

Article

PFAS in the Drinking Water Source: Analysis of the Contamination Levels, Origin and Emission Rates

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Abstract: Groundwater contamination caused by the use of the aqueous film-forming foam (AFFF) containing per- and polyfluoroalkyl substances (PFAS) was investigated in southern Sweden. Σ PFAS concentrations in groundwater ranged between 20 and 20,000 ng L⁻¹; PFAS composition was primarily represented by PFOS and PFHxS. The PFAS chain length was suggested to have an impact on the contaminant distribution and transport in the groundwater. PFAS profiling showed that the use of PFASs- and PFCAs/FTSAs-based PFAS-AFFF can be a contributor to PFAS contamination of the drinking water source (groundwater). PFAS emission was connected to PFAS-AFFF use during the fire-training and fire-fighting equipment tests at the studied location. PFAS emission per individual fire training was (semi-quantitatively) estimated as [1.4 < 11.5 ± 5.7 < 43.7 kg] (n = 20,000). The annual emission estimates varied as [11 < 401 ± 233 < 1125 kg yr⁻¹] (n = 1005) considering possible [2 < 35 ± 20 < 96] individual fire-training sessions per year.

Keywords: AFFF; PFAS; groundwater



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1. Introduction

PFAS-containing aqueous film-forming foam (PFAS-AFFF) has been used by firefighters for several decades [1]. PFAS-AFFF has a wide application in extinguishing hydrocarbon-fuel fires (class B fires); which is primarily due to the thermal stability of the active surfactant [2] and its ability to lower the surface tension at low concentrations [3]. From its early deployment in aircraft rescue in 1978 (Los Angeles International airport (3M Light Water)), PFAS-AFFF has become an effective and perhaps a universal solution in fire safety (in fuel fires) at airfields worldwide [1,4].

However, PFAS-AFFF use during fire-training activities, firefighting equipment tests, and emergency events can lead to contamination of the surrounding aquatic environment [5,6]. The ubiquitous occurrence of PFAS in surface water and groundwater in proximity to firefighting training locations has been reported in various studies worldwide [7–10]; and is the major historical and current source of PFAS in Sweden [11]. The source water contamination with PFAS can lead to human exposure via drinking water [12–14]. The evidence of negative health effects and cases of human exposure stresses the importance of further investigation on PFAS distribution in the environment [12,14–17]. Nearly 50% of Sweden's drinking water originates from groundwater reservoirs situated in highly permeable glaciofluvial deposits that are susceptible to contamination [11].

PFAS spread and distribution in the aquatic environment are often connected to the water solubility and persistence of the compounds [18,19]. Far-field transport conditions

have been studied in surface water [20,21] and groundwater [22,23]. Furthermore, the PFAS circulation in the urban (and/or industrial) water and waste handling cycles has been connected to the occurrence in water treatment plants [24,25] and landfills [26]. However, due to the surfactant nature of the contaminant and complex interaction mechanisms in the carrier-phase vs. media interface, the interpretation of the PFAS distribution in the corresponding transport domain requires further investigation [27–30]. Moreover, the PFAS distribution can be affected by various macroscopic conditions related to the environmental features of the area and the emission source. This can be connected to a large variation in the previously reported field-derived distribution predictors [22,31,32]. PFAS sorption and transport mechanisms in natural systems are yet subject to further investigation.

Assessment and interpretation of the water contamination by PFAS require an understanding of a cluster of interconnected processes, including analysis of the contamination levels, spatial distribution, as well as emission history. Furthermore, in contamination linked to PFAS-AFFF use, the application scenarios, equipment and purpose of the application can play a significant role in PFAS emission, as well as contamination levels and composition. PFAS-AFFF application can be continuous (historical) or related to an emergency event, thus affecting the consequent contaminant distribution [8,33]. It is often not known how and under which conditions or with which chemical speciation PFAS-AFFF has been practiced. Therefore, detailed analysis and understanding of the processes behind water contamination by PFAS and related distribution mechanisms are important.

Another important aspect in the assessment of PFAS contamination is the analytical limitations. Most of the conventional analysis methods are, in large measure, designed for quantification of the target substances, thus, despite the sufficient resolution, the detected PFAS composition is often restricted to the method [9,34]. In the contamination cases related to PFAS-AFFF, it is important to consider the presence of various structural isomers, as well as the structure of the functional group (e.g., a fraction of the branched isomers (PFOA and PFOS) can constitute 20–30% of the mass) [3,35,36].

The general purpose of the present study is to improve the knowledge in the analysis of the historical water source contamination by PFAS and address the related aspects of PFAS-AFFF application in fire-training activities and equipment tests. The conducted investigation included: (i) analysis of contamination levels and PFAS composition, (ii) assessment of the PFAS and PFAS-AFFF emission scenarios and (iii) assessment of the contaminant transport conditions.

