Paper product production identified as the main source of per- and polyfluoroalkyl substances (PFAS) in a Norwegian lake: Source and historic emission tracking☆

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ABSTRACT

The entirety of the sediment bed in lake Tyrifjorden, Norway, is contaminated by per- and polyfluoroalkyl substances (PFAS). A factory producing paper products and a fire station were investigated as possible sources. Fire station emissions were dominated by the eight carbon perfluoroalkyl sulfonic acid (PFSA), perfluorooctanesulfonic acid (PFOS), from aqueous film forming foams. Factory emissions contained PFOS, PFOS precursors (preFOS and SAmPAP), long chained fluorotelomer sulfonates (FTS), and per-fluoroalkyl carboxylic acids (PFCA). Concentrations and profiles in sediments and biota indicated that emissions originating from the factory were the main source of pollution in the lake, while no clear indication of fire station emissions was found. Ratios of linear-to branched-PFOS increased with distance from the factory, indicating that isomer profiles can be used to trace a point source. A dated sediment core contained higher concentrations in older sediments and indicated that two different PFAS products have been used at the factory, referred to here as Scotchban and FTS mixture. Modelling, based on the sediment concentrations, indicated that 42−189 tons Scotchban, and 2.4−15.6 tons FTS mixture, were emitted. Production of paper products may be a major PFAS point source, that has generally been overlooked. It is hypothesized that paper fibres released from such facilities are important vectors for PFAS transport in the aquatic environment.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a class of chemicals used in cosmetics, household products, medical devices, oil production, pesticides, aqueous film forming foams (AFFF), textiles and paper (Lindstrom et al., 2011; Wang et al., 2017b). Due to adverse environmental and human health effects (Knutsen et al., 2018; Lau et al., 2007; Stahl et al., 2011; Sunderland et al., 2019),

PFAS have received attention from the scientific community and regulatory authorities (Directive 2013/39/EU, 2013; Norwegian Government, 2006; Prevedouros et al., 2006; Wang et al., 2017b). The highest PFAS concentrations have been reported for sites contaminated by point sources such as AFFF from firefighting training (Anderson et al., 2016; Filipovic et al., 2015; Hu et al., 2016; Moody and Field, 2000). However, PFAS are ubiquitous in the environment and are even found at remote pristine locations (Ellis et al., 2004; Gao et al., 2019; Houde et al., 2011; Lescord et al., 2015; Liu and Mejia Avendaño, 2013; Tomy et al., 2004). Different data and techniques have been used to characterize sources, emissions, and the spread of PFAS pollution (Dorrance et al., 2017), including fate and transport properties, chemical profiles (where PFAS

composition in different samples is compared), spatial distribution, and PFAS history (Dorrance et al., 2017; Hu et al., 2018; Shi et al., 2015; Trier et al., 2011; Zhang et al., 2016).

As will be presented, PFAS mixtures in the environment can potentially be back-tracked to production methods and possibly the application of specific products. Two methods have been used for large scale PFAS production: electrochemical fluorination (ECF) and telomerization. ECF generates a mixture of linear and branched isomers in addition to impurities of other fluorinated compounds (Prevedouros et al., 2006), while telomerization primarily produces linear isomers (Buck et al., 2011). ECF has been used to produce PFOS and perfluorooctanesulfonyl fluoride (POSF) based products (Benskin et al., 2010; Paul et al., 2009; Prevedouros et al., 2006). PFOS and POSF based chemicals were phased-out in some parts of the world in the 2000s (Butenhoff et al., 2006), and PFOS was listed as a persistent organic pollutant (POP) by the United Nations Environment Programme (UNEP) Stockholm Convention in 2009 (UNEP - The Stockholm Convention, 2019). As an example, the use of PFOS in AFFF was phased out between 2006 and 2011 in Norway (Norwegian Government, 2006). Fluorotelomer based PFAS, produced by telomerization, are of the substances that have been used as replacements (Field and Seow, 2017; Hoke et al., 2015; Moe et al., 2012; Place and Field, 2012; The Swedish Chemicals Agency (KEMI), 2015; Wang et al., 2015).

Several commercial PFAS mixtures produced by ECF have been identified that contain compounds that can (bio)transform to PFOS in the environment, including the N-alkyl substituted perfluorooctane sulfonamides ($F_{17}C_8-\overset{\overset{O}{\|}}{\underset{\underset{R}{|}}{S}}-N-R$), for simplicity termed preFOS throughout this study, and their parent compounds such as the mono-, di-, and tri-substituted phosphate esters of N-ethyl perfluorooctane sulfonamido ethanol (referred to collectively as SAmPAP) (Armitage et al., 2009; Benskin et al., 2012a, 2012b; Lee and Mabury, 2011; Martin et al., 2010; Olsen et al., 2005; Paul et al., 2009). Due to the preferential biotransformation of branched precursor isomers, producing branched PFOS (Br-PFOS), observation of elevated ratios of Br-PFOS to linear PFOS (L-PFOS) has been suggested to indicate a major contribution from PFOS precursor compounds (Benskin et al., 2009b; Chen et al., 2015a; Peng et al., 2014; Ross et al., 2012). The ratio is therefore suggested to be a useful source tracking tool for precursor based sources (Benskin et al., 2009b; Gebbink et al., 2016). Br-PFOS has been reported to be more water soluble and have a lower depuration half-life in organisms compared to L-PFOS (Benskin et al., 2009a; Chen et al., 2015a; Zhang et al., 2013). This can increase the complexity of PFAS source tracking by leading to variations in branched to linear ratios (Martin et al., 2010).

Depending on the PFAS application and the industrial era, different mixtures of PFAS, with different chemical profiles, have been used (Trier et al., 2011; Wang et al., 2013; Zhang et al., 2016). These profiles can be used to track what mixtures were emitted, when one mixture was substituted for another, and provide information about current and historic sources (Land et al., 2018; Wang et al., 2017b; Xiao, 2017). For example, 6:2 fluorotelomer sulfonate (6:2 FTS), and other fluorinated telomer products with 6:2 configurations have been used as replacements for PFOS in AFFF (Hoke et al., 2015; Moe et al., 2012; Place and Field, 2012; The Swedish Chemicals Agency (KEMI), 2015; Wang et al., 2015). 6:2 FTS is a precursor of perfluoroalkyl carboxylic acids (PFCA) (Wang et al., 2011a), and the same is suspected to be the case for longer FTS (Simonnet-Laprade et al., 2019). Emission history, such as the shift from PFOS to 6:2 FTS in AFFF after the PFOS phase-out has previously been shown to be reflected in sediment cores (Lutz et al., 2009; Mussabek et al., 2019). Only the top 11 cm of sediments

from Tokyo Bay, Japan, was found to contain 6:2 FTS which corresponds to sediments settling from 2002 (Lutz et al., 2009).

