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Bioavailability and bioaccumulation of perfluorinated compounds (PFAS) in a polluted river sediment

Photo: NIVA

Norwegian Institute for Water Research

REPORT

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Summary

An experiment was performed to contribute to the environmental risk assessment of the perfluorinated compounds (PFAS) in sediments collected nearby a closed down factory in Oppland, Norway. The sediments were placed in replicate aquaria continuously flushed with freshwater. More than 30 different PFAS compounds were analyzed for determination of fluxes from sediment to water and concentrations in sediments, pore water and two benthic species (an oligochaete *Tubifex tubifex* and a mussel *Anodonta anatine*) after four weeks exposure in the sediments. Compared to aquaria with control sediments from Lake Årungen and a location in the river upstream of the old factory site, high fluxes and high concentrations of PFAS were found in sediments, pore water and oligochaetes, in aquaria with sediments collected nearby the factory site. Concentrations of PFOS were classified as class III "risk of chronic effects from longterm exposure" at the two most contaminated locations. Concentrations of PFOA did, however, not exceed class II "no toxic effects" in any of the sediments used in this study. By simple division of the estimated sediment reservoir by the fluxes observed in this study, depletion times were obtained ranging from 0.5 years for short chain carboxylates to more than 5000 years for the 12C fluorotelomer. Indications were found that the condition for both oligochaetes and mussels were reduced in the sediments collected in the river adjacent to the factory site. This was more likely due to high concentrations of petrogenic hydrocarbons than high concentrations of PFAS.

Four keywords	Fire emneord
1. PFAS	1. PFAS
2. Sediment	2. Sediment
3. Fluxes	3. Flukser
4. Bioaccumulation	4. Bioakkumulering

This report is quality assured in accordance with NIVA's quality system and approved by:

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Bioavailability and bioaccumulation of perfluorinated compounds (PFAS) in a polluted river sediment

Preface

In this project NIVA has worked on a contract for Rambøll, assignment 1350029503-03 “Sediment flux and bioaccumulation”. The assignment was part of a risk assessment of contaminated soil and sediments at an old factory site at Viul in Oppland, S.E.Norway, performed by Rambøll for Huhtamäki Oyj. The site has been used for production of coated paper items. The report describes set-up and performance of an experiment aimed at measuring fluxes and bioaccumulation of perfluorinated compounds from sediments collected in a river running by the old production site. The experiment was designed at the request and in cooperation with Tom Tellefsen and Aud Helland, Rambøll, and performed at NIVA Research Facility Solbergstrand with Morten Thorne Schaanning as NIVA’s project manager. Joachim Tørum Johansen and Pål Fasseland, NIVA, was responsible for collection and keeping of mussels and for the daily maintenance and sample collection during the experimental period. Marthe Torunn Solhaug Jenssen assisted during final sampling with particular responsibility for handling and preparation of the biological samples. Jan Thomas Rundberget was responsible for the chemical analysis of PFAS compounds performed at NIVAs laboratory in Oslo. Research Manager Marianne Olsen has quality assured the report.

Oslo, 26.02.2020

Morten Thorne Schaanning
Project Manager

Summary

An experiment was performed to assess the bioavailability of perfluorinated compounds (PFAS) in sediments from Randselva nearby an old factory site in Ringerike municipality, Oppland county, Norway. The work performed at NIVA's research station at Solbergstrand, was a contribution to Rambøll's risk assessment of soil and sediments at the site of the closed down factory.

Sediments potentially affected by contaminants from the closed down factory were collected in the river outside the factory site (Ot), ca 1 km further down in Svarthølen (Ba) and another 1 km downstream the Viul hydropower dam (Dw). The sediments were placed in three replicate aquaria continuously flushed with water from a large tank filled with groundwater from a local well. For comparison, aquaria with sediments from Lake Årungen, Akershus county, and an upstream location in Randselva (Up) were included in the set-up. Clams (*Anodonta anatine*) and sediments were collected at the same location in Lake Årungen. The oligochaete worms (*Tubifex tubifex*) were supplied from a commercial fish feed producer. Both organisms were added to all aquaria and retrieved for analyses of PFAS in soft tissues after four weeks of exposure in the sediments. Both organisms spend most of their time buried in the sediments, but the clams feed by filtering aquarium water supplied through a siphon and the hard shell protects the soft tissues from immediate uptake from sediment and pore water.