2. Materials and Methods

2.1. Study Site

2.1.1. Area Description

The studied area is located in Ronneby Municipality (Blekinge County) in Sweden (Figure S1 in Supplementary Materials). The landscape of the area is mainly represented by hilly terrain with woods surrounding the urbanized areas. There were two main objects of interest considered in the present study. These included the F17 airfield (Blekinge Air Force Wing) and Brantafors waterworks (Ronneby Municipality). The F17 (active since 1944) has been used by both military and civil air traffic. The airfield area is restricted and surrounded by wood in the north and west, and lake Sänksjön in the north. The F17 hosts both the flight exercise area and an operational wing [37]. The Brantafors waterworks has been in operation since the 1970s and at later development stages it supplied the Ronneby Municipality with drinking water. At Brantafors, source water for municipal water supply was extracted from the groundwater reservoir at four main extraction locations: north (GW1), south (GW3 and GW4), and east (GW2) (Figure S1 in Supplementary Materials). The waterworks have been reconstructed and modified during their operation. There was limited information available for the period prior to the 1990s. Prior to 2010, due to the water quality (according to former standards), the water treatment process included aeration, pH adjustment, and UV disinfection. Brantafors was later reconstructed, and the

treatment was extended with aeration, chemical precipitation, rapid sand filtration and UV disinfection.

2.1.2. Surface Water and Hydrogeology of the Area

There are several surface water bodies in the studied area, including the Hasselstadsbäcken creek, the Klintabäcken creek, the Ronnebyån river, and lake Sänksjön (Figure S1 in Supplementary Materials). The study area is located within the larger main catchment area of "Ronnebyån". Furthermore, the area is divided into two sub-catchment areas which are divided straight through the F17 airfield. Hasselstadsbäcken originates in the wetland area south-west of Lake Sänksjön and eventually discharges into Sörbybäcken creek, a tributary to Ronnebyån. Klintabäcken originates from the wetland areas in north and east of Sänksjön and discharges into Ronnebyån. Ronnebyån flows in a south direction and discharges into the Ronnebyfjärden Bay of the Baltic Sea. Lake Sänksjön is a kettle lake (formed by the retreating glacier) and is primarily fed by groundwater and surface runoff (Figures S1 and S4 in Supplementary Materials).

The study area is located in one of the largest delta formations in South-eastern Sweden (Bredåkradeltat) which was formed during the withdrawal of the last ice age. The area is mainly covered by glaciofluvial materials consisting of sand and silt. However, an esker formation is also present (Bredåkraåsen) and it runs parallel to the Klintabäcken (Figure S1 in Supplementary Materials). The esker mainly consists of sand, gravel, and rocks. It is located directly on the bedrock and is partially covered by glaciofluvial material.

The bedrock is primarily represented by Karshamngranit (a granite) consisting of quartz, feldspar and mica; the bedrock surface is characterized by deep and long fractures in the direction from north to south (Figures S4 and S5 in Supplementary Materials).

The area contains a groundwater reservoir which is represented by the esker and coarse-grained glaciofluvial material. It is mainly an unconfined aquifer; however local confined conditions can occur due to fine glaciofluvial material. The aquifer contains local subsystems and a transient groundwater divide is formed following the runoff area of Klintabäcken, which runs along the airfield to the east. To the west, the reservoir runs towards Ronnebyån where it is confined by the bedrock which is close to the surface at this area (Figure S4 in Supplementary Materials). The coarse-grained glaciofluvial material contains a lower groundwater extraction potential of $1\text{--}5\text{ L s}^{-1}$ whereas the esker formation contains a higher potential of $5\text{--}25\text{ L s}^{-1}$. Close to Ronnebyån the potential is even higher due to the added infiltration from the river (Figure S4 in Supplementary Materials). The primary groundwater flow direction is north to south following the flow of Klintabäcken (Figures S3 and S4 in Supplementary Materials).

2.1.3. PFAS Emission Source

The PFAS emission was primarily related to the use of PFAS-AFFF during fire-training activities and firefighting equipment tests at F17. The emission sources were connected to the designated fire-training and equipment test sites on the F17 airfield territory. The main emission source was identified at the fire-training facility (FTF), located in the east of the F17 airfield (Figure S1 in Supplementary Materials). However, there are several potential sources (training and test locations) suggested by former F17 personnel, including the north-end and south-end of the airstrip and the fire station in the south of the airstrip (Figure S1 in Supplementary Materials).

At FTF, the AFFF release was primarily connected to the aircraft rescue operation training (including open fire suppression). The training was mainly performed on a designated concrete platform (Figure S2 in Supplementary Materials). Prior to 2004, the fire training was conducted by military personnel. Since 2004, the operation was taken over by the civil fire rescue services. Due to several reasons, not least confidentiality rules, the exact training protocol prior to 2004, as well as possible changes in exercise routine and location at later stages, have not fully been clarified. Therefore, the PFAS-AFFF application in fire-training and equipment test activities was considered to continue until 2014, when

the PFAS contamination was detected. Later, PFAS-AFFF applications were consequently ceased, and training are further conducted with an alternative instead.

