martin, et al., 2003).

#### 4.3.1.2 Uptake kinetics in fish

The result from the uptake kinetics experiment of PFOS in the goldfish is shown in Figure 4.11, indicating a rapid linear uptake curve that has not reached equilibrium within the experimental sampling time. After 10 weeks of exposure the PFOS concentration in the goldfish was 32 ng/g fw muscle, which is in the same order of magnitude as the concentrations of 4-year old roaches residing in the lake (Norström & Viktor, 2012).

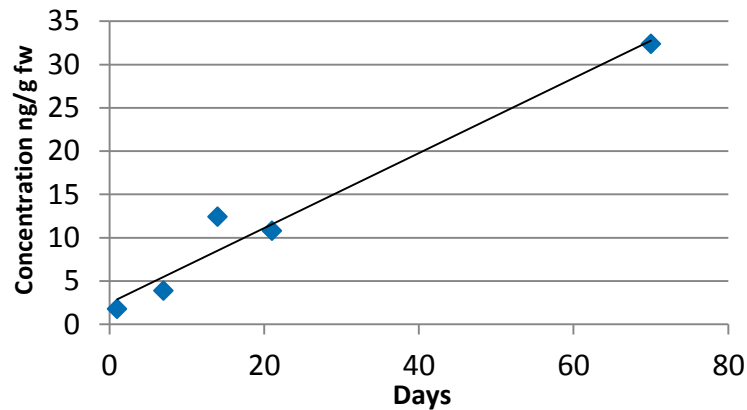


Figure 4.11. Uptake of PFOS in goldfish caged in Lake Halmsjön

#### 4.3.2 Mammals and birds

In addition to the extensive monitoring of PFOS in fish, a few samples of mammals and birds from the Landvetter area were also analysed (Table 4.2). However, these results only represent single individuals and information about their history is lacking.

**Table 4.2. PFOS concentrations in mammals and birds from the Landvetter area.**

Species	Tissue	n	C (ng/g fw)
Red fox, < 1 year	Muscle	1	5.5
Field mouse, <1 year	Muscle	1	<0.6
Tawny owl	Muscle	1	58
Tawny owl	Liver	1	690
Tawny owl	Heart	1	250

### 4.3.3 Ecotoxicology tests

#### 4.3.3.1 Standard toxicity tests

Results from the *standard toxicity tests* with embryo/larvae stages of zebrafish, perch and moorfrog exposed to selected pure PFAS compounds as well as to samples of AFFF are given in Table 4.3. The results are expressed as LC<sub>50</sub>-values in mg/l after 7 days exposure. The two individual tests for PFOS toxicity gave similar results, where zebrafish was the most sensitive species, followed by frog and perch. According to the acute toxicity rating scale<sup>3</sup> the results indicate that PFOS is highly toxic to zebrafish (i.e. LC<sub>50</sub> < 1 mg/L) and moderately toxic (1<LC<sub>50</sub> < 10 mg/L) to frog and perch. The LC<sub>50</sub> value obtained for zebrafish is lower than previously determined values in fathead minnow of 4.7 mg/L (KEMI, 2004). PFOA displays a moderately toxic effect (1<LC<sub>50</sub> < 10 mg/L) towards all tested organisms. The poor solubility in of PFOSA in water hinders a toxicity evaluation for this substance. The shorter carbon chain (6 carbon atoms) of 6:2 FTS seems to reduce the toxicity relative to the eight carbon chain of PFOS.

The relative toxicity to different species of firefighting foam products was similar for the four tested products (Table 4.3). The PFOS-free AFFF showed equal or slightly higher toxicity towards larvae of frogs and fish than the PFOS-containing AFFF, implying that PFOS itself is not the real cause of the observed toxicity, but that the effect is rather due to other components in the foam, such as the tensides or other PFAS substances present in the foam. The fluorine-free foam Moussol FF showed a lower toxicity than the fluorinated foams, whereas the detergent foam used for practicing purposes is the least toxic. All the products are either practically non-toxic (100<LC<sub>50</sub> < 1000 mg/L) or relatively harmless (LC<sub>50</sub>>1000 mg/l) according to U.S Fish and wildlife service 1996 and FFFC (2006). The measured concentrations of PFOS in the lakes were generally <200 ng/L, which is about a factor of 4000 lower than the observed effect levels in the standard acute toxicity tests. Thus, the risk for acute negative effects on the lake ecosystems can be considered to be low.

**Table 4.3. Results from standard toxicity tests with embryo/larvae stages of fish and moorfrog.**

Substance	LC <sub>50</sub> (mg/L)		
	Zebrafish	Perch	Moorfrog
PFOS	0.82 (-)*	>5,0**	2.4 (1.4-4.5)
	0.68 (-)*	9.6 (3.8->)	1.4 (<-2.8)
PFOA	2.9 (-)*	>5.0**	6.3 (2.4->)
6:2 FTS	>8.0**	14.8 (5.8->)	10.5 (6.8-62.2)
<b>Firefighting foams</b>			
PFOS-containing AFFF	260 (75-500)	740 (430-1800)	290 (200-430)
AFFF, PFOS free	300 (-)*	270 (150-430)	150 (90-240)
Moussol FF	1700 (-)*	1500 (-)*	500 (-)*
Detergent foam	3000 (-)*	2700 (-)*	4500 (-)*

\* Not sufficient mortality data for calculating confidence limits \*\* No toxic response in the maximum tested concentration

<sup>3</sup> Kemikalieinspektionens föreskrifter om klassificering och märkning av kemiska produkter; KIFS 2005:7 ISSN 0283-1937 [https://www.kemi.se/Documents/Forfattningar/KIFS/Ko5\\_7.pdf](https://www.kemi.se/Documents/Forfattningar/KIFS/Ko5_7.pdf)

#### 4.3.3.2 *In situ* tests on local perch

The *hatching frequencies* of perch roe are shown in Figure 4.12. As evident from the figure, no significant differences between lakes with a higher load of PFAS compared to the reference lakes were observed.

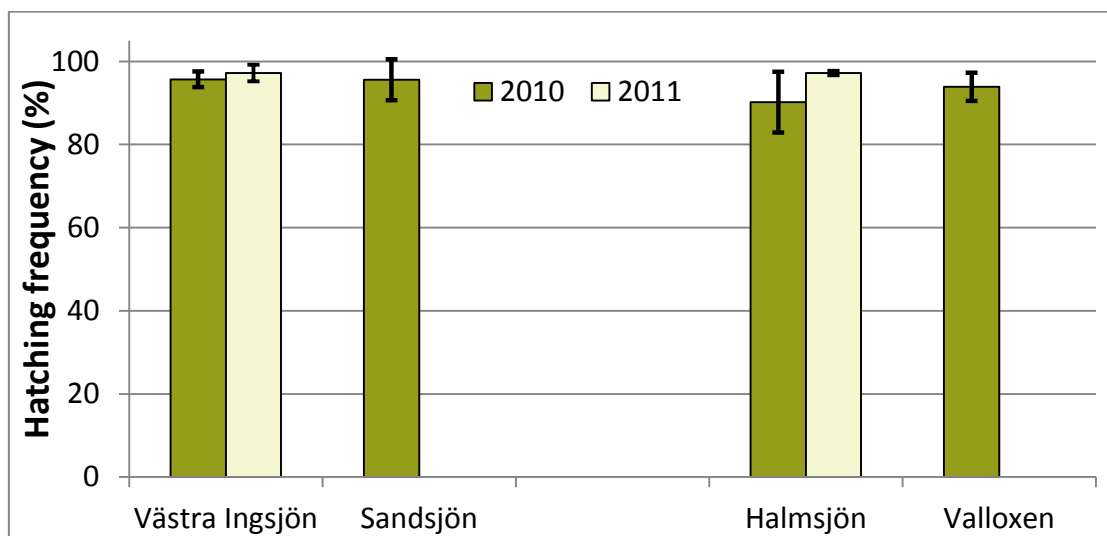


Figure 4.12. Hatching frequencies (average and 95% confidence intervals, n=3) of perch roe collected in lakes contaminated by PFOS and in reference lakes.

The frequency of *deformation* of perch fry varied between zero to 1.7 % for the studied lakes (Norström & Viktor 2012), which can be considered normal compared to e.g. perch fry from Lake Mälaren and the Baltic Sea coast, where deformation frequencies of 1.6 - 1.8 % have been observed (Sternbeck et al., 2005). The few deformations that were observed were four cases of mild scoliosis and three cases of oedema caused by osmosis regulation (Norström & Viktor 2012).

No significant differences regarding the *histopathology of perch liver* samples between lakes with a higher load of PFAS compared to the reference lakes were observed (Norström et al., 2011).

## 4.4 Outgoing amounts of PFAS from Stockholm Arlanda Airport and Göteborg Landvetter Airport

The concentrations of individual PFAS measured at the outlets of the two airports (A6 and L1 at Arlanda and Landvetter, respectively) were multiplied by the measured water flows (monthly average flows at Arlanda, daily average flows for the individual sampling days at Landvetter) to generate an estimate of the amounts of PFAS that leave the airport areas and contribute to the overall environmental burdens for these substances. The estimated amounts of  $\Sigma_{12}$ PFAS<sup>4</sup> leaving the two airports each month varied between 61 and 680 g at Arlanda airport and between 26 and 220 g at Göteborg Landvetter airport (Figure 4.13), with an annual discharge to the surrounding environment of **2.4 kg** and **1.3 kg** respectively. On average, PFOS accounted for 54% (1.3 kg) and 81% (0.9 kg) of total PFAS amounts leaving Arlanda and Landvetter, respectively.

<sup>4</sup> Including PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFBS, PFHxS, PFOD, PFDS, PFOSA, 6:2 FTS.

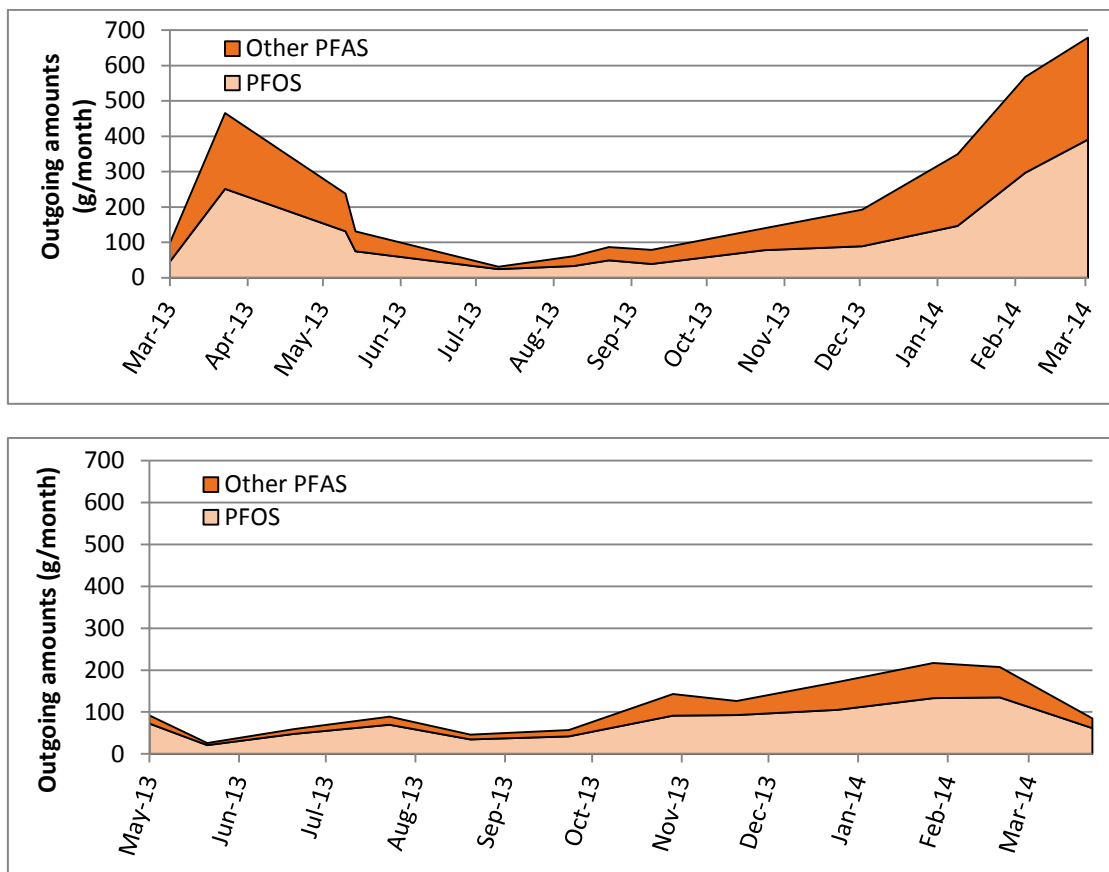


Figure 4.13. Estimated release of  $\sum_{12}$ PFAS from the airport areas at Stockholm Arlanda Airport (upper) and Göteborg Landvetter Airport (lower). The share of PFOS of the total amount of PFAS released is displayed by using a different colour of that substance.