Langberg et al. (2020) have previously reported that the entirety of the sediment bed in lake Tyrifjorden, Norway, is polluted by hydrophobic precursors (preFOS, SAmPAP, FTS) of perfluorinated alkyl acids (PFAA), resulting in substantial sediments and biota concentrations in all areas of this 138 km^2 lake, whilst concentrations in water are generally near or below detection limits (sum of L-PFOS and Br-PFOS of 0.22–0.28 ng L^{-1} in lake water) (Langberg et al., 2020). The objective of the present study was therefore to identify and better characterize the source of this pollution, as well as estimate the historic input of PFAS to the lake system. The two major likely point sources were a fire station where AFFF was used, and a factory producing PFAS coated disposable paper products. The present study builds on the work presented in Langberg et al. (2020), however as the objective differs from the previous study, data were interpreted using a different approach. In addition, water and sediments from the storm water system at the fire station, water and sediments sampled downstream a landfill filled with waste from the factory, a product from the factory (paper plate), sediment and fish samples from the river directly downstream to the fire station, and a dated sediment core from the lake were included in the present work. The present study uses source tracking methods (spatial distribution, PFOS isomer patterns and sediment core dating) to decipher which point source was responsible for the pollution. PFAS concentrations and profiles from the two source areas were compared to concentrations and profiles in river and lake sediments and water. Following the identification of the main source, a fate and transport model was employed to back-calculate historic emission volumes, to predict future sediment concentrations, and to draw hypotheses related to possible mechanisms that can explain the spreading of PFAS in the lake. This work is the first to use source tracking methods to positively identify the paper production industry as a major PFAS hot spot source and to estimate emission volumes and transport mechanisms from such industrial activity based on an environmental record.

2. Materials and methods

2.1. Case study site and sampling

Lake Tyrifjorden (60.03° N, 10.17° E), Norway is a freshwater lake with a surface area of 138 km^2 and a maximum depth of 288 m. Further details are given in the section *Site description - Lake Tyrifjorden* in the supplemental information (SI). In 2015, elevated PFOS concentrations (mean 183 ± 25 μg kg^{-1}, n = 5) were reported in perch livers (*Perca fluviatilis*) from the lake (sampled close to area L3 in the present study, see the description below) (Fjeld et al., 2016). Follow-up investigations identified two suspected major PFAS sources to the lake: a fire station that opened in the 1980s and used AFFF until 2007, and a shutdown factory that produced paper products from 1964 to 2013 (Slinde and Høisæter, 2017). The fire station and factory are located on the banks of a river flowing into the lake, with the fire station located 11 km upstream from the river mouth, and the factory a further 15 km upstream (Fig. S1 in the SI which shows all sampling locations). To simplify the presentation of data, the entire lake was divided into 6 regions: L1 to L6. L1 is the region closest to the river mouth and L4 to L6 the furthest away. L5 is connected to the outflow of the lake (Fig. S1). The sampling area in the river downstream the factory was termed the factory area. Sampling procedures are described briefly below and more information and details of quality assurance procedures, including sample storage and limit of quantifications (LOQ), are provided in the sections *Sampling and sample preparation* and *Quality assurance*

and sample storage in the SI.

2.2. Abiotic samples

Access to the factory itself was not possible so water and sediments were sampled in November 2018 from a creek located downstream a landfill used by the factory during the late 1980s to the 1990s (Fig. S1). The landfill is now closed. These samples were used to represent the PFAS emission profile of the factory. In addition, a paper plate produced at the factory in 2007 was analysed to determine the PFAS mixture used at that time. At the fire station, water was sampled (n = 2) from intermediate bulk containers and sediments were sampled (n = 2) from containers during cleaning of the storm water system (more information is given in the SI section *Sampling and sample preparation*). Downstream from the landfill, water (n = 1) and sediments (n = 1) were sampled from the creek. Water was sampled by submerging sample-rinsed high-density polyethylene (HDPE) bottles (1 L) directly in the water source. Sediments were sampled using a metal tube attached to a telescopic pole.

River and lake sediments were sampled from two locations upstream from the factory, nine locations in the river downstream the factory, four locations downstream the fire station, and 94 locations in the lake (shown in Fig. S3 and Table S1). Sediments were sampled with either a van Veen sampler or a Kajak-Brinkhurst sediment corer where the top two cm was carefully sampled if visually undisturbed. Sediment samples were transferred into pre-baked glass jars with HDPE lids. One core from sampling area L1 was divided in one cm intervals for determination of the vertical PFAS distribution profile and dating of sediments. Sediment traps (plexiglass, 10 cm internal diameter) were used to investigate PFAS concentrations in present day settling sediments (details in the *section Sampling and sample preparation* in the SI). Sediment in the river close to the factory were sampled in August 2018. Lake and river (downstream the fire station) sediment and pore water were sampled in September 2018. Abiotic samples were kept in insulated boxes and brought to the laboratory within 24 h of sampling. The samples were kept in the refrigerator (dark, at 4 °C) until analyses.

2.3. Biota

Fish (perch [*Perca fluviatilis*] and pike [*Esox lucius*]) were collected in the period June−October 2018 using fish nets (35−39 mm mesh size). Sampled biota varied between areas (n = 2−5), shown in Table S2. Whole organisms were carefully wrapped in three layers of clean aluminium foil and put in a clean plastic bag (polyethylene), before being frozen at −20 °C. Frozen biota samples were sent to the laboratory (in sealed, insulated boxes) for sample treatment and analysis.

2.4. Laboratory methods

The analytical methods to quantify extractable organic fluorine (EOF), pore water concentrations, and total organic carbon (TOC) are described fully in the SI section *Laboratory methods*. The sediment core from sampling area L1 was dated using unsupported ^{210}Pb, analysed via gamma spectrometry (details in the section *Laboratory methods* in the SI). Water was extracted using solid-phase extraction (SPE). Biota (fish livers) and sediment were extracted using acetonitrile and ultrasonication. Liquid chromatography quadrupole time-of-flight mass spectrometry (LC-qTOF-MS) was used for PFAS analyses (see all PFAS and acronyms in Tables S3 and S4). Authentic standards (i.e. a standard identical to the targeted substance) and internal standards were used to quantify 44 PFAS, while exact mass and retention time from

authentic standards were used to screen for 19 PFAS. In addition to this, peaks for Br-PFOS were identified (confirmed) using a standard mixture of Br-PFOS isomers. As the standards for the Br-PFOS isomers were in the form of a mixture, they could not be used for quantification purposes. Therefore, the standard for L-PFOS was used to quantify the peaks in the chromatogram which were made up of the different Br-PFOS isomers, and the sum of all the Br-PFOS isomers was reported. By using exact mass and estimated retention time, an additional 28 PFAS were screened for. Peaks in the chromatograms were observed at expected retention times for three substances, and these were quantified using the standard for a structurally similar compound (details in the section *Laboratory methods* in the SI). According to the literature, the detected compounds indicated the use of an EtFOSE based PFAS product, and therefore likely that SAmPAPs were one of the parent compound groups (Martin et al., 2010; Trier et al., 2017). The analytical range for most samples (m/z: 150−1100) did not include SAmPAP diester (m/z: 1203) and SAmPAP diester could therefore not be looked for in most samples. However, SAmPAP diester was analysed in the sediment core and the sediment sample used for EOF, described in the SI section *Laboratory methods* (PFAS names, acronyms and more details are given in Table S3).