According to former military personnel, during the period until 2004, airfield fire safety was provided by the use of a specialized emergency vehicle (heavy terrain vehicle type 4112). It was designed for fire safety and rescue missions and is commonly used in operations across the country. The main training objective included a simulation of the rapid-fire suppression and a crew rescue in the aircraft crash. The rescue training (including various activities) was carried out during one week (including several sessions per day) of the military service period and some occasional training activities throughout the period. Training with AFFF was conducted on 2 to 4 occasions a year (according to former personnel).

2.2. PFAS Measurements in Groundwater

PFAS concentration was measured in the groundwater samples collected at the extraction points GW1, GW3, and GW4. Duplicate water samples were collected directly in the well (from the water surface) in 1 L PP bottles. Samples were stored in dark at 4 °C. Prior to analysis, water samples were placed into the sonication bath for 20 min. Samples were consequently transferred into a 10 mL glass injection vial and spiked with an internal standard mix prior to analysis.

Additionally, PFAS measurements in groundwater were taken using Polar Organic Chemical Integrative Sampler (POCIS) with solid phase sorbent (Oasis HLB). The individual POCIS preparation was performed as described by Gobelius et al. [24]. Individual samplers (without cage) were set in a column configuration (Figure S6 in Supplementary Materials) and placed into the groundwater well. There were two sampler columns deployed with eight individual samplers (4 duplicates) and three individual samplers (1 triplicate), respectively. Sampler columns were deployed in groundwater extraction wells upstream (POCIS ×8) and downstream (POCIS ×3) of the groundwater aquifer at approximately 15 and 20 m depth at extraction points GW1 and GW3, respectively (Figure S4 in Supplementary Materials). For the well at GW3, a single deployment was set for 4 weeks, and at well GW1, four deployments were set for 1-to-4-week intervals. Individual samples were stored frozen prior to extraction and analysis. There were 13 samples extracted and analyzed (including 2 procedural blank samples). Sample extraction and analysis were performed according to the methods described by Gobelius et al. [24].

In total, 29 PFAS compounds were analyzed in the present study, including five perfluoroalkane sulfonates ($C_{4, 6, 7, 8, 10}$ PFASs) (PFBS, PFHxS, PFHpS, PFOS and PFDS), thirteen perfluoroalkyl carboxylates ($C_{3-13, 15, 17}$ PFCAs) (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFHxDA and PFOcDA), three perfluorooctane sulfonamides (FOSAs) (FOSA, MeFOSA and EtFOSA), two perfluorooctane sulfonamidoethanols (FOSEs) (MeFOSE and EtFOSE), three perfluorooctane sulfonamidoacetic acids (FOSAAAs) (FOSAA, MeFOSAA and EtFOSAA), and three fluorotelomer sulfonates FTSAAs (6:2 FTSA, 8:2 FTSA and 10:2 FTSA). A mix of 16 mass-labeled internal standards (IS) (i.e., $^{13}C_8$ -FOSA, d_3 -MeFOSAA, d_5 -EtFOSAA, d_3 -MeFOSA, d_5 -EtFOSA, d_7 -MeFOSE, d_9 -EtFOSE, $^{13}C_4$ -PFBA, $^{13}C_2$ -PFHxA, $^{13}C_4$ -PFOA, $^{13}C_5$ -PFNA, $^{13}C_2$ -PFDA, $^{13}C_2$ -PFUnDA, $^{13}C_2$ -PFDoDA, $^{18}O_2$ -PFHxS and $^{13}C_4$ -PFOS) was applied for internal calibration (<98% purity, Wellington Laboratories, Guelph, ON, Canada).

Duplicate samples and procedural blanks were analyzed using LC/UPLC system (Thermo Fisher Scientific, Waltham, MA, USA). A Hypersil GOLD aQ column (20 mm × 2.1 mm i.d., 12 µm particles, Thermo Fisher Scientific, Waltham, MA, USA) was used as an extraction column for online solid phase extraction. ACQUITY UPLC BEH-C18 column (100 mm × 2.1 i.d., 1.7 µm particles, Waters Corporation, Wilmslow, UK) was used as the analytical column. Injection volumes were 1.0 mL for all samples. A triple-stage quadrupole MS/MS TSQ Quantiva (Thermo Fisher Scientific, Waltham, MA, USA) was used as the detection sensor. Analysis data evaluation was performed using TraceFinder™ 3.3 software (Thermo Fisher Scientific, Waltham, MA, USA). The limit of quantification (LOQ) for the PFAS analysis

was determined as the lowest calibration point in the linear range (from 0.1 ng L⁻¹ to 2000 ng L⁻¹) if the S/N ratio was higher than 3.