The amounts of PFAS that are discharged from Arlanda Airport are further transported via the river Mårstaån to Lake Mälaren. For Landvetter Airport the amounts of PFAS is transported to the two lakes Lilla Issjön and Västra Ingsjön, and further out via the river Kungsbackaån to the Western Sea.

In the case of Lake Mälaren, Stockholm Arlanda airport is only one of multiple sources that contribute to the total burden of PFAS. Other contributing sources are e.g. the waste water treatment plant (WWTP) Kungsängsverket in Uppsala via river Fyris and via the previous firefighting training school used by the civil rescue service near Rosersbergsviken, where PFOS-containing firefighting foams have been in use (Lundgren och Karlsson, 2009).

## 4.5 Comparison with other sources

In order to estimate the relative contribution of PFOS and  $\sum_{12}$ PFAS from Arlanda Airport to Lake Mälaren, the annual outflow of PFOS (**1.3 kg**) and  $\sum_{12}$ PFAS (**2.4 kg**) from Arlanda Airport were compared to the contribution from the large river Fyris, from sewage treatment plants (STPs) and from the atmosphere. For Landvetter Airport, the annual outgoing amounts to Västra Ingsjön ( $\sum_{12}$ PFAS 1.3 kg, PFOS 0.9 kg) were compared to the estimated input via the atmosphere (see Table 4.4).

This comparison gives an indication of the magnitude of contribution to local PFOS/PFAS contamination by the two airports to the lakes. As evident from Table 4.4, the contribution of PFOS from Landvetter Airport to Västra Ingsjön exceeds the contribution from the atmosphere by a factor of 200, whereas the contribution of PFOS from Arlanda Airport to lake Mälaren is in similar order of magnitude as the contribution from the atmosphere and slightly lower than the inflow from river Fyrisån. Assuming that the

amounts of PFAS that enter Lake Mälaren from Arlanda Airport are further transported out to the Baltic Sea, with negligible sediment retention, this represents about 0.2 % of the total contribution of PFAS from Swedish rivers (Ahrens et al., 2014), and is exceeded by atmospheric deposition to the Baltic Sea by a factor of 330.

**Table 4.4. Comparison of flows of PFOS and PFAS (kg/year) to Lake Västra Ingsjön, Lake Mälaren and the Baltic Sea.**

Flow	PFOS (kg/year)	PFASs (kg/year)	Reference
<b>Contribution to Lake Västra Ingsjön</b>			
From Landvetter via Lilla Issjöbäcken	0.9	1.3 <sup>a</sup>	Re-Path data 2013-14
From atmospheric deposition	0.0044		Norström et al., 2013
<b>Contribution to Mälaren</b>			
From Arlanda via river Märstaån	1.3	2.4 <sup>a</sup>	Re-Path data 2013-14
From river Fyris	1.9	4.6 <sup>a</sup>	Re-Path data 2013-14
From the atmosphere	1.5		Ahrens et al., 2015
<b>Contribution to the Baltic Sea</b>			
From STPs at the Baltic coast	9		Andersson et al., 2012
From atmospheric deposition	238	804 <sup>b</sup>	Filipovic M. et al., 2013
Via 44 Swedish rivers		1150 <sup>c</sup>	Ahrens et al., 2014

<sup>a</sup>PFASs= 12 substances, <sup>b</sup>PFASs= PFHxA, PFOA, PFDA, PFOS, <sup>c</sup>PFASs= 13 substances

These estimates illustrate that the overall contribution from local sources such as Arlanda or Landvetter airport, is small in a broader context (e.g. contribution to the Baltic Sea), but they may still be the dominant sources in the local area (e.g. Västra Ingsjön, Lake Halmjön) where concentrations may be elevated by several orders of magnitude compared to background areas.

## 4.6 Modelling results for PFOS in the area of Stockholm Arlanda Airport

### 4.6.1 Estimated PFOS loads from a historical perspective

The estimated/predicted annual amounts of PFOS emitted to soil via firefighting foams and deposited from the atmosphere via atmospheric deposition to the Arlanda area in the time period 1980-2020 are shown in Figure 4.14. The estimated emission of PFOS in the area since 1980 is 38 kg, whereas the atmospheric deposition during the same time period is estimated to be negligible, only 0.013 kg, which corresponds to 0.03 % of the totally amounts applied.

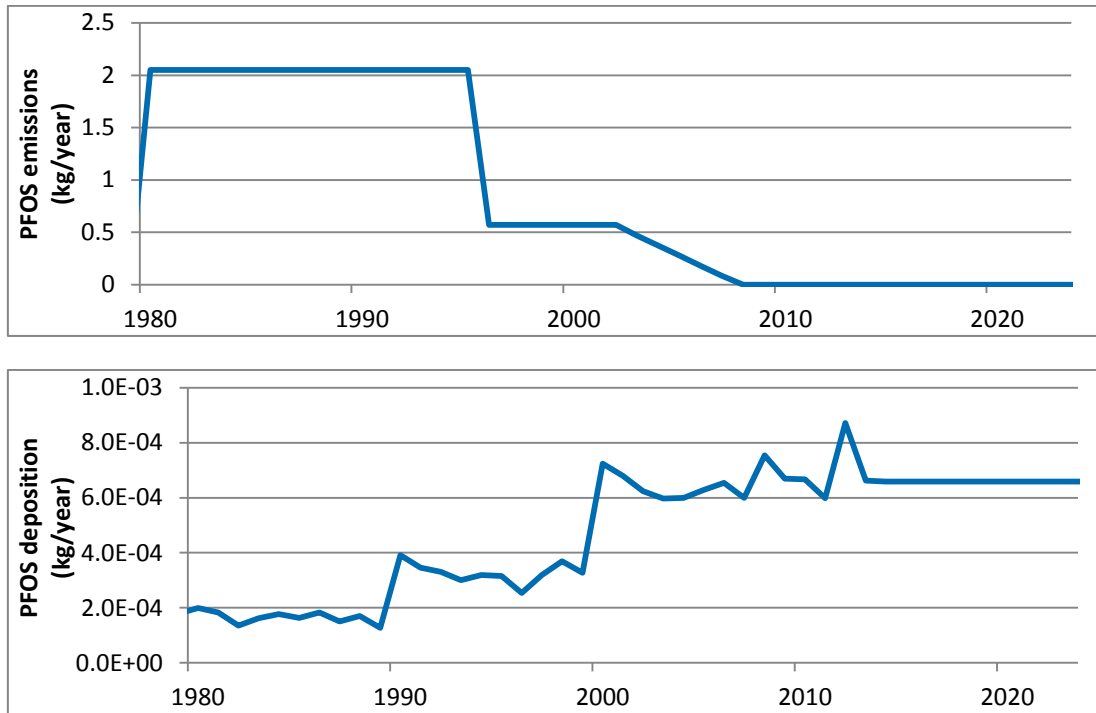
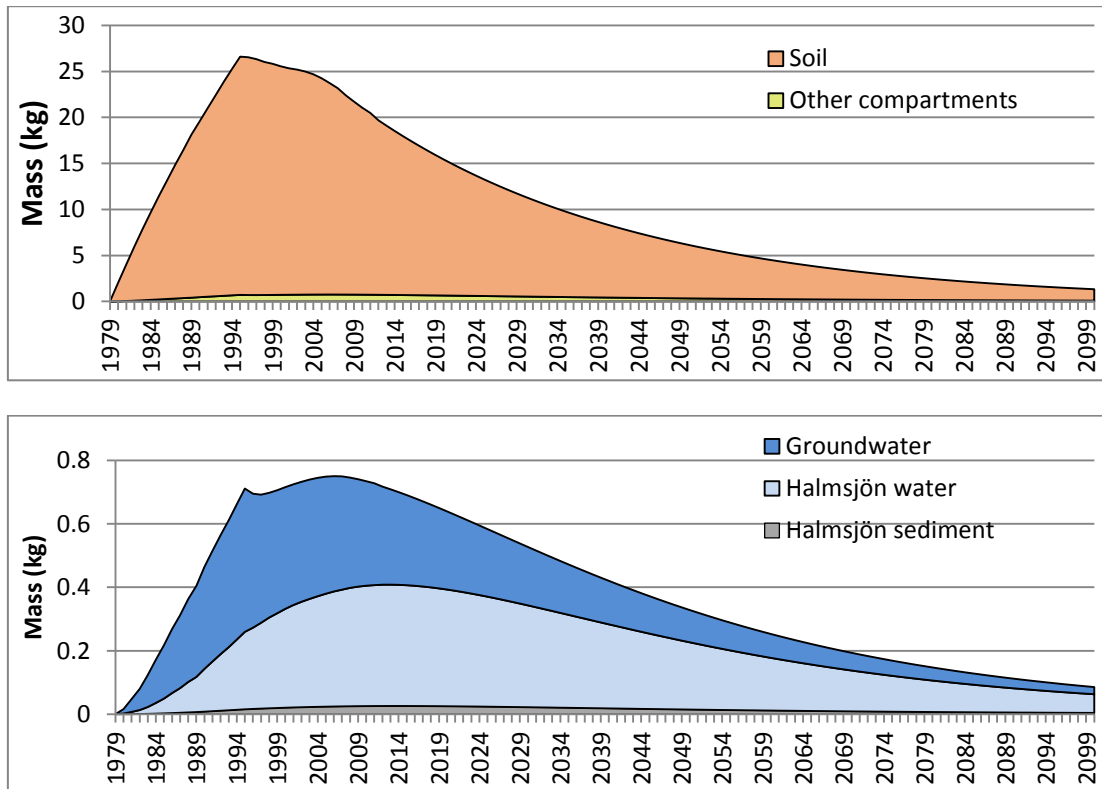


Figure 4.14. Estimated emissions of PFOS to soil (upper) and estimated atmospheric deposition of PFOS (lower) to the Arlanda area in the time period 1980 - 2020. Note the different scales on the y-axes.

#### 4.6.2 Accumulated amounts of PFOS in the Arlanda area

The model predictions indicate that the accumulated amount of PFOS were highest in the middle of the 90's, when 27 kg were estimated to be present in the area (in soil and water), after which the amounts are predicted to have decreased by 0.5 – 1 kg/per year. 20 kg is estimated to remain in the area in the year 2012, and predominantly reside in soil (Figure 4.15). Only 1.7 % is predicted to reside in the groundwater, and about the same fraction (1.9 %) in the water of Lake Halm sjön. The estimated amounts in the small streams and in the pond system are so small that they have been excluded from the figure.





**Figure 4.15. Estimated amounts of PFOS in soil (upper) and other compartments (lower) in the Arlanda area and their changes over time. Note the different scales on the y-axes.**

The overwhelming majority of all PFOS is estimated to remain in the soil and corresponds to about 50% of the total amount used at the airport. Earlier studies made in Norway have estimated that about 10 - 40% of the PFOS used in firefighting foam still resides in local soil (SFT, 2008). Axelström (2010) estimated the total amount of PFOS in the groundwater aquifer at the fire drill site to 4 kg, which is a factor of 12 higher than our estimates for 2010 of 0.33 kg in groundwater.