2.5. Quality assurance

Lab blanks were run following the same procedures as for field samples in each analysis batch. As the whole lake is polluted by PFAS (see Fig. S3), the use of a reference site in the lake system was not possible. Concentrations in the blank samples were low (<0.5 ng g^{-1} or ng L^{-1}) and consistent, indicating little cross contamination. Blank concentrations were subtracted from results when calculating sample concentrations. Recoveries in the present work were satisfactory (within the range of 70−110%, see Table S3). A random sample was selected from each matrix for duplicate analysis to control for repeatability.

2.6. Data handling and statistics

Arithmetic means and the standard error of the mean (SEM) are reported. Differences in PFAS concentrations and profiles for fish livers, sediments, and pore water at the different areas were tested using Kruskal-Wallis and Bonferroni correction. The significance level was set to 0.05. PFAS profiles in sediments and the paper product were compared using principal component analysis (PCA). Details related to the statistical analyses are in the section *Statistics and data analysis* in the SI.

2.7. Modelling

A fate and transport model of PFAS entering the lake was employed based on the previously reported Drammensfjord model (Arp et al., 2014). In principle, this model could be used for any lake/fjord or contaminant, provided the necessary input data is available (Arp et al., 2014; Oen and Arp, 2014). The model is a two-box water-sediment model that allows for changes in emissions of a pollutant within specified time-intervals following a first-order rate constant. The water domain describes all transport and transformation processes in the water phase over the entire lake, the sediment domain describes all transport and transformation processes in the sediment phase, including deep sediment burial (Fig. S6). These domains are modelled following coupled linear differential equations (Schwarzenbach et al., 2003) to account for the interdependency of sediment and water processes. Details are provided in the SI section *Modeling*.

3. Results and discussion

3.1. Lines of evidence for source tracking

3.1.1. Differences in PFAS profiles in samples

Concentrations and profiles of targeted PFAS in the water and sediment samples from the two suspected sources (fire station and paper production) varied (Fig. S7, concentrations in Tables S9–S10).

Water from the storm water system at the fire station was dominated by C5–C8 PFCA and perfluoroalkyl sulfonic acids (PFSA), while sediments were dominated by PFOS in addition to perfluorohexane sulfonic acid (PFHxS), FOSA, and 6:2 FTS. Relatively minor levels of C9 and C10 PFSA and PFCA, other preFOS compounds (FOSAA, EtFOSAA) and 8:2 FTS were detected, likely reflecting impurities, or that small amounts of different AFFF products have been used. PFAS profiles in fire station storm water and sediments are consistent with profiles previously reported for AFFF impacted areas (Backe et al., 2013; Dauchy et al., 2017; Filipovic et al., 2015; Langberg et al., 2019; Prevedouros et al., 2006).

Water from the creek downstream the factory landfill was dominated by PFOA, PFOS and EtFOSAA as well as a smaller proportion of C5–C7 and C9 PFCA, FOSA, and FOSAA. Sediment samples from the creek were dominated by 8:2 FTS and 10:2 FTS, smaller fractions of EtFOSAA and 12:2 FTS, in addition to some 14:2 FTS, FOSAA, EtFOSE, and PFOS. The paper plate was dominated by C6–C10 PFCA with smaller proportions of C12–C14 PFCA, 8:2 FTS, and 10:2 FTS. PFAS profiles in water and sediments in the creek downstream the landfill show the compounds, or their degradation products, that were used in paper production since the 1970s (i.e. SAmPAP and preFOS) (Olsen et al., 2005; Trier et al., 2011, 2017). The creek drains into lake Tyrifjorden and as such is a source of PFAS to the lake. However, as the landfill was filled with waste from the factory it is considered to represent factory emissions. Further, the total amount of PFAS in lake sediments (tons, according to an extrapolation of concentrations in the sediment core discussed

below and shown in Table 1) make it difficult to decipher a realistic estimate of the contributions from emissions via the creek to lake sediment concentrations. The national sum 28 PFAS emissions from Norwegian landfills have been estimated to be 0.017 tons per year (average per landfill was reported to be 0.00016 tons per year) (Knutsen et al., 2019). Similarly, other estimates of yearly PFAS emissions via landfill leachate (per landfill) are in ranges far below the volumes needed to account for the masses observed in lake Tyrifjorden sediments (Benskin et al., 2012b; Lang et al., 2017; Masoner et al., 2020). The profile in the paper plate from 2007, which did not contain PFOS above the LOQ, reflects that it is manufactured after the phase out of PFOS and related compounds (Butenhoff et al., 2006). The high percentages of PFCA and FTS might indicate that these substances were used as replacements at the time. The concentrations of PFCA in the paper plate were in the range of 6–7156 µg kg^{-1} (see Table S10), which is comparable to concentrations previously reported by (Xu et al., 2013). It is uncertain if the extraction method used (see description in the *Laboratory methods* section in the SI) extracted all the relevant PFAS (Schaider et al., 2017; Trier et al., 2011), however it is clear that large volumes of PFCA and FTS were used at the time. As a variety of different PFAS products have been used for paper products (Schaider et al., 2017; Trier et al., 2011), the analysed paper product does only represent a snapshot of the production at the factory. Nevertheless, the differences in PFAS profiles from the fire station and paper producing factory provide important source tracking information.

As previously reported, concentrations of targeted PFAS in river and lake water were low (i.e. the sum of L-PFOS and Br-PFOS in lake water was 0.22 and 0.28 ng L^{-1}) (Langberg et al., 2020). Thus, PFAS concentrations in lake water indicate limited ongoing emissions of PFAS to the lake.

Concentrations of targeted PFAS in sediments (dry weight; d.w.) sampled upstream the factory area were low and the only

Table 1
Model output for the two suspected PFAS products using three different log K$_{OC}$ values.

			Scotchban[a]			FTS Mixture[b]		
			log K$_{OC}$			log K$_{OC}$		
			5	**7**	**9**	**5**	**7**	**9**
Total emission estimates to lake Tyrifjorden								
Emissions entering the lake		(tons)	189.0	42.2	41.6	15.6	2.5	2.4
Emissions leaving the lake		(tons)	154.0	0.8	0.4	13.7	0.07	0.04
Estimated mass in sediments		(tons)	34.5	41.3	41.2	1.9	2.4	2.4
Mass in sediments extrapolated from the sediment core		*(tons)*	*40.7*			*2.3*		
Predictions								
C$_{Lake, sed}$[c]	2017	(ng g^{-1})	11	41	31	104	102	100
	Measured 2018	*(ng g^{-1})*	*25*			*68*		
	2030	(ng g^{-1})	6	24	18	60	58	57
		(% reduction)	44	42	42	43	43	43
	2060	(ng g^{-1})	6	21	16	57	56	55
		(% reduction)	48	48	48	45	45	45
C$_{Lake,w (total)}$[d]	2017	(ng L^{-1})	4	0.3	0.1	36	0.8	0.5
	Measured 2018	*(ng L^{-1})*	*0.2*			*<LOQ*		
	2030	(ng L^{-1})	2 × 10^{-3}	2 × 10^{-4}	8 × 10^{-5}	2 × 10^{-2}	4 × 10^{-4}	3 × 10^{-4}
		(% reduction)	~100	~100	~100	~100	~100	~100
	2060	(ng L^{-1})	2 × 10^{-3}	2 × 10^{-4}	7 × 10^{-5}	2 × 10^{-2}	4 × 10^{-4}	2 × 10^{-4}
		(% reduction)	~100	~100	~100	~100	~100	~100