2.3. PFAS Emission Estimates

There was limited information available on the AFFF use at the study site, as well as AFFF type or/and composition utilized in the past. PFAS emissions were estimated using Monte Carlo simulations and accessible historical records and accounts. The contaminant release was assessed in connection to suggested fire-training activities, equipment utilization, and AFFF release scenarios. PFAS was considered an active AFFF surfactant and its content in AFFF was estimated based on reported AFFF compositions [38]. The surfactant (PFAS) and water release per individual training session (fire-training and/or equipment test activity) were estimated according to the suggested equipment specifications and AFFF utilization scenarios. The emission scenarios were estimated as a cumulative release by individual training session scenarios per simulated period. An individual annual emission scenario was estimated from subsampled individual release scenarios considering a possible variation of the training sessions per day (k), number of exercises per period (j), and number of events per year (i):

$$PFAS_{annual} = \sum_{i=1}^i \sum_{j=1}^j \sum_{k=1}^k m_{per\ session} \quad (1)$$

Data evaluation, calculations, and analyses were performed using Microsoft Excel (Microsoft, proprietary), Argo (Booz Allen Argo, open source), and MATLAB (MathWorks, proprietary) software. There was limited information available (by the date) on an exact PFAS composition in related PFAS-AFFF formulations (Table S3 in Supplementary Materials) [34,39,40]. Although approximation on suspected PFAS composition was possible, due to the historical nature of the contamination and possible variation in AFFF types, the PFAS emission estimates were restricted to “blind” PFAS. Thus, simulation estimates are subject to uncertainty.

3. Results and Discussion

3.1. Groundwater Contamination

In the analyzed groundwater samples corresponding to extraction points GW1, GW3, and GW4, there were 12 out of 29 analyzed PFAS (PFBS, PFHxS, PFOS, PFDS, PFHpA, PFHxA, PFOA, PFNA, PFOcDA, 6:2 FTSA, 8:2 FTSA and FOSA) detected (Table S1 in Supplementary Materials). PFAS analysis indicated high groundwater contamination levels at GW1 and GW3 with \sum PFAS concentrations of 4200 ± 40 ng L⁻¹ and 20,000 ± 1900 ng L⁻¹, respectively, followed by relatively low levels at GW4 with \sum PFAS of 18 ± 3 ng L⁻¹.

In the groundwater corresponding to GW4, PFAS composition was primarily represented by PFOS (35%), PFHxS (32%) and FOSA (33%). At GW3, PFAS composition was dominated by PFOS (48%) and PFHxS (33%); followed by PFHxA (7%), PFOA (6%), PFBS (5%), and remaining compounds. Similarly, PFAS composition at GW1 was primarily represented by PFOS (72%) and PFHxS (19%), followed by remaining substances (<9%).

Based on the PFAS contamination levels, the concentration gradient is suggested to have propagated from GW1 (north) to GW3 (south), which agrees with primary groundwater flow direction from north to south (Figures S3 and S4 in Supplementary Materials). PFAS concentrations at GW1 and GW3 are suggested to indicate a unified emission source affecting the groundwater from the north.

A shift in relative composition was observed for PFASs and PFCAs (for GW1 vs. GW3) and suggestively connected to contaminant mobility. For PFCAs (PFHxA-PFHpA-PFOA), the observed composition shifted from 58-12-30% at GW1 to 46-13-41% at GW3 (downstream), whereas composition of PFASs (PFBS-PFHxS-PFOS) changed from 2-21-77% at GW1 to 5-39-56% at GW3. Furthermore, the difference in detected levels (GW1 vs. GW3) for PFDA, PFNA, 6:2 FTSA, 8:2 FTSA and FOSA is attributed to a possible effect of the

molecular chain length on PFAS transport. However, PFAS distribution in the groundwater can be affected by different transport processes (including groundwater extraction and groundwater table variation) and sorption processes related to the characteristics of the soil [22,27,41]. Therefore, until the contaminant transport conditions are sufficiently established, the suggested effect of the PFAS chain length on transport should be considered with some precaution.

Analysis of the POCIS deployed in groundwater wells at GW1 and GW3 showed a slightly better method sensitivity (Table S2 in Supplementary Materials). For PFCAs, detected in the samples corresponding to both GW1 and GW3, the observed PFAS composition was extended to PFPeA, PFHxA, PFHpA, PFOA, PFNA and PFDA. Furthermore, it was possible to conduct semi-quantitative measurements for some branched PFAS, thus extending the inventory to L-PFHxS, B-PFHxS, L-PFOS, B-PFOS, L-FOSA and B-FOSA. However, since the on-site calibration of POCIS was not possible and PFAS concentrations were estimated using the previously reported sampling rates [24], the analysis results were mainly considered for a qualitative assessment.

Overall, PFAS composition in POCIS was identical to previously measured in groundwater samples and primarily represented by PFSAs (81% (GW1) and 65% (GW3)) and PFCAs (18% (GW1) and 35% (GW3)). Based on detected PFAS composition (Figure 1) and cross-evaluation with reported PFAS-AFFF formulations (Table S3 in Supplementary Materials), there are a few possible PFAS-AFFF types in connection to the PFAS emission.