The distribution of PFOS between water and sediments in Lake Halmsjön is estimated to be 15:1. The amount of PFOS in the sediments of Lake Halmsjön (top 20 cm) has previously been estimated by Vectura (Axelström, 2010) to 0.08 kg in February 2010, and 0.21 kg in September the same year. This is 4-10 times higher than our model estimated amounts from the year 2010 of 0.025 kg.

Figure 4.16 shows a snapshot of the model predicted flows and amounts in 2012. The percentage distribution between the compartments in the area are:

Soil (96%) > Lake water (1.9%)> Groundwater (1.6 %) > Lake sediments (0.13 %) > Pond water (0.016 %) > Stream water (minor)

Due to the dynamics of the system (i.e. non-steady state), there is no exact mass balance for a single year. Therefore, for example, the outflow from the soil exceeds the inflow in 2012, which also reflected in the declining concentrations and quantities over time.

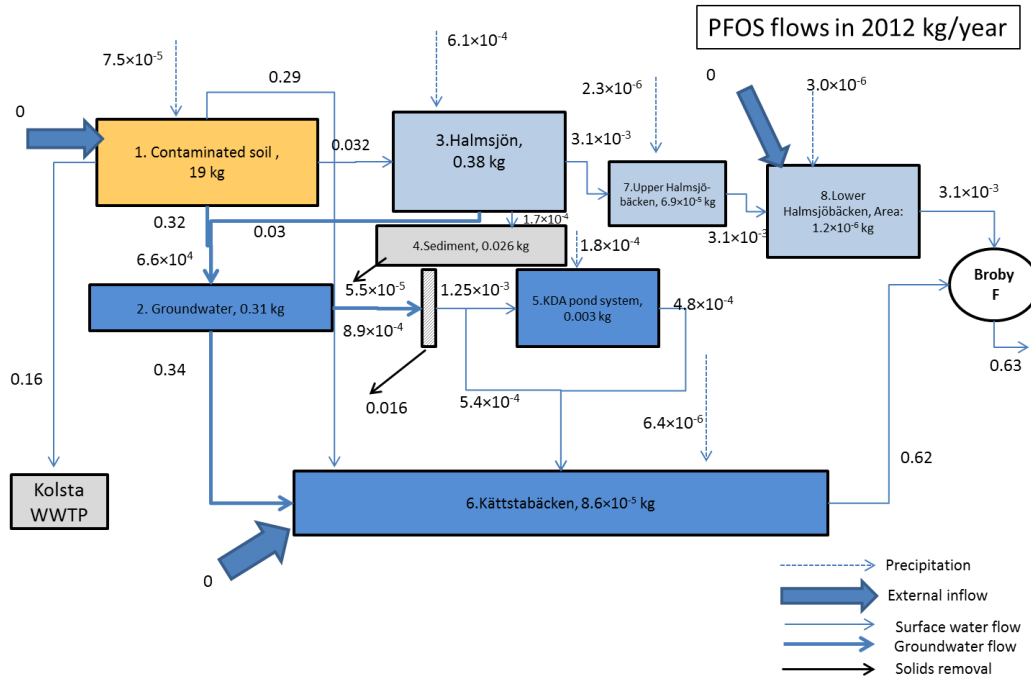


Figure 4.16. Model predicted amounts and flows of PFOS in the Arlanda airport area in 2012.

The estimated annual transport of PFOS out of the area is shown in Figure 4.17, assuming high cleaning efficiency of the carbon filter (90%). The predicted annual transport out to river Märstaån via Broby F (A6) was highest (~0.9 kg) in the late 90th century. In 2012, the transport to river Märstaån was about 0.6 kg according to these calculations, which is within a factor of 2 of the estimated outflow based on measured concentrations and flows at Broby F (see 4.4).

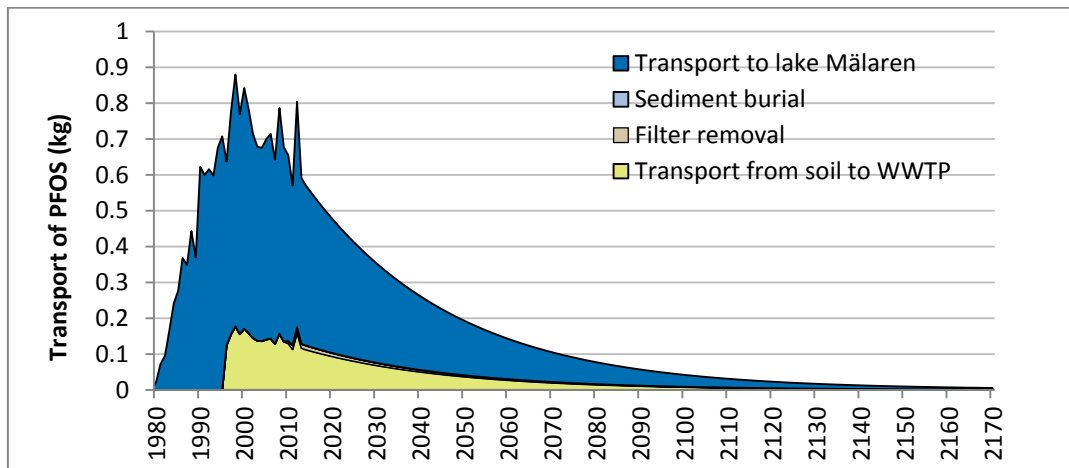


Figure 4.17. Estimated annual transport of PFOS out from the Arlanda area.

### 4.6.3 Comparison between predicted and measured concentrations

To evaluate the model predictions, the ratio between modeled and measured concentrations were calculated for the compartments where empirical data were available, i.e. soil, groundwater, surface water and sediments. Monitoring data were obtained from the summary report by Vectura (Axelström, 2010) and from annual reports from RE-PATH. Since measured concentrations often span over several orders of magnitude

(e.g. groundwater concentrations between 0.6 and 170 000 ng/L), median values were used in the calculation of ratios. Min and max values were also calculated based on the highest and lowest measured concentration of that year. The ratios are presented in Figure 4.18. As shown in the Figure the model

calculations compare relatively well with the measured concentrations of lake water (factor 1.7-3.2), for Kättstabäcken (factor 3.7-8.1) and for groundwater in 2009 (factor 1.2). In the other cases, the calculated concentrations deviate from measured by between a factor 22 for the stream Halmsjöbäcken and a factor of 4500 for soil. The comparison is hampered somewhat because the time series for each compartment is not complete. The large disagreement between modelled and measured concentrations in soil may be due to unrepresentative soil data (i.e. the most contaminated areas have not been sampled), or an underestimation of the soil volume in the model.

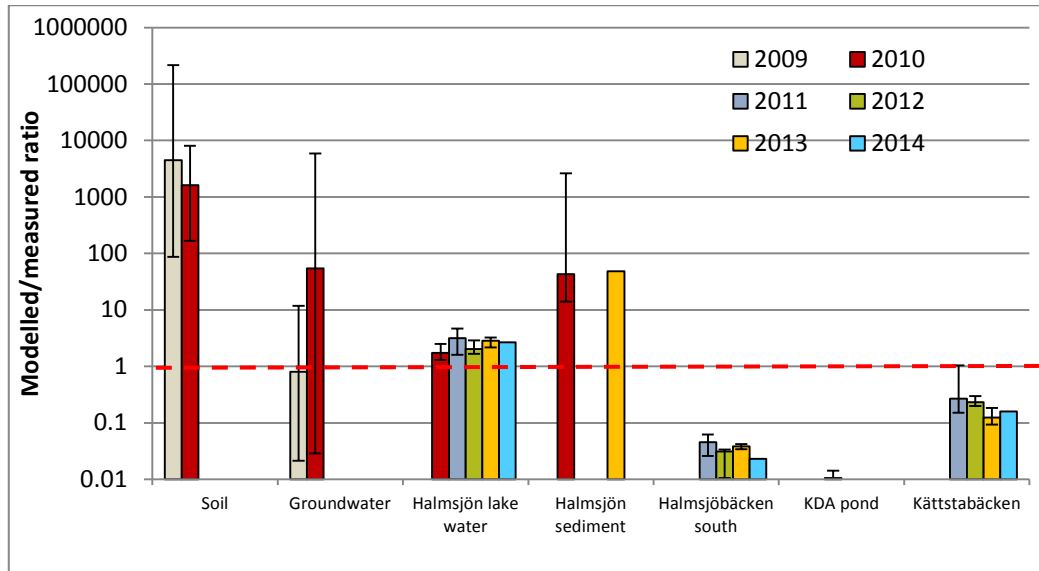


Figure 4.18. Comparison between modeled and measured median concentrations in different matrices in the Arlanda area between the years 2009 - 2014. The red dashed line shows the ratio 1, i.e. perfect agreement between modeled and measured concentrations. The error bars represent the min and max ratios, calculated based on the highest and lowest measured concentration a given year.

#### 4.6.4 The development of the concentrations in Lake Halmsjön over time

The predicted concentrations in Lake Halmsjön agree with the measured levels by about a factor of 2 as shown in Figure 4.18. Moreover, the predicted time trend agrees well with the monitoring data, indicating that the PFOS concentrations in Halmsjön may currently be at its peak level and that a gradual decrease is to be expected in the following years (Figure 4.19). With the predicted decrease rate, the concentration will have reached half its current level in the year 2055. Assuming that no particular remediation actions take place, the water concentrations will not have reached background levels (<2 ng / L) within the simulation period of >200 years even if the apparent overestimation by the model is taken into account (3.4 ng/L in the year 2260).

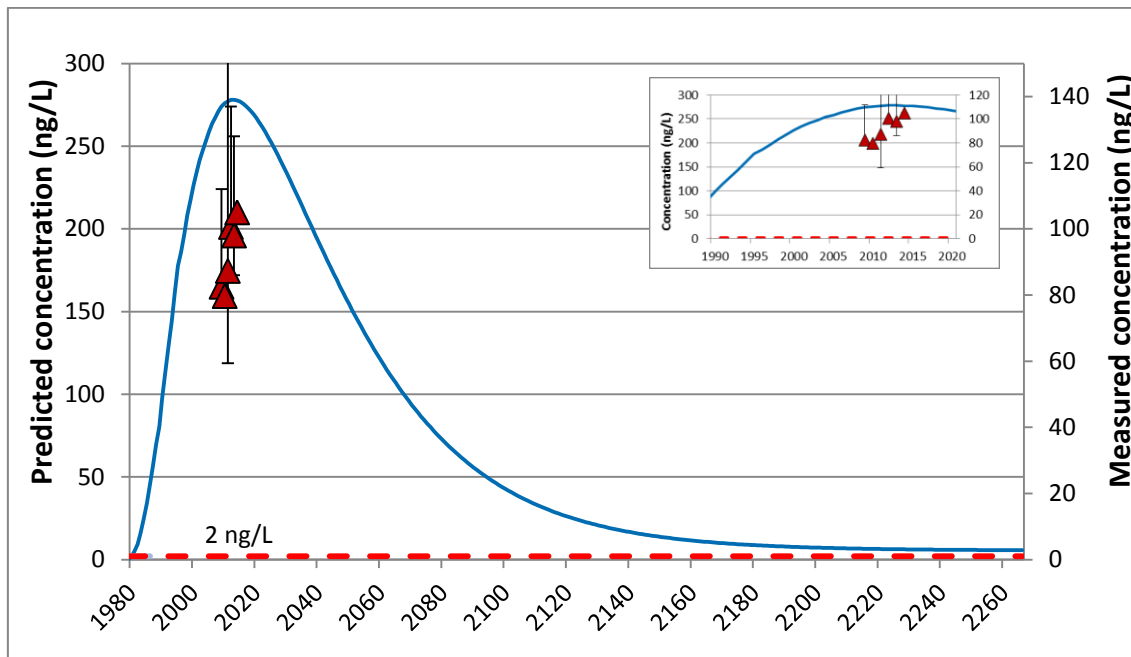


Figure 4.19. Model predicted concentration of PFOS in the bulk water of lake Halmsjön and the estimated change over time. Note the different scales on the two y-axes. The small picture to the upper right presents a close-up of predicted and measured concentrations in a shorter time span (1990-2020).