[a] Scotchban is considered sum of all SAmPAP diester, preFOS, PFSA, as well as PFCA (prior to 1990).
[b] FTS mixture is considered the sum of all FTS as well as PFCA (after 1990).
[c] Total sediment concentration i.e. sediment plus freely dissolved porewater (ng g^{-1}), and projected percentage reduction in top sediments in the future (2030 and 2060) compared to the concentration in 2017.
[d] Total water concentration i.e. freely dissolved phase plus particle/colloid bound (ng L^{-1}), and projected percentage reduction in water in the future (2030 and 2060) compared to the concentration in 2017.

substance above the LOQ was PFHxS (max 2.17 µg kg^{-1}). In contrast to this, elevated concentrations were found in sediments from the factory area and in the lake, as shown in Fig. 1A (all PFAS concentrations are listed in Table S11, sediment particle size distribution and TOC are shown in Table S12, and spatial distributions are shown in Figures S3 and S8-S11). The TOC content in river and lake sediments were between 0.3 and 4.5%. A thorough discussion of the effect of sediment characteristics on PFAS concentrations in sediments is provided in Langberg et al. (2020). The mean ΣPFAS 29 in river sediments from the factory area was 2450 µg kg^{-1}, and in lake sediments means ranged between 6.1 and 207 µg kg^{-1} (L6 and L2, respectively). As these areas collectively cover all main parts of the lake, it is clear that PFAS has been spread over the entire lake bed. Maximum concentrations of the dominating PFAS were 688−2150 µg kg^{-1} for C10−C16 FTS, 2455 µg kg^{-1} for EtFOSE, 1831 µg kg^{-1} for EtFOSAA, 1780 µg kg^{-1} for L-PFOS, 677 µg kg^{-1} for Br-PFOS, and 184−665 µg kg^{-1} for C10−C12 PFCA. PFAS profiles in sediments from the lake were dominated by the same compounds as from the factory area (especially FTS and preFOS), as shown in Fig. 1B. The ΣPFAS 29 concentration generally decreased with increasing distance from the factory area (significantly lower in area L5 [p = 0.02] and L6 [p < 0.01]) compared to the factory area. Concentrations in the sediments sampled in the river downstream the fire station were below the LOQ.

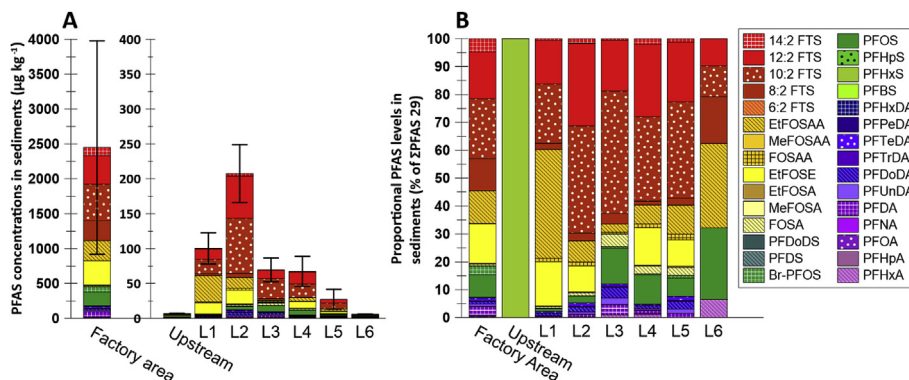Concentrations of ΣFTS (6:2, 8:2, 10:2, 12:2, and 14:2 FTS) and ΣpreFOS (EtFOSAA, MeFOSAA, FOSAA, ETFOSE, ETFOSA, MeFOSA, FOSA) in sediments followed the same trend as for ΣPFAS 29 where they generally decreased amongst the different lake regions with increasing distance from the factory area, however significantly lower ΣFTS concentrations compared to the factory area were only detected for L5 and L6 (ΣFTS; p = 0.03 and p < 0.01, respectively) and for L6 for ΣpreFOS (ΣpreFOS; p < 0.01). PFAS profiles did not show the same clear pattern, however the percentage of FTS as compared to total PFAS was significantly lower (p ≤ 0.01) at sampling area L6 compared to the factory area and all other sampling areas in the lake, and the percentage of preFOS at area L1 was significantly higher compared to areas L3 (p = 0.02), L4 (p = 0.02), and L6 (p < 0.01). Concentrations and profiles in the sediment traps in the river downstream both suspected sources and in the lake showed elevated concentrations of preFOS and FTS (Table S14). As previously reported (Langberg et al., 2020), concentrations in pore water were relatively high (the highest mean concentration of L-

PFOS was 392.2 ng L^{-1} at area L4), and reflect the higher solubility of PFAA compared to their larger precursors, shown in Table S15.

The low concentrations in sediments upstream the factory indicate that there are no significant PFAS sources further upstream that contributed to the observed PFAS loads in the lake. High concentrations at the factory area and decreasing concentrations with increasing distance into the lake clearly indicate a significant contribution of the PFAS pollution from the factory. Based on this, the concentrations in the sediments outside the fire station that were below the LOQ are unexpected (as the fire station is downstream the factory). One possible explanation could be that the high river current in the area prevented PFAS polluted particles from settling and transported them further into the lake. All PFAS profiles from the sediment samples and the paper plate were compared using PCA (Fig. S13). Samples from the fire station storm water system were grouped separately based on their content of 6:2 FTS and PFSA, while sediments from the factory area, landfill (with waste from the factory), and the lake grouped together. The paper plate did not group with either one, however its PC 1 score (x-axis) was similar to the PC 1 scores for sediments from the factory area, landfill, and the lake. PFAS profiles in the sediments downstream the factory and the landfill are similar to profiles in the lake sediments. The same compounds, that is preFOS (and related compounds) and precursors to FTS (based on their structure, such as FTMAP, also termed S-diPAP), have been reported to be used in paper production (Trier et al., 2011, 2017). PFAS profiles in the sediment traps in the river downstream both suspected sources and in the lake, reflect the dominate compounds in lake sediments, indicating that present day settling sediments are contaminated by the same source as sediments in the lake bed. Therefore, it is concluded that emissions originating from the factory are the main source of PFAS contamination in lake sediments.