More than 30 different PFAS compounds were analyzed in five different matrixes: clams, worms, sediments, pore water and aquarium water. All matrixes revealed high abundance of carboxylates (PFCAs), FOSAMs (degradation products of SAmPAP esters produced before 2002), and fluorotelomer sulfonates (FTSAs) which came into use mostly after 2002. Because the clams are less exposed to sediment and pore water than the worms, clams accumulated small amounts of PFAS compared to the worms. In accordance with the classification system for Norwegian freshwater, concentrations of PFOS corresponded to environmental class III "risk of chronic effects from long term exposure" in sediments collected outside the factory site (Ot) and in Svarthølen (Ba). PFOA did not exceed the upper boundary for class II in any of the sediments used in the present investigation.

Sediment:pore-water partition coefficients (K_d), biota:water concentration factors (BCFs) for clams and biota:sediment accumulation factors (BSAFs) for the worms were calculated. Consistent with cited literature, K_ds and BCFs, but not BSAFs, were positively correlated with increasing molecular size. Poor survival and anomalous BSAFs were found for the worms exposed in sediments from the most contaminated location (Ot). These sediments contained numerous small plastic fibres and were distinctly contaminated with petrogenic hydrocarbons. Both these factors may have contributed to the anomalous BSAFs.

Fluxes of PFAS determined in ng m^{-2} h^{-1} were calculated from concentrations in the aquarium water, water exchange rates and the sediment surface area. Depletion times were subsequently calculated from the flux and estimated contaminant loads in the sediment at Ot and Ba. Depletion times were positively correlated with molecular size and ranged between 0.5 years for short chain carboxylates to more than 5000 years for the 12C fluorotelomer.

Sammendrag

Tittel: Biotilgjengelighet og bioakkumulering av perfluorinerte forbindelser i et forurenset elvesediment.
År: 2020
Forfatter(e): Morten Thorne Schaanning, Jan Thomas Rundberget, Marthe Torunn Solhaug Jenssen
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Et eksperiment ble utført for å vurdere biotilgjengelighet av perfluorerte forbindelser (PFAS) i sedimenter fra Randselva i nærheten av den tidligere papirfabrikken på Viul i Ringerike kommune, Oppland. Arbeidet, som ble utført ved NIVAs forskningsstasjon på Solbergstrand, var et bidrag til Rambølls risikoanalyse av grunn og sedimenter i området rundt den nedlagte fabrikken.

Sedimenter med mulig påvirkning av forurensing fra fabrikkområdet ble innsamlet rett utenfor fabrikken (Ot), ca 1 km lenger ned i Svarthølen (Ba) og ytterligere ca 1 km nedstrøms demningen (Dw). Sedimentene fra hver lokalitet ble plassert i tre replikate akvarier med kontinuerlig tilførsel av vann fra en stor tank fylt med grunnvann fra en lokal brønn. Til sammenligning ble akvarier med sedimenter fra Årungen i Akershus og en oppstrøms lokalitet i Randselva (Up) inkludert i oppsettet. Dammuslinger (*Anodonta anatine*) innsamlet i Årungen fra samme sted som sedimentprøven i kontrollakvariet (Con) og fåbørstemarken (*Tubifex tubifex*) fra en kommersiell dyrefôrprodusent, ble tilsatt alle akvariene der de ble eksponert fire uker i sedimentene før innsamling og analyser av PFAS. Begge organismer oppholder seg mesteparten av tiden nedgravet i sedimentene, men muslingen har næringsopptak via sifong fra vannet over sedimentet og skjellene skjermer bløtdelene mot direkte opptak fra sediment og porevann.