## 4.7 Implications for exposure to humans and wildlife

### 4.7.1 Contamination of drinking water

One of the major concerns of PFAS is that their extreme persistence combined with their specific binding properties, cause them to follow water streams rather than stick to solids (in fact, they do both). This has resulted in contamination of groundwater in the vicinity of firefighting areas where they have been used in excess. In some of these areas (e.g. Kallinge and Uppsala), where groundwater is used as the main source for drinking water, elevated concentrations have been observed in drinking water, leading to high exposure of humans, especially young children (Jakobsson et al., 2014).

In the case of Arlanda and Landvetter, however, there are no large public drinking water sources in the close proximity to the contaminated area, and the few private groundwater wells that are located near the two airports have been controlled, showing only background levels (<1.5 ng/L, Swedavia, pers comm). The nearest public source of drinking water in the Arlanda area is Görvelns drinking water plant, located in north Mälaren. Although significant amounts of PFAS enter Lake Mälaren from multiple sources each year, the concentration reported by Görvelns drinking water plant (6 ng PFOS/L) is similar to background levels found in drinking waters in Sweden (Holmström et al., 2014) and Europe (Ullah et al., 2011).

### 4.7.2 Exposure to humans

The current value for tolerable daily intake (TDI) for PFOS is 150 ng/kg body weight and day (EFSA 2008). The Swedish National Food Agency has, based on typical levels (background levels) of PFOS in drinking water and food items, estimated the average daily intake (ADI) of PFOS for an adult person, to 0.6 ng/kg body weight and day with 71% of the contribution from food and 29 % from drinking water, (Livsmedelsverket 2014). A health risk characterization of PFAS in the Swedish population was performed in 2013 by Borg et al., which showed no concern for hepatotoxicity or reproductive toxicity in the general population.

The human exposure of PFOS with different exposure has been estimated based on data from Livsmedelsverket 2015 and the RE-PATH project. If the food and water sources are contaminated with higher PFOS levels than normal background, the daily exposure obviously increases. Table 4.1 illustrates estimated ADIs for individuals exposed to contaminated fish or water. In these examples it is assumed that the anglers would eat 1 portion contaminated fish per week, thus a very high consumption from these particular lakes. Adults exposed to high PFOS-levels in drinking water are still below the current safety levels, as long as they don't also eat contaminated fish. It is clear from the Table, that even a very high consumption of contaminated fish from Lake Halmsjön or Västra Ingsjön, would not lead to exposure exceeding the TDI. Such high consumption of freshwater fish is not recommended anyway, because of the high concentrations of mercury. However, if that individual is also exposed to contaminated water, e.g. by drinking water from a contaminated groundwater well, it is likely that the TDI would be exceeded. The most vulnerable human individuals are young babies fed on formula mixed with water from contaminated groundwater, such as in the area of Kallinge, Ronneby municipality in Blekinge, where high PFOS levels were detected in 2013. They would exceed the TDI by more than a factor of 3.

**Table 4.5. Estimated human exposure to humans with different food consumption patterns. ADI exceeding current TDI values are marked in red.**

Individual	Consumption	PFOS concentration	ADI (ng/kg bw day)
Average Swede	Background intake (food and drinking water)		0.6
Angler, Arlanda area	Background intake + Fish from Lake Halmsjön one portion per week	300 ng/g fw	94
Angler, Landvetter area	Background intake + Fish from Lake Västra Ingsjön once portion per week	40 ng/g fw	13
Adult, Kallinge	Background intake + Drinking water from Kallinge, 2 L per day	4000 ng/L	110
Formula fed baby, Kallinge	Drinking water Kallinge, 0.8 L per day (baby 4 month, 6.6 kg b.w.)	4000 ng/L	480

### 4.7.3 Exposure to fish and fish eating mammals

It thus appears as if human exposure to PFOS even in highly exposed areas is on the safe side, with the exception of formula fed babies in contaminated areas. What about other animals?

As mentioned in chapter 4.3.1, even if the fish in Lake Halmsjön are exposed to high levels of PFOS, no signs of dysfunctionality are shown yet. However, other studies have indicated that a high body burden of PFOS in fish in relation to the total body weight may have an effect on the physiological functions of the fish in the longer term. For example, it has been shown that PFOS can be transferred to developing embryos in zebrafish (*Danio rerio*) (Sharpe et al., 2010), where a chronic exposure to PFOS can have a negative effect on reproduction, embryonic growth, and subsequent offspring development in zebrafish (Wang et al., 2011b; Du et al., 2009). In addition, a PFOS-induced toxicity has been observed in gills in European bullhead (*Cottus gobio*) (Dorts et al., 2011) and in gonads in the female zebrafish (Du et al., 2009). Thus long-term effects on local fish cannot be excluded, although the current health status of fish in Lake Halmsjön appears to be good.

There are however, predators that live more or less exclusively of fish as a diet and even if the fish themselves are of good health status, higher organisms might be at risk due to biomagnification. Furthermore, such organisms do not distinguish between tissues, but eat the whole fish, which contains higher amounts of PFAS than the pure muscle normally consumed by humans (see Figure 4.9). High levels of PFAS have been observed in Scandinavian otters showing an increasing trend during the time period 1972-2011, up to 16000 ng/g f.w. (Roos et al., 2013). Persson et al. 2013 compared levels of PFAA in mink

liver from different areas in Sweden, and showed that minks from Märsta, in the vicinity of Stockholm Arlanda Airport, contained 4-20 times higher levels compared to the other regions that were examined.

## 5 Final remarks

The five year long monitoring programme of PFAS concentrations together with the ecotoxicological studies and assessment of fate and exposure conducted within the RE-PATH project has led to a number of specific conclusions:

- Firefighting training sites are significant point sources contributing to environmental contamination of PFAS in Sweden
- Individual firefighting training sites contribute only by a small fraction of the total load of PFAS to the environment, but can locally cause elevated concentrations of PFOS in fish and water by up to a factor of 100 in comparison to reference areas
- The current contribution of PFOS and PFAS from Stockholm Arlanda Airport to Lake Mälaren is 1.3 kg and 2.4 kg/year respectively, and 0,9 and 1,3 kg/year respectively from Göteborg Lanvetter Airport (reference year 2013).
- No risk for human health effects caused by intake of PFOS via water or fish has been identified in the studied areas, using the current level of TDI (150 ng/kg bw).
- No acute ecotoxicological effects on local fish (perch) have been observed, at the prevailing conditions.
- The concentrations of PFOS in the local environment surrounding Stockholm Arlanda Airport are expected to decline slowly. If no measures are taken, background levels will not be reached in the near future
- A risk for secondary poisoning of species higher up in the food chain such as fish eating species cannot be excluded

Although no acute effects of PFOS in fish have been observed in the current study at environmental levels, it cannot be excluded that effects could be observed after long-term exposure. To investigate this, more long-term studies are required.

One of the major concerns of PFAS for humans today is associated with contamination of groundwater, which leads to pollution of drinking water. Individuals who are exposed to contaminated drinking water may exceed the levels of consumption that are considered safe. Thus, it is of utmost importance to ensure that existing contaminated areas do not result in such pollution now, or in the future. For this, mapping of PFAS concentrations at contaminated areas as well as their surroundings is needed, and there is a need to establish site-specific limit values for soil and groundwater depending on local conditions.

Even if the contribution of local sites to the overall contamination of PFOS on a larger scale is small, all sources together may still add up to important contributions in total. A consistent regulatory practice, on all potential sources is therefore important.

Even more important is to learn from experience. The main reason for the problems we see today is the extreme persistence of some of the PFAS, and the fact that most of the non-persistent PFAS degrade to form extremely stable compounds, combined with their tendency to be transferred to groundwater via their specific properties, which makes it extremely challenging to implement cost-effective remediation techniques. In many of the current AFFFs the fluorinated content comprises precursor compounds to perfluorinated alkylacids (PFAA), which are released to soil and groundwater, where they have been shown to degrade to persistent PFAAs (Houtz et al., 2012, Houtz et al., 2013). Thus, just removing the persistent

substances themselves from the products does not necessarily prevent environmental contamination of PFAAs from firefighting foams.

Since October 2014 several PFAS (PFDS, PFDA, PFNA and PFHxS) have been added to the 'SIN list' ([www.chemsec.org](http://www.chemsec.org)), which is a concrete tool to speed up the transition to a world free from hazardous chemicals. The chemicals on the SIN List have been identified by ChemSec as Substances of Very High Concern based on the criteria established by the EU chemicals regulation REACH.

This illustrates that the problem with environmental contamination of PFAS is not limited to PFOS in AFFF, but concerns the entire chemical group, which all have multiple application areas.



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## 7 References

- Ahrens, L., Norström, K., Viktor, T., Cousins, A. and Josefsson. (2015) Stockholm Arlanda Airport as a source of per- and polyfluoroalkyl substances to water, sediment and fish. *Chemosphere, In press*
- Ahrens, L., Ribéli, E., Josefsson, S., Gustavsson, J., Nguyen, M. and Wiberg, K. (2014) Screening av perfluoroalkylerade ämnen och flamskyddsmedel i svenska vattendrag. Rapport Naturvårdsverket NV-2213-13-026.
- Andersson H, Cousins AP, Brorström-Lundén E, Wickman T, Pettersson M, Holmström K, et al. 2012. Summary report SWEDEN Work package 4: Identification of sources and estimation of inputs/impacts on the Baltic Sea, COHIBA Project Consortium. [http://www.cohiba-project.net/sources/results/en\\_GB/reports/](http://www.cohiba-project.net/sources/results/en_GB/reports/)
- Arias, V., Mallavarapu, : and Naidu, R. (2015) Identification of the source of PFOS and PFOA contamination at a military air base site. *Environ Monit Assess*, 187, 4111.
- Award, E., Zhang, X., Bhavasar, S., Petr, S., Crozier, P., Reiner, E., Fletcher, R., Tittlemier, S., Braekevelt, E. (2011). Long-term environmental fate of perfluorinated compounds after accidental release at Toronto Airport. *Environmental Science & Technology*, 45 (19), 8081–8089.
- Axelström, K. PFOS-förekomst på Arlanda flygplats. Vectura Delrapport 2010-11-09.
- Berger, U., Glynn, A., Holmström, K., Ankarberg, : and Törnkvist, A. (2009) Fish consumption as a source of human exposure to perfluorinated alkyl substances in Sweden – Analysis of edible fish from Lake Vättern and the Baltic Sea. 76 (6), 799-804.
- Borg, D., Lund., B-O., Lindquist, N-G. and Håkansson, H. (2013) Cumulative health risk assessment of 17 perfluoroalkylated and polyfluoroalkylated substances (PFASs) in the Sedish population. *Environment International*, 59, 112-123.
- Buck, R., Franklin, J., Berger, U., Conder, J., Cousins, I., Voogt, P., Jensen, A., Kannan., K., Mabury, S. and Leeuwen, S. (2011) Perfluoroalkyl and polyfluoroalkyl substances in the environment: Terminology, classification, and origins. *Integrated Environmental Assessment and Management*, vol 7, no 4, 513-541.
- D'Agostino, L. and Mabury S. (2014) Identification of novel fluorinated surfactants in aqueous film forming foams and commercial surfactant concentrates. *Environmental Science and technology*, 48, 121-129.
- Defoort, C., Lindberg. M, Woldegiorgis, A. (2012). Rapport: PFOS Tullinge grundvattentäkt – Nulägesanalys Slutrapport. WSP Uppdragsnummer: 10158302
- de Solla, S.R., De Silva, A.O., Letcher, R.J. (2012). Highly elevated levels of perfluorooctane sulfonate and other perfluorinated acids found in biota and surface water downstream of an international airport, Hamilton, Ontario, Canada. *Environment International*, 39, 19-26.
- Dorts, J., Kestemont, P., Marchand, P.A., D'Hollander, W., Thézenas, M.L., Raes, M., Silvestre, F., 2011, Ecotoxicoproteomics in gills of the sentinel fish species, *Cottus gobio*, exposed to perfluorooctane sulfonate (PFOS). *Aquat. Toxicol.* 103, 1–8.
- Du, Y., Shi, X., Liu, C., Yu, K., Zhou, B., 2009, Chronic effects of water-borne PFOS exposure on growth, survival and hepatotoxicity in zebrafish: A partial life-cycle test. *Chemosphere* 74, 723–729.