3.1.2. PFAS in biota

Concentrations of targeted PFAS for perch livers (wet weight; w.w.) are presented in Fig. 2. Data for the different stations for both perch and pike is shown in the SI (Tables S16−17). 21 PFAS (+Br-PFOS) were detected in perch liver. The same number of PFAS was detected in pike livers, however EtFOSA was detected in pike but not perch, while PFHpA was detected in perch but not pike. The concentrations for PFOS in perch livers at sampling area L3 (188 ± 85 µg kg^{-1}) were comparable to the concentrations reported

Fig. 1. Average PFAS concentrations (d.w.) (A) and distribution profiles (B) in sediments at the different regional stations (i.e. sampling areas) in the river and lake (n = 2−25, shown in Table S1). FTS (6:2, 8:2, 10:2, 12:2, and 14:2 FTS) are coloured red, preFOS (EtFOSAA, MeFOSAA, FOSAA, ETFOSE, ETFOSA, MeFOSA, FOSA) are yellow, PFSA are green, while PFCA are blue and purple. For concentrations, the scale on the y-axis are different for the factory area and the other sampling areas. Distribution profiles are given as relative concentrations (of ΣPFAS 29). Only compounds detected above the LOQ in at least one sample are included in the data analysis. In samples where compounds were not present above the LOQ, concentrations are taken as half the LOQ for plot A. For the distribution profiles in B, concentrations below the LOQ are treated as 0. Statistically significant differences between areas are shown in Fig. S12. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)
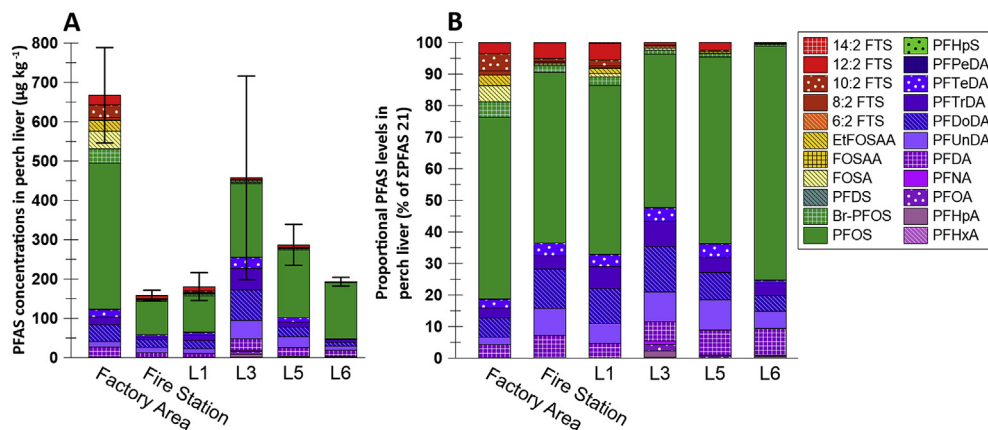
Fig. 2. Average PFAS concentrations (w.w.) (A) and distribution profiles (B) in perch livers at the different sampling areas (n = 2−5, shown in Table S2). FTS (6:2, 8:2, 10:2, 12:2, and 14:2 FTS) are coloured red, preFOS (EtFOSAA, FOSAA, FOSA) are yellow, PFSA are green, while PFCA are blue and purple. Distribution profiles are given as relative concentrations (of ΣPFAS 21). Only compounds detected above the LOQ in at least one sample are included. For concentrations (A), values below the LOQ are treated as half the LOQ. For distribution profiles (B), values below the LOQ are treated as 0. Statistically significant differences between areas are shown in Fig. S14. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

in the study in 2015 (183 ± 25 µg kg^{-1}) (Fjeld et al., 2016). The highest PFAS concentrations were generally found in fish from the factory area, similar to the sediment results. Maximum concentrations of the dominating PFAS in the perch livers from the factory area were 25−96 µg kg^{-1} for C10−C14 PFCA, 640 µg kg^{-1} for L-PFOS, 88 µg kg^{-1} for Br-PFOS, 195 µg kg^{-1} for FOSA, 64 µg kg^{-1} for EtFOSAA, and 14−56 µg kg^{-1} for C10−C14 FTS. Mean ΣPFAS 21 in perch liver was 667 µg kg^{-1} at the factory area, 158 µg kg^{-1} at the fire station, 181 µg kg^{-1} at L1, 458 µg kg^{-1} at L3, 287 µg kg^{-1} at L5, and 193 µg kg^{-1} at L6. Perch liver ΣPFAS 21 concentrations were significantly lower at the fire station (p = 0.03), L1 (p = 0.02), and L6 (p = 0.03), compared to the factory area, shown in Fig. 2A. Similar trends were observed for pike livers (Table S17).

Concentrations of ΣFTS and ΣpreFOS as well as their relative percentages compared to ΣPFAS 21 generally decreased with distance from the factory area (Fig. 2). ΣFTS concentrations as well as relative percentages were significantly lower (p < 0.01) in perch livers from sampling areas L3, L5, and L6 compared to perch from the factory area. ΣpreFOS concentrations were significantly lower in the fire station (p = 0.03), L3 (p = 0.01), L5 (p < 0.01), and L6 (p < 0.01) areas, compared to the factory area (Fig. 2A). Relative percentages of ΣpreFOS compared to ΣPFAS 21 were significantly lower in perch livers from areas L3 (p = 0.01), L5 (p < 0.01), and L6 (p < 0.01) compared to perch livers from the factory area.

The dominant PFAS in lake perch are consistent with those in factory area perch, in factory area and landfill sediments and water, and in lake sediments. This collectively indicates that the factory is a major source of the observed PFAS in biota. Dominance of PFOS, in addition to the presence of other PFSA and comparatively low concentrations of PFCA have previously been reported for perch sampled at AFFF impacted sites (Ahrens et al., 2015; Kwadijk et al., 2014). There is little evidence for bioaccumulation of C4−C7 PFSA, which if present could be attributed to AFFF used at the fire station. The C7 PFSA (PFHpS), was the only of the C4−C7 PFSA above the LOQ in biota. PFHpS was detected in all perch livers from the factory area (n = 5), in none of the perch from the fire station (n = 2), and in seven perch livers from the lake (n = 20). The C6 and C7 PFSA, PFHxS and PFHpS, are previously reported to bioaccumulate in fish, however bioaccumulation potentials are smaller, and half-lives are shorter, compared to PFOS (Falk et al., 2015; Labadie and Chevreuil, 2011; Lescord et al., 2015; Zhong et al., 2019). The lower bio-accumulation potentials complicate the use of these compounds as

indicators of AFFF contamination, however PFHxS has previously been reported in whole fish, fish liver and fish muscle at AFFF polluted sites (Filipovic et al., 2015; Kärrman et al., 2011; Langberg et al., 2019; Lanza et al., 2017). The lack of PFHxS in biota from lake Tyrifjorden, indicates that emissions from the fire station do not result in detectable PFHxS accumulation. The results taken together show that the biota profiles also reflect PFAS emissions from the factory and not the fire station.