	<u>6:2 FTSA</u>			<u>8:2 FTSA</u>		
	<1%			<1%		
<u>PFPeA</u>	<u>PFHxA</u>	<u>PFHpA</u>	<u>PFOA</u>	<u>PFNA</u>	<u>PFDA</u>	
6–16%	8–11%	1–2%	2–5%	<1%	<1%	
<u>PFBS</u>		<u>PFHxS</u>		<u>PFOS</u>		
6–11%		37%		17–37%		
				<u>FOSA</u>		
				<1%		
				<u>MeFOSA</u>		
				<1%		

Figure 1. PFAS profile detected in the POCIS deployed at GW1 and GW3 (sorted left to right according to fluorinated chain length).

The PFSAs based PFAS-AFFF (often referred as Legacy foam) is suggested to be the major contributor to PFAS emission. This is in connection to the PFSAs (PFBS, PFHxS and PFOS) profile observed in groundwater samples. The PFCAs composition, on the other hand, is attributed to PFCAs/FTSAs based PFAS-AFFF and alternatively to FTSAs based PFAS-AFFF (assuming possible FTSAs to PFCAs transformation) [3,36,39]. Furthermore, traces of perfluoroalkane sulfonamide and sulfonamido substance such as FOSA and MeFOSA were considered as an indicator of the fluorination method used for the production of the corresponding PFAS. FOSA and MeFOSA can be linked to the PFAS synthesis involving electrochemical fluorination [36]. Thus, the detected FOSA and MeFOSA are associated with the material used in the production of detected PFSAs (PFBS, PFHxS and PFOS) and indirectly confirm the PFSAs-based formulation of PFAS-AFFF. PFCAs (PFPeA,

PFHxA, PFHpA, PFOA, PFNA and PFDA) detected in the groundwater samples can be attributed to electrochemical fluorination, as well as to fluorotelomer (i.e., telomerization) processes [36]. FTSA, on the other hand, can be associated with the fluorotelomer process-based origin. Assessment of the PFSA vs. PFCAs and FTSA traces, as well as analysis of the perfluoroalkane sulfonamides and sulfonamido substances (PFAS precursor compounds), are useful in the identification of the synthesis processes and related PFAS-AFFF origins [9,40,42].

PFAS-AFFF identification based on measured PFAS composition in groundwater, however, neglects possible effects of the transport and retention. Thus, the detected PFAS composition may not fully represent the actual PFAS composition in PFAS-AFFF. The assumptions on the PFCAs/FTSA based PFAS-AFFF should be considered with precaution. Further investigation is necessary to establish the transport conditions and secure the connection between PFAS in groundwater and at the emission source.

Furthermore, in the present study, PFAS analysis was conducted considering a group of target substances (29 PFAS) and, despite the quantification of some branched isomers (PFHxS, PFOS and FOSA), the detected substances do not fully represent the PFAS composition. The analytical representation (of the composition) can be fortified by use in combination with methods such as total organic fluoride (TOF) and total oxidable precursors (TOP) assay; furthermore, with the use of non-targeted analysis/screening.

3.2. Assessment of the PFAS Emission

The individual fire-training session scenarios were simulated considering possible variations in the AFFF stock solution composition, surfactant composition (PFAS content), AFFF solution (used for foam aggregation), and equipment utilization. For the AFFF stock solution composition, the prior population boundaries were assigned as $[0.5 < 0.065 \pm 0.21 < 0.9]$ for water content, $[0.1 < 0.17 \pm 0.054 < 0.24]$ for surfactant content (not restricted to PFAS), and $[0.5-1]$ as an additional parameter for the surfactant composition (or PFAS content) [34,38]. AFFF (stock solution) dilution scenarios were estimated based on reported and suggested by F17 former personnel (with a prior population set as $[0.01 < 0.03 < 0.04]$) [43]. With suggested equipment utilization scenarios, the PFAS emission per individual training session [kg] was estimated as $[1.4 < 11.5 \pm 5.7 < 43.7]$ ($n = 20,000$, average deviation (adev) = 4.5). Similarly, the release of water [L] corresponding to content in AFFF, was estimated as $[945 < 1985 \pm 594 < 3032]$ ($n = 20,000$, adev = 515). Furthermore, with suggested exercise routine, the annual PFAS emission rate $[\text{kg yr}^{-1}]$ was estimated as $[6 < 414 \pm 436 < 5462]$ (note: values above correspond to one individual simulation ($n = 20,000$)).

The annual release scenarios were evaluated based on samples from the simulated individual emission scenarios (per fire-training session). Subsampling and calculations were conducted using suggested fire-training exercise routines (including 2–6 individual sessions per day, 1–5 training days, with 1–5 occurrences per year). Consequently, 1005 annual release scenarios were evaluated (Figure 2).

The estimated annual emission scenarios varied in the range of $[11 < 401 \pm 233 < 1125 \text{ kg yr}^{-1}]$, corresponding to $[2 < 35 \pm 20 < 96]$ individual fire-training sessions per year ($n = 1005$) (Figure 2).