EFSA. 2008. PFOS, PFOA and their salts. Scientific opinion of the Panel on Contaminants in the Food Chain. The EFSA Journal 653, 1-131.

FFFC (2006). AFFF update. [www.ffc.org](http://www.ffc.org)

Filipovic, M., Berge, U. and McLachlan, M. (2013) Mass Balance of Perfluoroalkyl Acids in the Baltic Sea. Environmental Science and Technology, 47(9), 4088-4095.

Filipovic, M., Woldegiorgis, A., Norström, K., Bibi, B., Lindberg, A. and Österås, A. (2015) Historical usage of aqueous film forming foam: A case study of the widespread distribution of perfluoroalkyl acids from a military airport to groundwater, lakes, soils and fish. Chemosphere In press

Herzke, D., Olsson, E., Posner, P. (2012) Perfluoroalkyl and polyfluoroalkyl substances (PFASs) in consumer products in Norway A pilot study. Chemosphere, 88, 980-987.

Hetzer R.H., Kümmerlen F., Wirz K., Blunk D. (2014). Fire Testing a New Fluorine-free AFFF Based on a Novel Class of Environmentally Sound High Performance Siloxane Surfactants Proceedings of the eleventh international symposium IAFSS  
<http://www.iafss.org/publications/fss/11/42/view>

Holmström, K., Wetterstrand, and Hedenberg, G. (2014) Nationell screening av perfluorerade föreningar (PFAA) i dricksvatten. Svenskt Vatten, rapportnummer 2014-20.

Houtz, E. and Sedlak, D. (2012) Oxidative conversion as a means of detecting precursors to perfluoroalkyl acids in urban runoff. Environmental Science and Technology, 46, 9342-9349

Houtz, E., Higgins, C., Field, J. and Sedlak, D. (2013) Persistence of perfluoroalkyl acid precursors in AFFF-impacted groundwater and soil. Environmental Science and Technology, 47 (15), 9342-9349.

Jakobsson, K., Kronholm Diab, K., Lindh, C., Persson, B. och Jönsson, B. (2014). Exponering för perfluorerade ämnen (PFAS) i dricksvatten i Ronneby kommun. Rapport nr 8/2014 Arbets- och Miljömedicin – Lund, Region Skåne

KEMI 2004. Rapport 3/04 - Riskbedömning för PFOS Bilaga 3

KEMI 2006. Rapport nr 6/06 ([www.kemi.se](http://www.kemi.se))

KEMI 2014. PM 3/14 ([www.kemi.se](http://www.kemi.se))

Kissa, E., 2001. Fluorinated surfactants and repellents (2nd edition revised and expanded) (Surfactant science series 97). New York, NY.

Livsmedelsverket 2014 Glynn, A., Cantillana, T and Bjermo, H. (2013). Rapport 11. Livsmedelsverket, National Food Agency, Sweden

Livsmedelsverket 2015 [www.livsmedelsverket.se](http://www.livsmedelsverket.se)

Lundgren N, Karlsson H, 2009, "Delrapport C Perfluorerade ämnen i miljön – en bedömning av risker för människa och miljö vid f.d.Räddningsverkets skola i Rosersberg", Uppdragsnummer: 218854B, Tyréns (på Uppdrag av Avvecklingsmyndigheten).

Martin, J.W., Mabury, S.A., Solomon, K.R., Muir, D.C.G., 2003, Bioconcentration and tissue distribution of perfluorinated acids in rainbow trout (*Oncorhynchus mykiss*). Environ. Toxicol. Chem. 22, 196–204.

Mood, C. and Field A. (2000) Perfluorinated surfactants and the environmental implication of their use in fire-fighting foams. *Environmental Science & Technology*.34, 3864-3870.

Moody, C., Martin, J., Kwan, W., Muir D., Mabury, S. (2002). Monitoring perfluorinated surfactants in biota and surface water samples following an accidental release of fire-fighting foam into Etobicoke Creek. *Environmental Science and Technology*, 36, 545-551.

Moody, C., Hebert, G., Strauss, S., Field, J. (2003). Occurrence and persistence of perfluorooctanesulfonate and other perfluorinated surfactants in groundwater at a fire-training area at Wurtsmith Air Force Base, Michigan, USA. *Journal of Environmental Monitoring*, 5, 341-345.

Norström, K., Viktor, T. och Magnér, J. (2011) Årsrapport 2010 för projektet RE-PATH. Mätningar av PFAS i närområdet till Stockholm Arlanda Airport och Göteborg Landvetter Airport. IVL Svenska Miljöinstitutet, IVL rapport B1984.

Norström, K. och Viktor, T. (2012) Årsrapport 2011 för projektet RE-PATH. Mätningar av PFAS i närområdet till Stockholm Arlanda Airport och Göteborg Landvetter Airport. IVL Svenska Miljöinstitutet, IVL rapport B2060.

Norström K., Viktor, T., Cousins, A. and Benli, C. (2013) Årsrapport 2012 för projektet RE-PATH. Mätningar av PFAS i närområdet till Göteborg Landvetter Airport och Stockholm Arlanda Airport. IVL Svenska Miljöinstitutet, IVL rapport B2148.

Persson, S., Rotander, A., Kärrman, A., Bavel, B and Magnusson, U. (2013) Perfluoroalkyl acids in subarctic wild male mink (*Neovision*) in relation to age, season and geographical area. *Environ International*, 425-430.

Prevedouros, K., Cousins, I.T., Buck, R.C., Korzeniowski, S.H., 2006. Sources, Fate and Transport of Perfluorocarboxylates. *Environ. Sci. Technol.* 40, 32–44. doi:10.1021/es0512475

Roos, A., Berger, U., Dijk, J. and Bignert, A. (2013) Increasing concentrations of perfluoroalkyl acids in Scandinavian Otters (*Lutra lutra*) between 1972 and 2011: A new threat to the otter population? *Environmental Science and Technology*, 47, 11757-11765.

Sharpe, R.L., Benskin, J.P., Laarman, A.H., MacLeod, S.L., Martin, J.W., Wong, C.S., Goss, G.G., 2010, Perfluorooctane sulfonate toxicity, isomer-specific accumulation, and maternal transfer in zebrafish (*Danio rerio*) and rainbow trout (*Oncorhynchus mykiss*). *Environ. Toxicol. Chem.* 29, 1957–1966.

SFT (2008). Screening of polyfluorinated organic compounds at four fire training facilities in Norway No. 2444/2008.

Sternbeck, J., Munthe, J. och Viktor, T. 2005. Risk assessment of sedimentary mercury at Snäckviken, Södertälje channel. IVL Svenska miljöinstitutet, IVL report B1630, 2005.

Svensson, H. (2014) Sammanställning av PFOS förorening vid Svedavias flygplatser. Swedavia report. 2013-11-14, D02012-. Göteborg Landvetter Airport

Ulla, S., Alsberg, T. and Berger, U. (2011). Simultaneous determination of perfluoroalkyl phosphonates, carboxylates, and sulfonates in drinking water. *Journal of Chromatography A*, 1218 (37), 6388-6395.

United States Department of Agriculture Forest Services (1996), Evaluation of wild and fire chemicals, standard test procedures, STP 1.5 – Fish toxicity.

Vestergren, R. Thesis Human exposure to perfluoralky acids. 2011. Doctoral thesis in Applied Environmental Science at Stockholm University.

Wang, N., Liu, X., Buck, R., Korzeniowski, S., Wolstenholme, B., Folsom, P and Sulecki, L. (2011a) 6:2 Fluorotelomer sulfonate aerobic biotransformation in activated sludge of wast water treatment plants. Chemosphere, 82, 853-858.

Wang, M., Chen, J., Lin, K., Chen, Y., Hu, W., Tanguay, R.L., Huang, C., Dong, Q. (2011b), Chronic zebrafish PFOS exposure alters sex ratio and maternal related effects in F1 offspring. Environ. Toxicol. Chem. 30, 2073–2080.

Woldegiorgis, A., Andersson, J., Remberger, M., Kaj, L., Ekheden, Y., Blom, L. and Brorström-Lundén, E. (2006) Results from the National screening program 2005. Subreport 3: Perfluorinated alkylated substances. IVL Svenska Miljöinstitutet, IVL rapport B1698.

Woldegiorgis, A. och Viktor, T., Studie av perfluorerade alkylsulfonater (PFAS) i fisk och vatten från Västra Ingsjön. IVL Svenska Miljöinstitutet, IVL rapport U2308, 2008.

Woldegiorgis, A., Norström, K. och Viktor, T. Årsrapport 2009 för projektet RE-PATH. Mätningar av PFAS i närområdet till Stockholm-Arlanda Airport och Göteborg Landvetter Airport. 2009 IVL Svenska Miljöinstitutet, IVL rapport B1899, 2010.

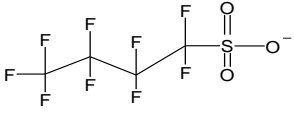
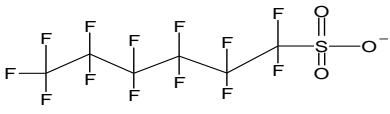
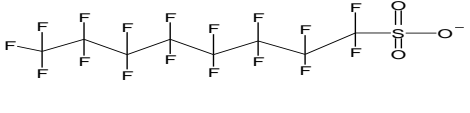
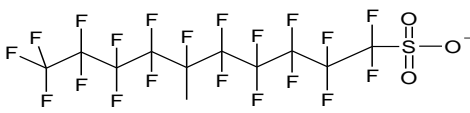
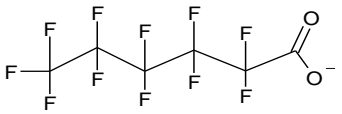
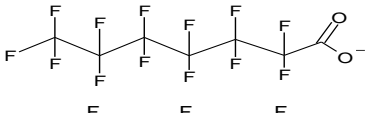
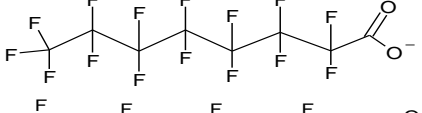
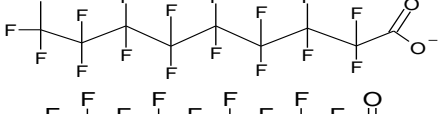
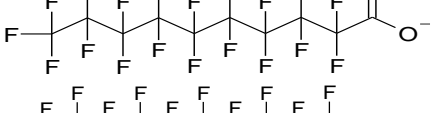
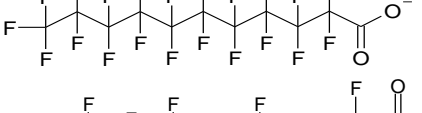
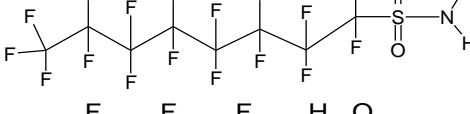
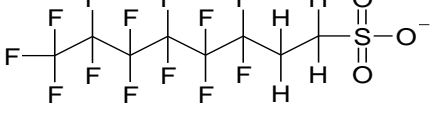
Vägverket Konsult AB, 2007. Undersökning av PFOS i sediment, jord, grund- och ytvatten. Göteborg-Landvetter Airport. Rapport Oktober 2007.

## 8 APPENDIX

### 8.1 Chemical structures

Table 8.1 shows the PFAS that have been included within the project. All compounds have been detected in different AFFF products (Herzke et al. 2012).

Table 8.1. Chemical name and structure included in the RE-PATH project.

Abbreviation	Chemical name	Structure (deprotonated form)
PFBS	Perfluorobutane sulfonic acid	
PFHxS	Perfluorohexane sulfonic acid	
PFOS	Perfluorooctane sulfonat	
PFDS	Perfluorodecane sulfonic acid	
PFHxA	Perfluorohexanoic acid	
PFHpA	Perfluoroheptanoic acid	
PFOA	Perfluorooctanoic acid	
PFNA	Perfluorononanoic acid	
PFDA	Perfluorodecanoic acid	
PFUnDA	Perfluoroundecanoic acid	
PFOSA	Perfluorooctane sulfonamide	
6:2 FTS	6:2Fluorotelomer sulfonate	

## 8.2 Methods

### 8.2.1 Sampling

Sampling of **surface waters** in the catchment areas of both airports has been performed each spring and fall every year between 2009 and 2013, see Figure 3.1 for sampling sites. The reference Lake Valloxen was sampled in 2009 and 2012. In 2011 all samples in Figure 4.1 were sampled in one day to get an overview of the PFAS distribution and concentrations at one moment.