The decreasing fish liver concentrations of FTS and preFOS in regions further away from the factory echo the same trend in sediments. This is interpreted as an indication that the fish reflect PFAS concentrations in the abiotic environment in the part of the lake in which they were sampled. The relatively large distances between sampling areas (13−17 km between area L1 and areas L4, L5, and L6, see the section *Sampling and sample preparation* in the SI) and the relatively short (days) depuration half-lives of PFAA in fish (Martin et al., 2003) is likely the explanation for this observation even though fish are expected to move around in the lake. However, the same clear trend for preFOS and FTS was not observed for relative percentages, with a clear decreasing trend for percentages of FTS and preFOS in perch livers but not in sediments. The reason for this discrepancy is unclear but could be due to differences in factors such as partitioning coefficients and exposure pathways between the two media (i.e. perch livers and sediments). The decreasing proportions of FTS and preFOS in perch liver with distance from the factory area could be due to FTS and preFOS being less mobile in the environment compared to the PFAA, or it might indicate more complete transformation of these compounds with distance from the source (preFOS to PFOS and FTS to PFCA (Armitage et al., 2009; Paul et al., 2009; Simonnet-Laprade et al., 2019; Wang et al., 2011a)). A significant proportion of environmental PFAS not covered by targeted analyses, which are potential PFAA precursors are expected to be present (Barzen-Hanson et al., 2017; D'Agostino and Mabury, 2014). Corresponding to this, as reported previously (Langberg et al., 2020), extractable organic fluorine (EOF) in fish liver decreased with distance from the factory, and the sum of organic fluorine in the targeted PFAS as a percentage of EOF in fish livers generally increased with distance from the factory (Fig. S15 and Table S18). This might indicate lower proportions of unknown precursor compounds (and unknown intermediate transformation products) further from the factory due to a more complete transformation. Thus, biotransformation of PFAA

precursors might explain the high PFAA levels observed in areas furthest from the factory.

3.1.3. Branched and linear PFOS

PFOS products produced by the 3M Company using ECF have been reported to consist of approximately 70% linear and 30% branched isomers (Benskin et al., 2010; Jiang et al., 2015; Vyas et al., 2007). The percentage of L-PFOS (of ΣPFOS) were 68.2% and 64.3% in the lake water from L4 and L6, respectively (Langberg et al., 2020). Percentages of L-PFOS were 74.3−89.3% in pore water, 92.0−99.3% in perch liver, and 97.0−99.6% in pike liver and increased with distance from the factory area ($p < 0.05$) (Fig. S16, individual p values are shown in Table S19). The percentage of L-PFOS in water and fish at the factory area is comparable to previously reported percentages for both point and diffuse sources, including sources where precursor compounds could contribute to PFOS levels (urban runoff and sewage, water from a firefighting training area, and wastewater discharge) (Boulanger et al., 2005; Houde et al., 2008; Kärrman et al., 2011; Labadie and Chevreuil, 2011). The increasing percentages of L-PFOS with distance from the factory area observed in the present study are likely due to the higher water solubility and faster elimination in organisms of most Br-PFOS congeners compared to L-PFOS (Benskin et al., 2009a; Chen et al., 2015a, 2015b; Zhang et al., 2013). These processes result in environmental fractionation whereby L-PFOS is retained in biota and sediments, while Br-PFOS is removed with water exchange. Thus, over time and increasing distance from point sources, the amount of L-PFOS relative to Br-PFOS is expected to increase (for this type of environmental transport scenario). The faster transformation of branched isomers could also contribute to this, i.e. that more Br-PFOS precursors are transformed earlier/closer to the source compared to L-PFOS precursors (as all the ECF based preFOS have both branched and linear compositions) (Benskin et al., 2009b; Chen et al., 2015a,b; Peng et al., 2014; Ross et al., 2012). Thus, PFOS isomer profiles in perch livers, pike livers, and pore water represent further evidence that the factory is the main point source of PFOS to the lake.

3.1.4. Historic concentrations in lake sediments

The dated sediment core at L1 presents PFAS concentrations in sediments that settled between 1934 and 2017 (Fig. 3 and Table S20). High concentrations of SAmPAP diester were detected in the core (max: 3383 µg kg^{-1}), shown in Fig. 3A. This is in agreement with concentrations previously reported for top sediments in Lake Tyrifjorden (max: 1872 µg kg^{-1}), including the sample analysed for EOF (850 µg kg^{-1}) (Langberg et al., 2020). Core data was used to explore the introduction and phase-out of the different PFAS products. Peaks in the sediment core varied for different PFAS substances: PFOS peaked at approximately 1960; preFOS and SAmPAP diester peaked around 1984, with a smaller peak around 1960; PFCA and FTS peaked at the second half of the 1990s. The low concentrations in top (recent) sediments compared to deeper (older) sediments likely reflects lower levels in settling sediments after the factory was shut down in 2013. PreFOS and SAmPAP were detected in sediments dated to the 1950s. Production at the factory began in 1964 and preFOS based phosphate surfactants were commercialised in the late 1960s and introduced for use in food contact paper and packaging in 1974 (Olsen et al., 2005). Concentrations observed in the period between 1950 and 1970 could be due to uncertainties related to the dating, sampling, or bioturbation of PFAS in sediments. The concentration peaks of SAmPAP diester and preFOS in the 1980s correspond well to the history of the factory as well as reported use of PFAS in paper products. Therefore, the accuracy of the dating varies throughout the core, but appears more uncertain with depth.