With further approximation, the annual PFAS emission can be estimated for the suggested most-likely exercise routine scenario as 3 consequent training days per occurrence and 2–4 occurrences per year. Thus, for the scenario with 6–60 individual sessions carried out per year (with 1–5 sessions, 3 training days, and 1–4 occurrences per year), the annual emission range is approximately 100–700 kg per year. However, since the annual emission scenarios are based on a rather limited sample, given estimates are strictly relative to the evaluated population. Further investigation is necessary to validate the estimate's accuracy.

Due to limited data available on both possible PFAS-AFFF composition and historical records of AFFF use at the studied site, it was not possible to estimate the individual PFAS emission. The estimates of PFAS release were conducted with no restriction to the actual PFAS composition in the PFAS-AFFF stock solution. Furthermore, the overall

surfactant composition in the PFAS-AFFF was considered to include fluorinated (PFAS) and non-fluorinated surfactants.

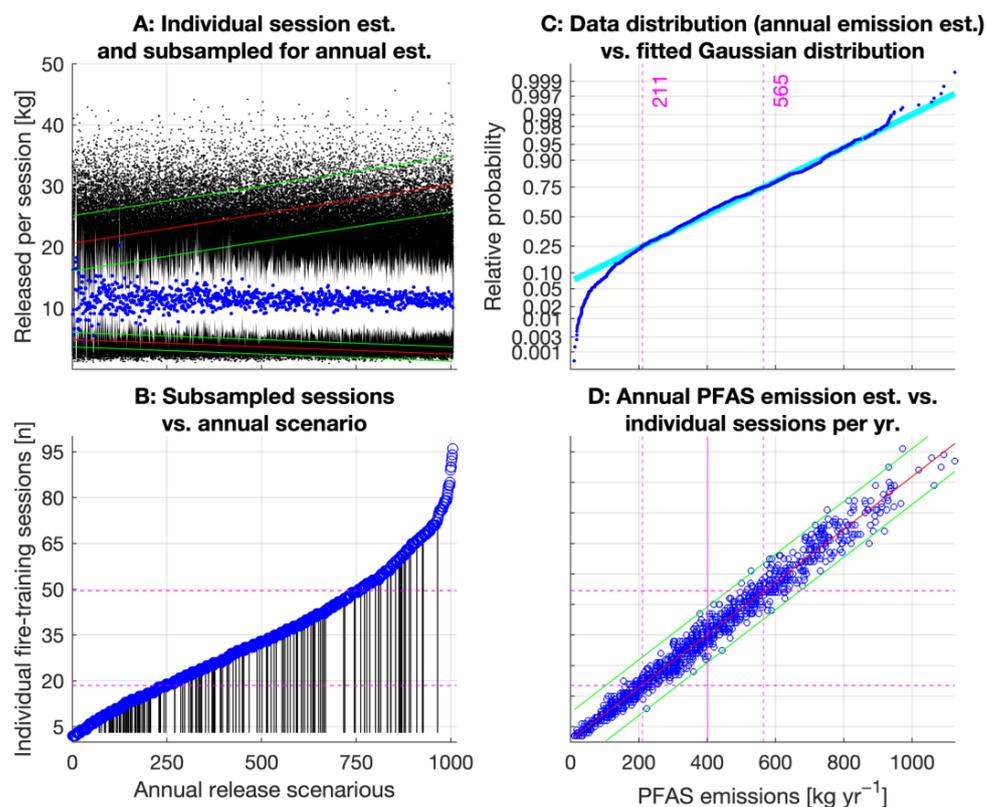


Figure 2. Evaluation the PFAS emission scenarios. (A) black dots represent simulated individual PFAS emission scenarios (corresponding to an individual fire-training session); blue dots represent the mean of the subsampled n -scenarios with white area corresponding to \pm one standard deviation; red line at top and bottom represent the maximum and minimum of the subsampled n -scenarios, respectively, with green lines corresponding to \pm one standard error. Vertical axe corresponds the variation in individual emission scenarios and horizontal axe corresponds to set of individual samples ($n = 20,000 \times 1005$). (B) blue circles indicate the number of subsampled session scenarios in A; vertical black lines correspond to suggested most-likely exercise routine with 3 consequent training days per annual exercise occurrence. (C) comparison of the distribution of the simulated annual release scenarios (blue dots) vs. fitted Gaussian distribution (cyan); vertical dashed magenta lines correspond to first and third quantiles of the data. (D) Estimates on annual PFAS emission (blue line) vs. number of individual training sessions, shown as a linear fit with green line corresponding to \pm three standard deviations; vertical magenta line corresponds to estimated median, vertical black line corresponds estimated mean, and dashed magenta lines correspond to first and third quantiles of the data.

In the present study, provided PFAS emissions were based on available records/suggestions and conducted considering a range of both PFAS-AFFF contents and application rates. However, an accurate assessment of emissions, requires a thorough clarification of the AFFF application history. This includes the PFAS-AFFF stock solution formulations, PFAS/surfactant composition in AFFF, and AFFF application routine (frequency and duration). To the authors' knowledge, this is the first study attempting to reconstruct the historical PFAS emission as in connection to PFAS-AFFF release scenarios and fire-training activities.