The water was collected in clean plastic bottles and has been stored in 4 °C until extraction.

**Sediment samples** collected were stored in a freezer (-20°C) until extraction.

**Fish sampling** has been performed each year between 2009 and 2013 at Lake Halmsjön and Lake Västra Ingsjön, see Figure 3.1. At the reference lakes Sandsjön and Lake Valloxen, fishing was performed during 2009 and 2010. Fishing was performed in Lilla Issjön in 2009, and in Steningeviken 2010 and 2013.

The fish were put on carbonic acid ice during transport to the lab where they were continually stored in the freezer. Length, weight and sex were decided for all individuals. Permission was issued by the ethical board of animal testing in Gothenburg (Dnr 54/2008) and concerned all lakes included.

Muscle tissue from European perch (*Perca fluviatilis*), roach (*Rutilus rutilus*) and pike (*Esox lucius*) have been examined within this project. From several individuals of perch also blood, liver, gill and gonad tissues were used for PFAS determination.

A few **mammals and birds** were found dead within the area of Gothenburg Landvetter Airport. At 2010 a red fox and a field mouse were found, both younger than 1 year, and muscle tissue was prepared. Also a night owl was found in the same area where muscle, liver and heart were prepared for analyses.

### 8.2.2 Uptake of PFOS in goldfish

Juvenile Goldfish (*Carassius auratus*) were acclimatized at clean conditions at the lab for three month. They were fed daily with fish feed which were in advance examined not to contain PFAS. 80% of the water was exchanged three times a week.

Ten individuals of Goldfish were placed in a cage in Lake Halmsjön and were fed with same food as they were at the lab. They were not able to feed from the bottom of the lake. The individuals were sampled after 1,2,3 and 10 weeks of exposure in the lake. After each sampling the fish was transported to the lab where they were put down and the muscle tissue were removed for analyses of its PFOS content. A control group were held in a tank situated nearby equipped with a continuous flow of natural water with PFOS concentrations below detection limits.

### 8.2.3 Outgoing amounts

During May 2013 to June 2014 (April and May missing), surface water were sampled each month at Göteborg Landvetter Airport (site LO, Utloppet), and at Stockholm Arlanda Airport (Broby (A6) and Märstaån (A7)) from March 2013 to April 2013. At the same time also three locations in the Uppsala region (Fyrisån Vindbron, Fyrisån Klastorp and Vendelån Lena kyrka). In the project only data from Fyrisån Vindbron have been used.

Data of the water flow at the sampling sites was provided by Swedavias own measurements at the sites LO (Utloppet, Landvetter) and site A6 (Broby F, Arlanda). At the site A7 (Märstaån), a flow meter was placed.

Flow data from Uppsala (Fyrisån) was provided by the Swedish University of Agriculture Science. All data are shown in Table 8.2.

**Table 8.2. Concentrations and flow measurements at Broby (Arlanda), Utloppet (Landvetter) and Fyris (Uppsala) during 2013-2014.**

RE-PATH cod			PFOS (ng/L)	$\Sigma$ PFAS <sub>12</sub> (ng/L)	Flow on Date (L/s)	Flow Mean of the month (L/s)
699	Broby	2013-03-22	196	423	90.0	52.4
700	Broby	2013-04-13	153	283	634	721
685	Broby	2013-05-31	270	487	188	58.8
658	Broby	2013-06-04	337	589	86.0	29.3
701	Broby	2013-07-31	182	231	52.4	190
690	Broby	2013-08-30	140	257	92.0	82.1
703	Broby	2013-09-13	221	389	86.3	52.4
692	Broby	2013-09-30	181	363	83.8	61.2
693	Broby	2013-11-14	136	244	221	249
741	Broby	2013-12-23	161	348	214	202
742	Broby	2014-01-30	196	468	288	128
768	Broby	2014-02-26	284	542	404	495
769	Broby	2014-03-23	357	620	422	614
657	Utloppet	2013-05-22	296	376	94.3	
681	Utloppet	2013-06-11	358	445	22.5	
682	Utloppet	2013-07-11	412	510	44.8	
683	Utloppet	2013-08-13	257	328	105	
674	Utloppet	2013-08-29	366	457	31.9	
695	Utloppet	2013-09-10	308	409	43.3	
696	Utloppet	2013-10-14	286	389	56.4	
697	Utloppet	2013-11-19	273	428	129	
698	Utloppet	2013-12-11	166	225	216	
763	Utloppet	2014-01-15	295	482	137	
764	Utloppet	2014-02-17	209	341	246	
765	Utloppet	2014-03-12	319	491	163	
782	Utloppet	2014-06-23	283	389	84	
732	Fyrisån Vindbron	2013-03-13	6.02	10.8	4320	4238
735	Fyrisån Vindbron	2013-04-17	4.65	7.96	33800	30927
737	Fyrisån Vindbron	2013-05-16	3.12	7.77	13800	17368
709	Fyrisån Vindbron	2013-06-14	7.96	22.4	3350	3246
712	Fyrisån Vindbron	2013-07-18	11.9	36.8	1490	1611
715	Fyrisån Vindbron	2013-08-12	25.8	56.1	1240	1194



RE-PATH cod			PFOS (ng/L)	ΣPFAS <sub>12</sub> (ng/L)	Flow on Date (L/s)	Flow Mean of the month (L/s)
718	Fyrisån Vindbron	2013-09-18	23.2	53.2	990	1007
721	Fyrisån Vindbron	2013-10-15	15.9	46.7	1130	1365
724	Fyrisån Vindbron	2013-11-12	10.7	24.2	7540	4867
727	Fyrisån Vindbron	2013-12-11	10.0	21.5	7060	10084
772	Fyrisån Vindbron	2014-01-20	6.84	22.9	12600	13620
774	Fyrisån Vindbron	2014-02-14	9.7	24.1	17300	15479
777	Fyrisån Vindbron	2014-03-18	7.62	17.5	16000	15615
780	Fyrisån Vindbron	2014-04-16	6.69	20.8	11500	10146

## 8.2.4 Extraction methods

The **water samples** were filtered using glass fiber filters (GF/C, diameter 47 mm, Whatman) prior to extraction by using Oasis WAX cartridges (Waters, 150 mg, 6 mL, 30 µm) according to ISO/DIS 25101:2009. In short, the dissolved phase was spiked with 25 ng of each of the internal standards. The GF/C filters were extracted using 5 mL methanol which were combined with the water sample. The cartridges were preconditioned with 4 mL of 0.1% ammonium hydroxide in methanol, 4 mL of methanol, and finally 4 mL of Millipore water and the cartridges were loaded with the samples at approximately one drop per second. The cartridges were then washed with 4 mL of 25 mM ammonium acetate buffer (pH 4) in Millipore water and the analytes eluted with 4 mL of methanol and 4 mL of 0.1% ammonium hydroxide in methanol.

**Fresh fish muscle samples** (1 g) were homogenized and extracted based on the solid-liquid extraction method described by Powley et al. (2005) with a few modifications (Verreault, et al., 2005). In short, aliquots of 1–2 g homogenates were spiked with 25 ng of each of the internal standards and then extracted twice with 5 mL of acetonitrile for 15 min in an ultrasonic bath. After centrifugation, the combined extract was gently concentrated by nitrogen blow down to 1 mL and cleaned using a dispersive clean up with ENVI-Carb (100 mg, 1 mL, 100–400 mesh, Supelco, USA) and glacial acetic acid (Powley, et al., 2005). A volume of 0.5 mL of the supernatant was transferred to a vial and 0.5 mL of 4 mM ammonium acetate in Millipore water was added. The extract was stored in a freezer until instrumental analysis.

The **sediment samples** were freeze dried overnight and aliquotes of 1 g soil was extracted by solid-liquid extraction method described by (Powley, et al., 2005). The sediment sample were spiked with 25 ng of each of the internal standards 5 mL 0.2 M sodiumhydroxide was added. The samples were then extracted twice with 5 mL methanol in an ultrasonic bath for 30 min. After centrifugation, the extracts were combined and 50 µL hydrochloric was added for neutralization. The extract was concentrated to 1 mL before analysis and then cleaned using ENVI-Carb as described for the fish tissue samples.

## 8.2.5 Chemicals

The target analytes included 11 PFASs. The PFCAs (C<sub>6</sub>–C<sub>11</sub>) were: PFCAs, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA) and the PFSA (C<sub>6</sub>, C<sub>8</sub>, C<sub>10</sub>) were PFHxS, PFOS, PFDS). PFOA and 6:2 FTS were also included. [<sup>13</sup>C<sub>4</sub>]-PFOS and [<sup>13</sup>C<sub>4</sub>]-PFOA were used as mass-labeled internal standards (IS). All chemicals were purchased from plus (Wellington Laboratories, ON, Canada).

## 8.2.6 Instrumentation

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 temperature of the LC column was held constant at 40 °C. Aliquots of 10 µL were injected on a C<sub>8</sub> column (50 mm x 3 mm, 5 µm particle size, Thermo Scientific, Dalco Chromtech) using a gradient of 400 µL min<sup>-1</sup> methanol and water (both with 2 mM NH<sub>4</sub>OAc). The initial gradient was set at 40/60 (v/v) methanol/water (hold for 0.5 min), then increased to 95/5 methanol/water (hold for 5 min) and then decrease to 40/60 methanol/water (hold for 8 min). The MS/MS was operated at the most sensitive transition from precursor ion to product ions in the multiple-reaction monitoring (MRM) mode. The isotope dilution method was used for quantification.

## 8.2.7 QA/QC

For all matrices, as standard procedure, laboratory blanks, method detection limits (MDLs), and recoveries were examined. Analytes were identified using retention time and two MRM transitions for each compound. The ration between the areas of the two fragment from each compound were calculated and compared with authentic reference standards. All samples were blank corrected. Method detection limits (MDLs) were calculated as 3 times standard deviation of the blank samples.

## 8.3 Sediment - water equilibrium calculations

Z-value (mol/m <sup>3</sup> Pa)	Term definition
$Z_W = 1/H = Z_A/K_{AW}$	$K_{AW}$ = air-water partition coefficient: negligible ( 1E-9)
$Z_{sed} = v_W \times Z_W + v_{DS} \times Z_{DS}$	$v_W$ , $v_{DS}$ =volume fraction water (0.91) and particles (0.09) in sediment
$Z_A = 1/RT$	R = Gas constant = 8.314 Pa m <sup>3</sup> K <sup>-1</sup> mol <sup>-1</sup> T = Temperature in K
$Z_{SS} = K_D \times Z_W \times \rho/1000$	$K_D$ = sediment-water partition coefficient =240 L/kg (Ahrens et al., 2015), $\rho$ = particle density (2400 kg/m <sup>3</sup> )
$Z_{sed} = v_W \times Z_W + v_{DS} \times Z_{DS}$	$v_W$ , $v_{DS}$ =volume fraction water (0.91) and particles (0.09) in sediment

## 8.4 Data analysis

The data used for the score plots in Figure 4.8 are shown in Table 8.3 and Table 8.4.

### 8.4.1 Principal Component Analysis (PCA)

A brief description of PCA is given here; more details are given in literature e.g. Martens and Naes, 1989. PCA decomposes a data matrix X according to:

$$X = TP^T + E$$

PCA can be considered a co-ordinate transformation from the original variable space to a model hyper-plane of much lower dimensionality that captures the variance in the data in the most efficient way. The scores, denoted t or T, are the co-ordinates in the new orthogonal co-ordinate system and thus describe the objects (here: chemical substances). The loadings, denoted p or P, describe the relation between the latent variables (principal components) that span the model space and original variables. The matrix E in the

equation above contains the residuals, i.e. the part of the data not captured by the model hyper-plane. The substantial dimensionality reduction achieved by applying PCA leads to enhanced interpretation abilities which facilitate classification and clustering of substances. PCA is not a regression method and cannot be used for finding quantitative relationships between descriptors and responses.