Mer enn 30 forskjellige PFAS-forbindelser ble analysert i fem forskjellige matrikser: dammusling, børstemark, sedimenter, porevann og akvarievann. Alle matrixer viste høye konsentrasjoner av karboksylater (PFCA), FOSAMs (PFOS og andre nedbrytningsprodukter av SAmPAP estere produsert før 2002), og fluorotelomer sulfonater (FTSA) produsert hovedsakelig etter 2002. Sammenlignet med det norske klassifiseringssystemet for ferskvann, tilsvarte konsentrasjonene av PFOS klasse III «risiko for kroniske effekter fra langtids eksponering» i sedimenter innsamlet utenfor og nedstrøms fabrikkområdet (stasjon Ot og Ba). Konsentrasjonene av PFOA overskred ikke grenseverdiene for klasse II i noen av sedimentene benyttet i denne undersøkelsen.

Sediment:porevann partisjonskoeffisienter (K_d), biota:vannkonsentrasjonsfaktorer (BCF) for muslinger og biota:sediment akkumulasjonsfaktorer (BSAF) for børstemarkene ble beregnet. I overenstemmelse med referert litteratur, var K_d og BCF men ikke BSAF, positivt korrelert med økende molekylstørrelse. Dårlig overlevelse og uvanlige BSAF-verdier ble observert for børstemark eksponert i sedimenter fra den mest kontaminerte lokaliteten (Ot). Disse sedimentene inneholdt også plastikkfiber fra den tidligere produksjonen ved fabrikken og var tydelig kontaminert av petrogene hydrokarboner som kan ha større akutt toksisk effekt enn PFAS.

Flukser av PFAS målt som ng $m^{-2}h^{-1}$ ble beregnet fra konsentrasjonene målt i akvariumvannet, utskiftingshastigheten av akvariumvannet og arealet av sedimentoverflaten. Tømmetider (den tiden det vil ta før det ikke er forurensing igjen i sedimentet) ble deretter beregnet fra de målte fluksene og estimerte forurensningsmengder i sedimentreservoarene utenfor fabrikken (Ot) og i Svarthølen (Ba). Tømmetidene var positivt korrelert med molekylstørrelsen og varierte fra 0,5 år for kortkjedede karboksylater til mer enn 5000 år for 12C fluorotelomerene. De beregnede tømmetidene er underestimert i den grad fluksene vil avta etter hvert som konsentrasjonene i sedimentene avtar.

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1 Introduction

The work described in this report is based on sediment samples collected at a closed down paper mill at Viul between Jevnaker and Hønefoss in Ringerike municipality, Oppland county, Norway. The location has been identified as a potential point source for perfluorinated compounds (PFAS) (Grøndal et al., 2019).

The common structure of PFAS molecules is a fluorinated alkyl chain with a hydrophilic end group -X in Figure 1. The PFAS compounds are divided into sub-groups based on the hydrophilic end group. This may be carboxylic acid (-COOH), sulfonic acid ($-HSO_3$), sulfonamides ($-SO_2NH_2$), or more complex structures. The acids are often referred to as dissociated forms, carboxylates ($-COO^-$) or sulfonates ($-SO_3^-$). Another important group is the fluorotelomeres (Wang et al., 2011) which apparently came into production after production of PFAS and higher homologues was terminated in 2002. In fluorotelemors an alkyl with variable carbon chain length is inserted between the fluorinated chain and the X-group.

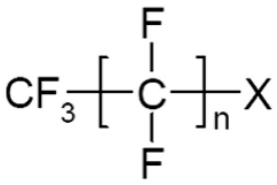


Figure 1. General structure of a perfluorinated compound. (After Arvaniti and Stasinakis, 2015).