3.3. PFAS Transport Considerations

The PFAS emission was connected to the use of PFAS-AFFF in fire-training and equipment tests at FTF. The spatial distribution of PFAS and contamination of the aquatic environment was primarily associated with transport in the dissolved state, including transport with the surface runoff, transport with groundwater, and further distribution with raw water extraction (Figure 3).

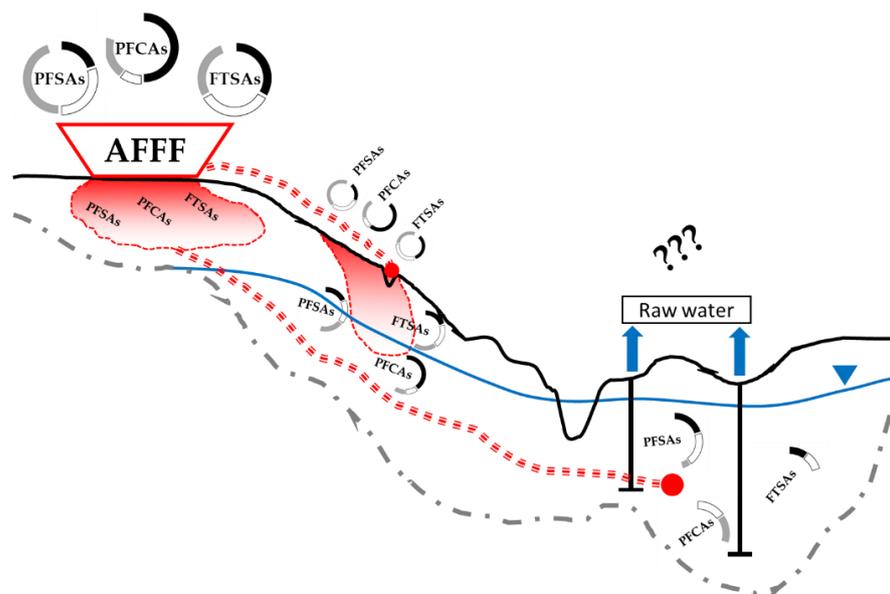


Figure 3. Conceptualization of the PFAS release, transport, and distribution at the studied site.

The initial stage of emission was associated with fire-training activities at FTF (including the simulation of the aircraft rescue mission). Primary AFFF application was connected to the dispatch of the rescue team/vehicle and putdown of an open fuel fire. According to former personnel, the exercise included an initial blast (on-vehicle) of the rescued aircraft with AFFF. Consequently, when the simulated aircraft fire is partially contained, a secondary suppression was conducted (off-vehicle) with a targeted application of AFFF. It was also suggested, that fire training sessions were possibly followed by clean-up stages where the aircraft crash simulation area was flushed with water. It is therefore assumed, that an individual training session could contribute to the generation of a substantial amount of liquid. Thus, including AFFF release, AFFF dissociation and possible use of water (in area flushes), the mass generated during the exercise session is considered sufficient for possible PFAS transport with surface runoff. The transport and distribution with runoff, however, can be affected by topographical features of the area surrounding the FTF and soil saturation. Furthermore, the PFAS transport with surface runoff is suggested to have been affected by the re-direction of surface flows with the drainage system at FTF. However, limited information was available on the drainage operation period as well as its transport capacity. It is presumed that the initial PFAS transport with surface runoff is followed by distribution processes associated with hydrological processes (distribution and retention) of much longer response time. Further PFAS distribution, associated with infiltration processes and transport in unsaturated soil, is considered as a primary contributor to PFAS accumulation and retention in the porous media, as well as to the shift in PFAS composition, prior to further advective transport. However, due to complex interaction mechanisms in the porous media, the PFAS transport in unsaturated soil is difficult to decipher with certainty, not least due to limited data available on the contaminant emission. The advective stage of the PFAS transport is primarily subject to transport in a dissolved state (with groundwater). Although the PFAS transport is assumed to primarily occur in soil of high permeability and mostly connected to free-water mobility, the retar-

dation/sorption process is relevant to certain soil fractions and compositions. Ultimately, the PFAS sorption and transport in aqueous-solid, solid-air, and aqueous-air interfaces are complex and not well-studied processes. The PFAS sorption/interaction can be affected by the mineral composition of the heterogeneous media as well as porosity and pore-size distribution [22,28,30,41]. Further investigation is required to better understand the PFAS distribution in soil and groundwater. Consequent PFAS transport was primarily subjected to groundwater extraction, processing, and distribution with drinking water. Since water treatment, prior to plant modification, was restricted to treatment barriers insensitive to PFAS, the drinking water is assumed to have been contaminated for an extended period.