The interpretation of the score and loading plots are illustrated in Figure 8.1 below.

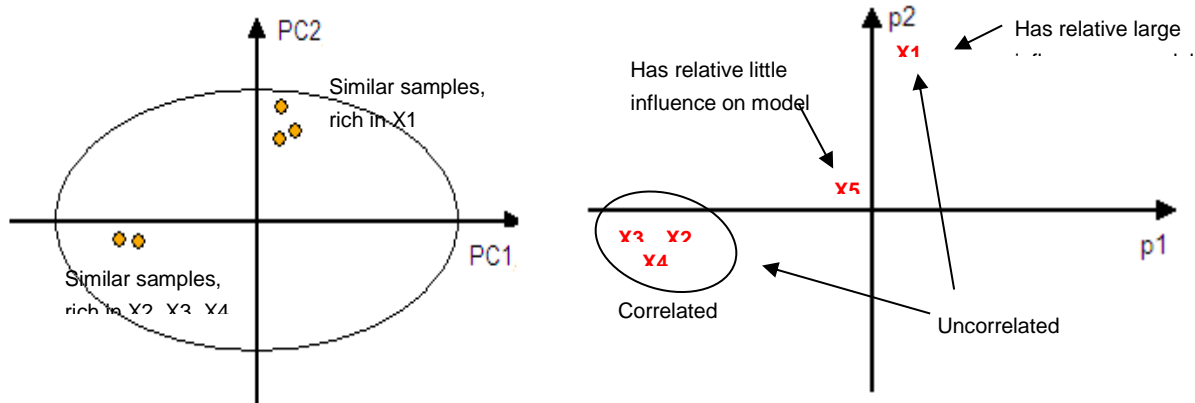


Figure 8.1. To the left an example of a score plot and to the right an example of the corresponding loading plot.

Table 8.3. Ingoing data used for the PCA score plot in Figure 4.8 for Lake Halmsjön.

RP-no	Date	Area	PFOS	PFOSA	Sex	Age
125	2009-04-30	Halmsjön	543	3.23		2
127	2009-04-30	Halmsjön	266	2.09	1	2
129	2009-04-30	Halmsjön	581	2.13	0	5
134	2009-04-30	Halmsjön	694	2.77	0	7
138	2009-04-30	Halmsjön	792	3.10	1	8
192-1	2009-04-30	Halmsjön	498	1.06	0	6
192-2	2009-04-30	Halmsjön	528	1.52	0	6
192-3	2009-04-30	Halmsjön	489	1.01	0	6
205	2009-11-10	Halmsjön	90	0.494	0	5
209	2009-11-10	Halmsjön	113	0.813	1	4
212	2009-11-10	Halmsjön	222	1.36	0	5
359	2010-04-28	Halmsjön	253	1.23	1	1
363	2010-04-28	Halmsjön	329	0.893	0	5
525	2011-09-19	Halmsjön	216			1
526	2011-09-19	Halmsjön	240			1
527	2011-09-19	Halmsjön	315			1
528	2011-09-19	Halmsjön	384	0.384		1
529	2011-09-19	Halmsjön	259		1	3
530	2011-09-19	Halmsjön	166		0	2
531	2011-09-19	Halmsjön	115		0	2

RP-no	Date	Area	PFOS	PFOSA	Sex	Age
627	2012-10-12	Halmsjön	279	0.214	0	9
634	2012-10-12	Halmsjön	273		0	11
639	2012-10-12	Halmsjön	233		0	9
667	2013-06-05	Halmsjön	447	0.477	0	8
668	2013-06-05	Halmsjön	376	0.429	0	9
669	2013-06-05	Halmsjön	219	0.646		2
670	2013-06-05	Halmsjön	121	0.523	0	3
671	2013-06-05	Halmsjön	103	0.596	0	4

Table 8.4. Ingoing data used for the PCA score plot in Figure 4.8 for Lake Västra Ingsjön, Lake Lilla Issjön and Lake Sandsjön.

RP-no	Date	Area	PFOS	PFOSA	Sex	Age
64	2009-09-09	Lilla Issjön	217	16.9	0	5
68	2009-09-09	Lilla Issjön	306	20.7	1	5
71	2009-09-09	Lilla Issjön	240	22.5	1	5
74	2009-09-09	Lilla Issjön	266	20.3	0	6
79	2009-09-09	Lilla Issjön	287	19.8	1	7
81	2009-04-17	Sandsjön	1.83		0	6
85	2009-04-17	Sandsjön	3.53	0.0198	0	8
97	2009-04-17	Sandsjön	2.96	0.0113	1	1
99	2009-04-16	V:a Ingsjön	31.0	1.70	0	2
105	2009-04-16	V:a Ingsjön	30.2	1.64	1	3
111	2009-04-16	V:a Ingsjön	39.0	0.979	0	4
323	2010-04-22	V:a Ingsjön	61.3	1.31	0	3
349	2010-04-22	V:a Ingsjön	54.6	5.04	0	3
352	2010-04-22	V:a Ingsjön	100	9.41	1	4
496	2011-09-03	V:a Ingsjön	45.2	3.86	1	3
499	2011-09-03	V:a Ingsjön	49.6	3.57	1	5
502	2011-09-03	V:a Ingsjön	46.4	1.92	0	7
507	2011-09-02	V:a Ingsjön	37.7	0.424	0	7
510	2011-09-02	V:a Ingsjön	35.2	0.0968	0	6
513	2011-09-02	V:a Ingsjön	58.2	0.350	0	7
518	2011-09-02	V:a Ingsjön	78.8	0.312	0	9
616	2012-09-10	V:a Ingsjön	210	0.724	0	1
662	2013-08-29	V:a Ingsjön	18	0.878	0	1
663	2013-08-29	V:a Ingsjön	17.4	1.75	0	1
664	2013-08-29	V:a Ingsjön	33.5	2.47	0	1
665	2013-08-29	V:a Ingsjön	31.4	2.00	0	3
666	2013-08-29	V:a Ingsjön	51.0	5.96	1	3

## 8.4.2 Trend analysis

To see if there are any trends in the concentration data between years a regression analysis has been performed for the different sampling locations and matrices. One assumption made is that the trends we are trying to detect are linear. For each location a linear regression is performed by the least square method. If the resulting parameters, in this case the slope of the regression line, is significant at a p level of 0.05 we say that there is a linear trend in the data set studied.

## 8.5 Modelling

### 8.5.1 Estimating water flows

Large efforts were devoted to estimating the water flows, especially to elucidate how the different water flows are interrelated, since these largely govern the transport of PFOS in the area. Precipitation data ( $\text{m}^3 / \text{m}^2 \text{ year}$ ) between the years 1979-2010 were taken from SMHI's database, and distributed on the surfaces that are included in the model area. Rainfall for the years 2010 onwards was estimated at the same magnitude as in 2010. According to SMHI the normal annual evaporation rate is in the range of 400 mm ( $0.4 \text{ m}^3 / \text{m}^2 \text{ year}$ ), corresponding to about 65% of the gross rainfall in 2011. This value has been assumed to apply throughout the simulated period. The other water flows were then estimated based on the net precipitation as follows (see Figure:

- Irrigation/Drill water: According to the fire chief at Arlanda, Lars Johansson, fire drills took place approximately once a month during the 80-90's, each time with a consumption of about 300 L firefighting foam (AFFF). The foam was used in a 3% mixture, corresponding to a water consumption equivalent to 9000 L per month or  $120 \text{ m}^3$  per year. This consumption pattern has been assumed to be valid for the years 1980-1996. After 1996 fire drills were conducted using lower volumes of extinguishing agents, and Lars Johansson estimates an annual consumption of about 1000 L, still in a 3% blend, which corresponds to about  $33 \text{ m}^3 / \text{year}$ . This volume has been considered to remain valid from 2002 and onwards. Only the liquids used for extinguishing have been considered for establishing water balance. Other liquids such as propane, fire drill fuel, etc. have been assumed to be used in more closed systems and therefore should not have contributed significantly to the overall water balance.
- Water collection and transportation to Kolsta WWTP: In 2012 the incoming water to the treatment plant averaged at  $1427 \text{ m}^3$  per month (measured January-September), which is equivalent to  $1.7 \times 10^4 \text{ m}^3 / \text{year}$ . It is unclear whether all of this water originates from the contaminated soil region or if the catchment area to Kolsta WWTP is larger than the surface area defined in the model. To achieve a water balance consistent with other measured flows, we have assumed that 30% of the total irrigation water (applied during fire drills) and rainfall in the contaminated area is collected, which corresponds to a volume of  $2.5 \times 10^3 \text{ m}^3 / \text{year}$  for 2011. This 30 % fraction has been assumed to be valid from the year 1996. Prior to that, the collection of water was considered to be negligible.
- Surface Water Runoff: 50% of the remaining water after collection to Kolsta treatment plant is assumed to run off as surface water partly to Halmsjön (10%) and partly to Kättstabäcken (90%). This assumption is based on the fact that the contaminated area consists of a combination of natural areas and paved surfaces. For the latter, a runoff coefficient of 0.85 has previously been suggested in the stormwater model StormTac (Karlsson, 2002).
- Groundwater flow: 50% of the remaining water after collection to Kolsta treatment plant is assumed to infiltrate into the contaminated soil and form groundwater, i.e. everything that does

not run off as surface water. For 2011, this corresponds to an annual groundwater flow of  $2.9 \times 10^3 \text{ m}^3$  or 70 mm/year. This can be compared to the estimated annual groundwater recharge determined by SWECO to 50-250 mm/year (Ekstrand et al., 2013). Thus the model estimated groundwater flow is considered to be of reasonable magnitude.

- Groundwater inflow from Halmsjön: A new groundwater model for the Arlanda area was developed in 2012 and was completed in early 2013 (Ekstrand et al. 2013). According to calculations using this model, there is a leakage from Halmsjön to the groundwater aquifer equivalent to an average of 2 L / s or  $6.3 \times 10^4 \text{ m}^3$  / year. In the model, this influx has been allocated to the groundwater reservoir with further flow to Kättstabäcken and the KDA pond system. In reality, the groundwater pathways are probably more complex.
- Pumping of groundwater: In August 2010, a pump was installed in the area at the fire drill site to pump and filter contaminated groundwater, with a pumping speed of 400 L / h or  $3.5 \times 10^3 \text{ m}^3$  / year. For 2011, this corresponds to about 5% of the annual estimated groundwater flow (see above), which is assumed to have prevailed since the middle of 2010. The pumped water is transported through an active carbon filter for purification either to the KDA pond system (70%) or directly to Kättstabäcken (30%).
- Other groundwater: Non pumped groundwater is assumed to reach Kättstabäcken (100% prior to 2010, 95% afterwards), which represents a simplified assumption about the direction of the contaminated groundwater pathways. It is likely that part of the groundwater flows out of the model system through other routes.
- External inflow from Måbydalen: According to Norling (2011) Måbydalens agricultural area is assumed to supply water to the lower part of the stream Halmsjöbäcken, corresponding to approximately 25% of the total flow at the point Broby F. Based on the measured outflow in 2011, this corresponds to an influx from Måbydalen of  $1.76 \times 10^6 \text{ m}^3$  / year. This flow is assumed to be constant throughout the years.
- External supply to Kättstabäcken: Based on the above described flows, the external inflow to Kättstabäcken adjusted to obtain consistency with the measured outflow at Broby F for the year 2011, which resulted in an estimated inflow of  $5.15 \times 10^6 \text{ m}^3$  / year.

All assumptions regarding the connections of the water flows are shown in Figure 8.2 and in Table 8.5. As an example, also resulting flows for the year 2011 are shown in Figure 8.3 Figure 7. The model has been designed so that rainfall and irrigation during fire drills constitutes adjustable input to the model, as well as the external inflows to Kättstabäcken and lower Halmsjöbäcken (runoff from Måbydalen).