The PFAS profile observed in sediments dated to pre-1995 corresponds to a 3M product called Scotchban which was used for paper products and contained a mixture of SAmPAP and preFOS (Martin et al., 2010; Trier et al., 2017). As commercial SAmPAP formulations were dominated by diester, with much less mono- and tri-ester (Lee and Mabury, 2011), this compound was prioritized for analysis. However, the presence of SAmPAP mono- and tri-ester in sediments were expected as well, as reported previously (Zhang et al., 2018). The decreasing concentrations of SAmPAP, preFOS, and PFOS from the late 1980s/early 1990s occurs before the phase out of PFOS and related compounds in 2002 (Martin et al., 2010). However, the peak and subsequent decline in PFOS concentrations is in agreement with previous studies (Furdui et al., 2008; Holmström et al., 2005; Kwadijk et al., 2010; Martin et al., 2004; Verreault et al., 2007). In the present study, high levels of FTS were detected downstream the landfill which was filled with waste from the factory during the late 1980s to the 1990s. Concentrations of 10:2 and 12:2 FTS dominate the sediment core between 2000 and 2010, peaking in 2006, indicating that the use of Scotchban was phased out at the site by the late 1990s. Thus, the reported decline of PFOS in the environment before the phase out might be due to a shift from SAmPAP and preFOS to other PFAS mixtures.
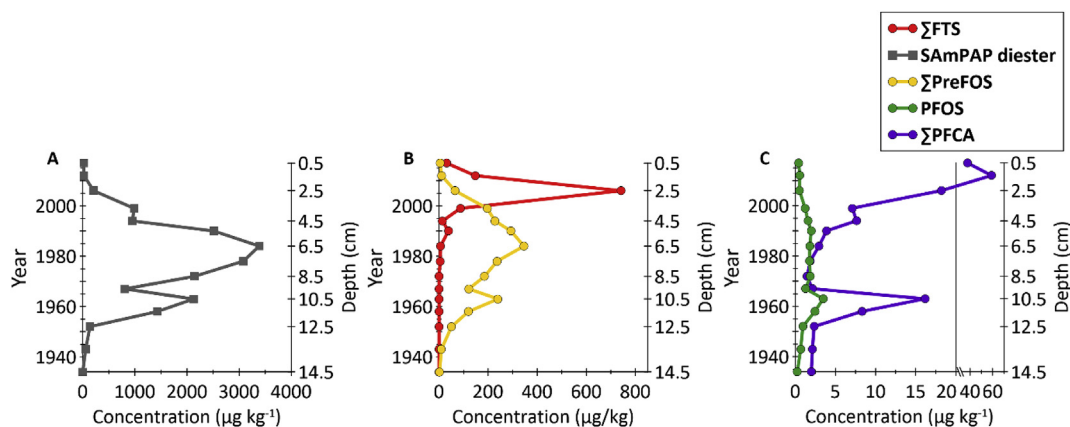
Fig. 3. PFAS concentrations in sediments from the dated core sample from area L1. Panel A: concentrations of SAmPAP diester. Panel B: Concentrations of ΣFTS and ΣpreFOS. Panel C: Concentrations of PFOS, and ΣPFCA (PFOS was the only PFSA above the LOQ). The black vertical line in C shows that the x-axis is split at the interval 20−35 µg kg^{-1}. Concentrations for individual compounds are shown in Fig. S18 and Table S20.

Fluorotelomer mercaptoalkyl phosphate esters (FTMAP, also termed S-diPAP and known by the tradename Lodyne P208E) have been used in food packaging since 1995, and are based on their structure likely precursors to FTS (Lee and Mabury, 2011; Trier et al., 2011). Other possible FTS precursors are groups containing the same (suspected) FTS precursor moiety as FTMAP such as 3-[2-(perfluoroalkyl)ethylthio] propionate (tradename: Zonyl FSA) (Trier et al., 2011), fluorotelomer sulfonamide alkylbetaines (FTAB) (Field and Seow, 2017; Moe et al., 2012), and others (Barzen-Hanson et al., 2017). However, it is not known to what extent these have been used in paper coatings, and research in this area is scarce. The main focus in the literature of FTS compounds are those with 6:2 and 8:2 structure (Field and Seow, 2017); detailed information regarding 10:2, 12:2, and 14:2 FTS and their potential precursors is not currently available. Nevertheless, the results indicate two eras of product emissions: 1) Scotchban (considered sum of all SAmPAP, preFOS, PFSA, as well as PFCA prior to 1990), and 2) the FTS dominated product(s), termed the FTS mixture (considered the sum of all FTS and their precursors as well as PFCA after 1990). Only the targeted compounds were included in calculations and for the modelling (described below), therefore SAmPAP mono- and triester and potential precursors to FTS were not included. Interestingly, another group of ester phosphates reported to be used in paper products, fluorotelomer alcohol (FTOH) mono- and di-substituted phosphates (Trier et al., 2011), were analysed for in water, sediments and biota, but not detected (neither were their expected degradation products, FTOHs), indicating that these compounds were not used at the factory (Table S3).

The sediment core profile corresponding to PFAS used in paper products gives further evidence to that the majority of the PFAS pollution in the lake originates from the factory. To the best of our knowledge, this (together with the results reported by Langberg et al., 2020) is the first-time that production of PFAS coated paper products has been reported to be a significant PFAS point source. Extrapolating the concentrations in the core to the entire lake except area L6 (due to lower concentrations in this area) gives a total mass residing in the sediment bed of 40 660 kg Scotchban and 2341 kg FTS mixture (*equation VII* in the SI). This extrapolation is based on the observation that surface sediments of the core match well with the mean and geometric average of all PFAS in all other surface sediments except area L6 (see Table S13), and this core is therefore assumed to be representative of the lake for the purpose of modelling (as described in the *Modelling* section in the SI). These calculations are based on the results from the targeted analyses, therefore, the unidentified fraction of organic fluorine, which is approximately 50% in the sediment sample which was analysed for EOF (Fig. S15), is not included.

3.2. PFAS fate and transport modelling

To understand how emissions from the factory may have resulted in PFAS pollution over the entire lake bed, a fate and transport model was employed. The purpose of this model was threefold: 1) to back-calculate emission volumes of the two suspected PFAS products, Scotchban and the FTS mixture, 2) to account for how much of the emissions of Scotchban and FTS mixture were likely dissolved or particulate bound, and 3) to extrapolate towards future predictions of emissions and sediment surface concentrations. Emissions in the model assume two eras of pollution that were calibrated to the sediment core data (using the method of least squares): the Scotchban era and the FTS mixture era. The Scotchban era was assumed to begin in 1950, followed by a yearly increase in emissions until 1984. After this, Scotchban emissions were assumed to decrease yearly to the present day. For the FTS mixture, emissions were assumed to begin in 1994, reach a peak in

2006 before being phased out, with emissions declining yearly. Assumptions, as well as details for the modelling are described in the *Modelling* section in the SI.

Details regarding the production methods at the factory are not known, however it is widely acknowledged that the paper industry produces effluent wastewater containing organic, suspended solids, rich in paper/cellulosic fibers (Ali and Sreekrishnan, 2001; Lacorte et al., 2003). As Scotchban and the FTS mixture were used as paper coaters, it is not unreasonable to assume they were sorbed to paper fibres when they were released (and therefore associated with organic carbon, which is a dominant parameter affecting sorption of PFAS (Higgins and Luthy, 2006)). As part of a sensitivity analysis, the model was run assuming three different organic carbon-water partitioning coefficients (Log K_{OC}: 5, 7 and 9), to describe sorption of Scotchban or FTS mixture to the particles entering the lake, as well as sorption to sediments. The value of 5 was chosen as it corresponds to locally measured values for preFOS and FTS in lake Tyrifjorden sediment (Table S21), which dominate the PFAS profile. The value of 9 was chosen to represent the very strong hydrophobic sorption of parent compounds (SAmPAP and FTMAP), which has not yet been measured but could be much higher (Wang et al., 2011b). A value of 7 was chosen as the mid-point.

Modelled emission volumes and sediment and water concentrations are shown in Table 1 for the two eras of product emissions, and for the three log K_{OC} values. When log K_{OC} was 5, emissions of Scotchban and the FTS mixture summed over all modelled years were 189 tons and 15.6 tons, respectively, with 154 and 13.7 tons leaving the lake by the downstream river. However, when log K_{OC} was 7, the back calculated emissions dropped drastically, to 42 and 2.5 tons respectively, and (in comparison) relatively minor emissions downstream: 0.8 and 0.07 tons, respectively. There was no substantial change in calculated emissions when log K_{OC} was 7 or 9, as at this point PFAS are essentially particulate bound (Table 1).