In the present study, several corresponding transport estimates were introduced, including PFAS sorption and transport in soil and far-field transport with groundwater. However, an accurate estimate was not possible due to data limitations on PFAS sorption parameters, emission rates and durations, and inconsistency in observed PFAS concentration and composition. Further investigations are necessary for the validation of the transport estimates.

In cases of historical contamination, as considered in the present study, the observed contamination levels can be represented by a series of inconsistent AFFF release events. Thus, representing a series of short-lasting contaminant spikes of different magnitude (levels) and related PFAS composition. The far-field transport processes, however, are strongly connected to the hydrogeological features of the area and related response time variability. It is therefore suggested important to consider the scale-related features of both PFAS emission and distribution processes.

3.4. Remarks on Regulations

There are currently several regulations on PFAS; these are, however, implemented on different institutional/industrial levels. The initial attempts on regulating the substances came from the manufacturers, possibly, as a response to the rising awareness of the environmental and health impacts of PFAS (e.g., a gradual phase-out of PFOS and PFOA) [44–46]. The UN Stockholm Convention on Persistent Organic Pollutants and EU registration, evaluation and authorization of chemicals (REACH) was established in the 2000s and remains as a primary regulatory tool (for listed individual substances) [47]. In the latest amendment by REACH (EU 2021/1297) the regulations were expanded to include long-chain (C9–C14) PFCAs, their salts and related substances [48]. To date, there are, however, no regulations on PFAS as a group/class of chemicals [47].

In the context of PFAS-AFFF, the European Chemicals Agency (ECHA) is aiming for an EU-wide restriction on PFAS in firefighting foam [49]. However, identification and disposal of the PFAS-AFFF, as well as a safe transition and replacement, are yet to be addressed [50]. On the other hand, as in relation to the public health and risks of human exposure to PFAS, a revision of the acceptable level in water can be a strong factor, e.g., the EU drinking water directive (EU 2020/2184) limiting the drinking water levels to 100 ng L⁻¹ (PFAS 10) and 500 ng L⁻¹ (PFAS total) [51]. In Sweden, an additional limit has been set to 4 ng L⁻¹ for PFOS, PFOA, PFHxS, and PFNA [51,52]. Ultimately, work regulating PFAS is an ongoing process that evolves with an increase in knowledge, as well as in public awareness. Both regulatory actions and an actual practical implementation are challenging and demanding tasks that require international efforts.

4. Conclusions

PFAS emission at the studied site was connected to PFAS-AFFF application during fire-training and fire-fighting equipment tests. The groundwater contamination was studied and PFAS contamination levels were established. Measured Σ PFAS concentrations in groundwater ranged between 20 and 20,000 ng L⁻¹. The PFAS composition in analyzed samples was dominated by PFOS and PFHxS. The PFAS chain length is suggested to have an impact on the distribution and transport of groundwater. Based on PFAS profiling, PFAS contamination of groundwater was primarily connected to the application of PFSA-

based and PFCA-/FTSA-based PFAS-AFFF. AFFF utilization and PFAS release scenarios were studied, and PFAS emissions per individual fire-training session were estimated as $[1.4 < 11.5 \pm 5.7 < 43.7 \text{ kg}]$ ($n = 20,000$). The PFAS annual emissions were estimated as $[11 < 401 \pm 233 < 1125 \text{ kg yr}^{-1}]$ ($n = 1005$, corresponding to $[2 < 35 \pm 20 < 96]$ individual fire-training sessions per year). Further investigation is necessary regarding the PFAS-AFFF formulations and application routine. Distribution of PFAS in the aquatic environment is associated with transport in surface runoff, vertical advective-diffusive transport in unsaturated soil, advective transport with groundwater, and further distribution with raw water extraction. Further investigation is necessary for the assessment of the PFAS transport conditions.

Supplementary Materials: The following supporting information (in supplementary materials) can be downloaded at: <https://www.mdpi.com/article/10.3390/w15010137/s1>, Figure S1: study area description, including: airfield territory, PFAS emission sources, surface water bodies and groundwater extraction locations; Figure S2: topography of the studied area, including: airfield territory, surface water bodies, PFAS emission sources and spread area; Figure S3: description of the soil layer depth at the studied area, including: airfield territory, PFAS emission sources and spread area, surface water bodies and groundwater extraction locations; Figure S4: hydrogeology of the studied area, including: airfield territory, PFAS emission sources and spread area, surface water bodies and groundwater extraction locations, groundwater confinement and capacities; Figure S5: bedrock lithology at the studied area, including: airfield territory, PFAS emission sources and surface water bodies; Figure S6: photo of the POCIS prior to deployment into the groundwater well; Table S1: PFAS concentrations measured in triplicate groundwater samples collected from groundwater extraction wells corresponding to GW1, GW3 and GW4; Table S2: PFAS detected in POCIS and corresponding PFAS concentrations estimated for groundwater measured at GW3 and GW1; Table S3: example of analyzed PFAS compositions corresponding to PFAS-AFFF reported for US market and measured PFAS concentrations and relative composition in PFAS-AFFF reported for on Swedish market.

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