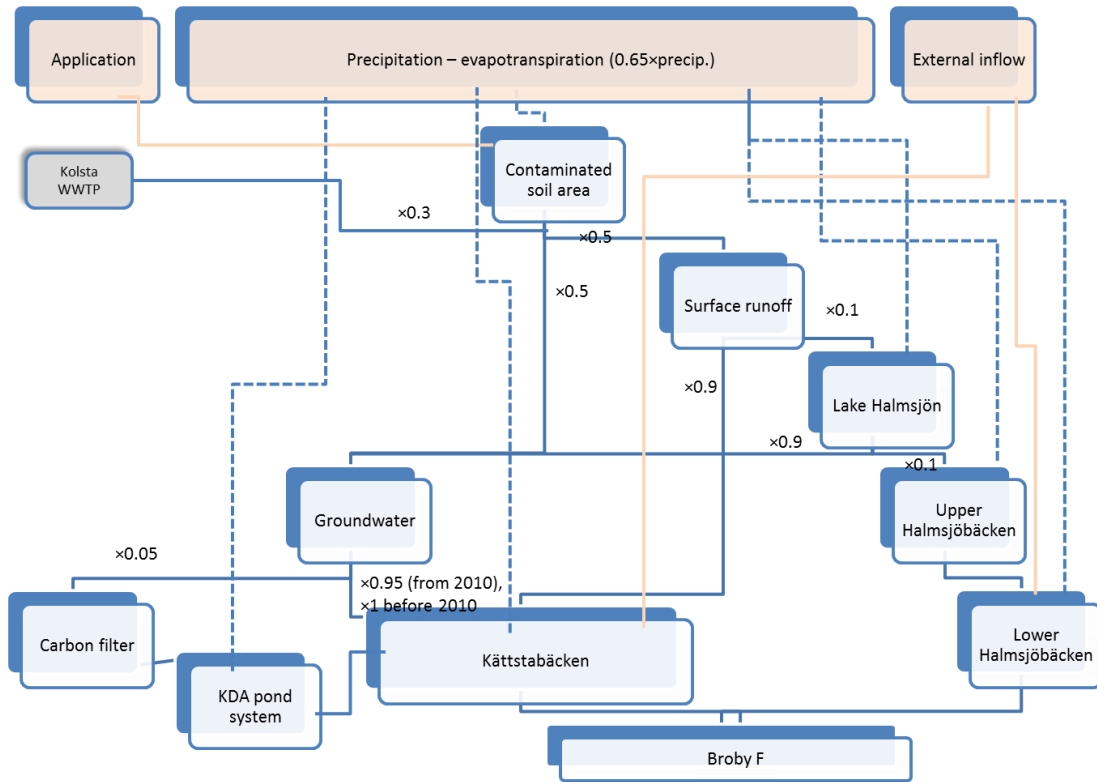


Figure 8.2. Assumptions and estimates of the connectivity of water flows in the Arlanda airport area.

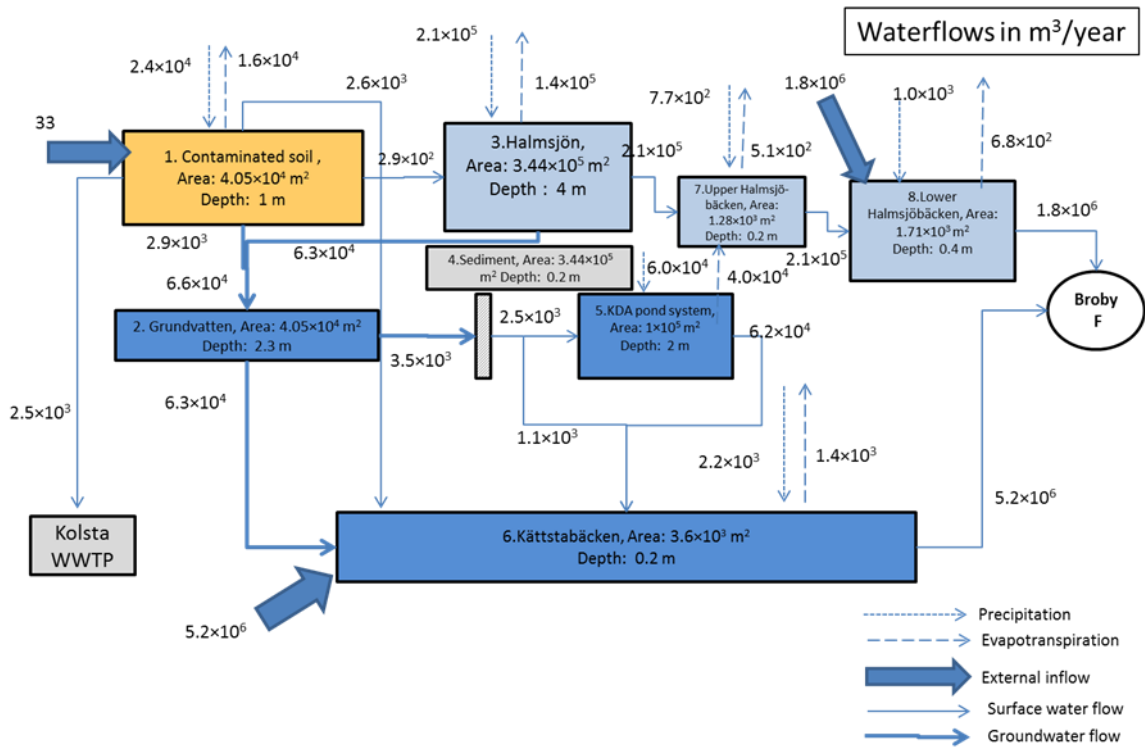


Figure 8.3. Estimated water flows in the Arlanda airport area in 2011.

**Table 8.5. Parameter values and assumptions used to quantify the water flows in the Arlanda airport area.**

Water flows	Symbol	Value	Unit	Comment/Reference
Precipitation	$U_{prec}$	0.63	$m^3/m^2$ year	SMHI's statistical precipitation register (station Arlanda)– example for year 2010
Irrigation in connection to fire drills	$U_{irr}$	108	$m^3/year$	Assuming 9000 L/month – information from Swedavia
Fraction of soil surface runoff (precipitation plus irrigation) to STP	$f_{STP}$	0.2	$m^3/m^3$	Calculated from measured inflow to Kolsta WWTP in 2012 and annual precipitation and irrigation
Fraction of external water supply [precipitation + irrigation – transport to STP] that recharges groundwater	$f_{GW}$	0.5	$m^3/m^3$	Assumed runoff coefficient of 0.85 for paved surfaces (STORMTAC).
Groundwater recharge	$U_{GW}$	10000	$m^3/year$	$f_{GW} \times ((1 - f_{STP}) \times (U_{irr} + U_{prec} \times A_{soil}))$
Fraction of soil surface runoff transported to Kättstabäcken	$f_{Kättsta}$	0.9	$m^3/m^3$	Assuming that 90 % of remaining surface runoff $((1 - f_{STP}) \times (U_{irr} + U_{prec} \times A_{soil}) - U_{GW})$ goes to Kättstabäcken
Fraction of soil runoff that goes to Lake Halmsjön	$f_{Halmsjön} = 1 - f_{Kättsta}$	0.1	$m^3/m^3$	Assumption that 10 % of remaining surface runoff $((1 - f_{STP}) \times (U_{irr} + U_{prec} \times A_{soil}) - U_{GW})$ goes to Lake Halmsjön
Fraction of groundwater to carbon filter purification	$f_{CF}$	0.05	$m^3/m^3$	Assumption that 5 % of the groundwater is pumped up for purification since August 2010
Fraction av groundwater to Kättstabäcken	$f_{GV,H}$	$1 - f_{CF}$	$m^3/m^3$	Assumption that remaining groundwater reaches Kättstabäcken
Fraction of carbon filtered water to KDA pond system	$f_{KDA}$	0.7	$m^3/m^3$	Assumption–the remaining amounts are allocated to Kättstabäcken
External inflow to Kättstabäcken	$U_{ext,Kättsta}$	4.98+06	$m^3/year$	
External inflow from Måbydalen agricultural area	$U_{ext,Måby}$	1.76E+06	$m^3/year$	
Precipitation	$U_{prec}$	0.63	$m^3/m^2$ year	SMHI's statistical precipitation register (station Arlanda)– example for year 2010
Irrigation in connection to fire drills	$U_{irr}$	108	$m^3/year$	Assuming 9000 L/month – information from Swedavia
Fraction of soil surface runoff (precipitation plus irrigation) to STP	$f_{STP}$	0.2	$m^3/m^3$	Calculated from measured inflow to Kolsta WWTP in 2012 and annual precipitation and irrigation
Fraction of external water supply [precipitation + irrigation – transport to STP] that recharges groundwater	$f_{GW}$	0.5	$m^3/m^3$	Assumed runoff coefficient of 0.85 for paved surfaces (STORMTAC).
Groundwater recharge	$U_{GW}$	10000	$m^3/year$	$f_{GW} \times ((1 - f_{STP}) \times (U_{irr} + U_{prec} \times A_{soil}))$
Fraction of soil surface runoff transported to Kättstabäcken	$f_{Kättsta}$	0.9	$m^3/m^3$	Assuming that 90 % of remaining surface runoff $((1 - f_{STP}) \times (U_{irr} + U_{prec} \times A_{soil}) - U_{GW})$ goes to Kättstabäcken



Water flows	Symbol	Value	Unit	Comment/Reference
Fraction of soil runoff that goes to Lake Halmsjön	$f_{\text{Halmsjön}} = 1 - f_{\text{Kättsta}}$	0.1	$\text{m}^3/\text{m}^3$	Assumption that 10 % of remaining surface runoff $((1 - f_{\text{STP}}) \times (U_{\text{irr}} + U_{\text{prec}} \times A_{\text{soil}}) - U_{\text{GW}})$ goes to Lake Halmsjön
Fraction of groundwater to carbon filter purification	$f_{\text{CF}}$	0.05	$\text{m}^3/\text{m}^3$	Assumption that 5 % of the groundwater is pumped up for purification since August 2010
Fraction av groundwater to Kättstabäcken	$f_{\text{GV,H}}$	$1 - f_{\text{CF}}$	$\text{m}^3/\text{m}^3$	Assumption that remaining groundwater reaches Kättstabäcken
Fraction of carbon filtered water to KDA pond system	$f_{\text{KDA}}$	0.7	$\text{m}^3/\text{m}^3$	Assumption—the remaining amounts are allocated to Kättstabäcken
External inflow to Kättstabäcken	$U_{\text{ext,Kättsta}}$	4.98+06	$\text{m}^3/\text{year}$	
External inflow from Måbydalen agricultural area	$U_{\text{ext,Måby}}$	1.76E+06	$\text{m}^3/\text{year}$	

## 8.6 References

Ekstrand N., et al. Akviferlager Arlanda – reviderad matematisk grundvattenmodell. Kalibreringar och driftsimuleringar. SWECO Rapport 2013-01-03.

Herzke, D., Olsson, E., Posner, P. (2012) Perfluoralkyl and polyfluoralkyl substances (PFASs in consumer products in Norway A pilot study. Chemosphere, 88, 980-987.

Karlsson M. 2002 Beskrivning och utvärdering av recipientmodellen i dagvattenmodellen StormTac. Examensarbete Industriellt Miljöskydd, Kungliga Tekniska Högskolan, TRITA-KET-IM 2002:22, STOCKHOLM 2002, ISSN 1402 7615.

Martens, H. and Naes, T. , *Multivariate calibration*, John Wiley and Sons, Chichester 1989.

Norling M. 2011. Märstaån – ett vattenlandskap. Är våtmarker och dammar vägen framåt? Kandidatuppsats. Geografi C. Institutionen för Livsvetenskaper, Södertörns Högskola.

Powley, C.R., George, S.W., Ryan, T.W., Buck, R.C., 2005. Matrix Effect-Free Analytical Methods for Determination of Perfluorinated Carboxylic Acids in Environmental Matrixes. Anal. Chem. 77, 6353–6358.

Verreault, J., Berger, U., Gabrielsen, G.W. 2007. Trends of perfluorinated alkyl substances in herring gull eggs from two coastal colonies in northern Norway: 1983-2003. Environmental Science and Technology, 41, 6671-6677.



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