Regardless of the log K_{OC}, predicted total amounts of PFAS in sediments in the lake (ca. 35–41 tons of Scotchban and ca. 1.9–2.4 tons of the FTS mixture) agreed well with the extrapolated amount based on sediment measurements used for calibration (41 and 2.3 tons for Scotchban and the FTS mixture, respectively, see *equation VII* in the section *Modeling* in the SI for the calculation of measured volumes in the sediments). These emission volumes are substantial considering that previous estimates by Wang et al. (2017a) of total global emissions of PFOS, preFOS (*x*FOSA/Es) and POSF between 1958 and 2015 are in the ranges of 1228–4930, 1230–8738, and 670 tons respectively (Wang et al., 2017a). However, these emissions do not cover SAmPAP itself, but rather the building blocks and degradation products thereof (*x*FOSA/Es), and were mostly for air, which are most relevant for global distribution in a short time frame, and not lake sediments (Wang et al., 2017a). Thus, the global emission amounts could be much higher than estimated, when including local SAmPAP emissions and emissions to sediments, as discussed in Wang et al. (2017a). It has been reported that between 1.0 and 1.5% fluorochemical concentrations (based on the dry weight of the fibres) are needed for paper protection, and that approximately 32% of total PFOS produced in the European Union before 2000 was used for paper coating (United Nations Environment Programme Persistent Organic Pollutants Review Committee, 2010). In this context, modelled emissions for log K_{OC} values of 7 or higher seem more realistic, but still imply that the amount of SAmPAP and FTS in lake Tyrfifjorden is in the range of 0.5–3% of estimated global *x*FOSA/Es emissions. If correct, this would imply that, globally, local emissions of large PFAS such as SAmPAP in sediments and soils could be much larger than global emissions of *x*FOSA/Es, and therefore represent a continuous source of xFOSA/Es emissions in the future as these degrade.

Projected water concentrations in the years 2030 and 2060 were very low for all scenarios (below present day LOQ), which matches well with the assumption of strong sorption to particles. PFAS in top sediments originating from the use of Scotchban was projected to decrease up to 44% in 2030 and 48% in 2060 when compared to modelled concentrations in 2017. Similarly, the projected decrease was 43% in 2030 and 45% in 2060 for the FTS mixture. Rate constants (change in emission volumes in the lake via river input k_{in}), for the simulations at log K_{OC} 7.0/9.0 (Table S22) when concentrations of Scotchban and FTS mixture increased, were small, indicating near steady-state emissions when these products were used. Rate constants for the periods when concentrations of Scotchban and the FTS mixture, decreased, were larger, indicating a rapid decrease in emissions. Emission half-lives from the peak were on the scale of 4 years for Scotchban and 3 years for the FTS mixture. The extent to which these explain a decrease in direct factory emissions and a transition to diffuse emissions (e.g. from resuspension of river sediments or emissions from landfills) is unclear and should be re-evaluated in the future.

Both diffuse emissions from the landfill and soil as well as resuspension from top sediments are likely sources of long-term pollution to lake Tyrifjorden. Thus, despite the current low aquatic concentrations, PFAA exposure to biota is expected to be an issue for the foreseeable future. This is confirmed by the PFAS profiles seen in the sediment samples and the sediment traps, which exhibit a combination of both Scotchban and the FTS mixture composition, despite the likely shift from Scotchban to the FTS mixture around 1990 and the closure of the factory in 2013. The presence of both PFAS products in present day settling material in all lake sampling areas shows that mobilisation of the sediment is still occurring, resulting in a wide distribution of the PFAS pollution. Field results indicating a more complete transformation of PFAS furthest from the point source suggest that (some of) these legacy sources are subject to very little biotransformation, and that this occurs after PFAS are emitted from these sources.

4. Conclusions

PFAS profiles in samples representing emissions from the factory and PFAS profiles in river sediments directly downstream the factory were similar to PFAS profiles in lake sediments. In contrast, PFAS profiles in samples representing emissions from the fire station differed. PFAS profiles in biota were dominated by the same compounds and/or their expected biotransformation products as in lake sediments. The spatial distribution of concentrations and profiles (including PFOS isomer patterns) showed clear trends with distance from the factory, as expected based on PFAS physiochemical properties and biotransformation governing fate and transport. The dated sediment core showed distinct differences in the emitted PFAS mixture with time of release, and historical PFAS profiles matched well with known historical use of different PFAS for paper products (including SamPAP diester). Therefore, it is concluded that the factory is the main source to the PFAS contamination in lake Tyrifjorden. Results of the model show that emission volumes were very high, however due to strong sorption to particles, aquatic concentrations are low. Concentrations in top sediments will decrease over time, nevertheless, PFAS exposure to biota is expected to be an issue for the foreseeable future.

4.1. Environmental implications

The body of evidence in the present study indicates that production of paper products can be a major, largely overlooked, PFAS source to the environment. Both the overall environmental release, and local impacts on the environment and human health (e.g. from fish consumption) at such sites, point to the need for investigation of similar paper product production sites, as well as paper recycling and disposal facilities where these products might end up. Exploiting similar chemical profiling methods that allow source tracking and identification, as demonstrated here, is recommended. The inclusion of precursor compounds to PFAA in monitoring campaigns is necessary to capture the full environmental load. Based on these findings, there is a clear need for more rigid regulation of the use of PFAS in paper products and their potential release from the paper recycling industry.

Follow up studies should focus on the role that paper fibres can play in the fate, transport, and exposure of PFAS, as this information could be of importance for the assessment of PFAS related risks. There are currently no studies focusing on this specifically, however based on the literature of microplastic fibres (Thompson et al., 2004; Willis et al., 2017), studies on fibres in the oceanic water columns (Bagaev et al., 2017), as well as fluid dynamic theory (Wiens and Stockie, 2015), they appear to be readily suspended throughout the water column and are easily distributed through large water bodies like lakes, and only settle when aggregated. Hall (2003) presented a summary of research related to pulp mill effluent-induced coagulation and flocculation in rivers, showing that suspended solids downstream pulp mill discharges undergo coagulation and flocculation. The sedimentation of effluent fibres and their contaminants by coagulation and flocculation processes results in apparent decreasing concentration gradients in water and increased concentration gradients in sediments with downstream distance from the pulp mill (Hall, 2003). Similar research could not be found for paper production facilities, but it seems largely consistent with that observed in lake Tyrifjorden, warranting further research on the relevant coagulation and flocculation processes. It is reasonable therefore to hypothesize that the main transport mechanism by which the entirety of the sediment bed in lake Tyrifjorden was contaminated by PFAS, was products such as Scotchban and the FTS mixture being sorbed to emitted paper fibres. These fibres could then have been widely distributed throughout the entire volume of the lake until finally settling. Follow-up studies should thus explore if such paper fibres could be a major transport and exposure vector of PFAS pollution.

Associated content

Supporting information

The Supporting Information is available online.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.116259.

Credit author statement

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