PFAS are man-made chemicals which may enter the aqueous environments during manufacturing, use and disposal of various industrial and consumer products (Ahrens, 2011). Being persistent, bioaccumulating and potentially toxic to humans, PFAS comply to the definition of an environmental contaminant. The two most common congeners, often accounting for more than 50% of the total PFAS in European rivers, are perfluoro-octanoic acid (PFOA) ($C_7F_{15}COO^-$) and perfluoro-octane sulfonic acid (PFOS)($C_8F_{17}SO_3^-$). PFOS and its derivatives has been identified by the European Union as priority hazardous substance and prohibited for general use since 2008 (EU directive, 2006/122). Environmental quality standards (EQS) for inland surface waters and biota were defined in EU directive 2013/39. EQS values implemented in Norway for PFOS and PFOA are shown in Table 1.

Table 1. Environmental quality standards (EQS) for PFOS and PFOA in inland water and sediment as given in Norwegian guidelines (Veileder 02:2018).

	Class I Background	Class II AA-EQS No toxic effects	Class III MAC-EQS Chronic effects from longtime exposure
Water			
PFOA (ng L^{-1})		0-9100	>9100
PFOS (ng L^{-1})		0-0.65	0.65 – 36000
Sediment			
PFOA (ng g^{-1} d.wght.)		0-713	>713
PFOS (ng g^{-1} d.wght.)		0-2.3	2.3-360

In addition, the guidelines provide a quality standard for PFOS in organisms of 9.1 ng g^{-1} ww and PFOA in biota of 91.3 ng g^{-1} ww.

In order to better understand the risk posed by the contaminated sediments to downstream ecosystems, an experiment was performed to assess the bioavailability of PFAS stored in the contaminated sediments.

Bioavailability is the tendency of a contaminant to move from a solid phase with no harmful effects into biological tissues within which it may cause harmful effects to the organism. Bioavailability is often described in terms of partitioning equilibria between different environmental matrixes and calculated as the concentration in one matrix (sediment, organism) divided by the concentration in another matrix (water, pore water). Desorption from the solid phase is the first step towards uptake in an organism. Therefore, a high sediment water partitioning coefficient ($K_d = C_{sed}/C_{water}$) indicates low bioavailability of a given contaminant. More direct measure of bioavailability is the BCF (Biota Water Concentration factor) or BSAF (Biota Sediment Accumulation Factor) determined for a given specie or group of species. The flux from the sediment to the overlying water is a useful parameter for biogeochemical modelling but may also be considered an indicator on bioavailability. A high flux from the sediment to the overlying water indicates high risk to organisms living both in the sediments and in free water masses. Because these parameters often depend on organic carbon and other environmental parameters, the estimated coefficients are often site-specific and strictly valid only for the specific site studied.

In this study, PFAS were measured in five matrixes: sediments, pore water, aquarium water, clams and worms. The clams (*Anodonta anatine*) were protected from the sediment by a hard shell and feeding from aquarium water through a siphon. The worms (*Tubifex tubifex*) were living buried in the sediments exposed to the pore water through a thin body membrane and feeding by ingesting whole sediment. Bioavailability was indicated both by the abiotic parameters

1. flux from sediment to aquarium water, and
2. sediment:pore water partitioning coefficients (K_d)

and the biotic parameters

3. BSAF for the worms, and
4. BCF for the clams.

2 Material and methods

2.1 Source sediment collection

Sediment samples were collected by Rambøll 26.-28.06.2018 at four locations – in Randselva upstream (Up) and outside (Ot) the closed down factory site, in the Svarthølen basin (Ba) and in the river downstream the Viul hydropower dam (Dw) (Figure 2). About 10 kg of the upper 2 cm of the sediment from each location was collected in four transport boxes with lids. When sampling at Ot, an oil film occurred on the water surface and the sediment smelled distinctly of hydrocarbons that easily evaporate (Tom Tellefsen, pers.com.). The samples were kept cool in the shadow during field work and transportation, and in cold-storage chambers at +4 ^{o}C between sampling and start of the experimental work. The samples were delivered at NIVA Research Facility Solbergstrand the week after collection.

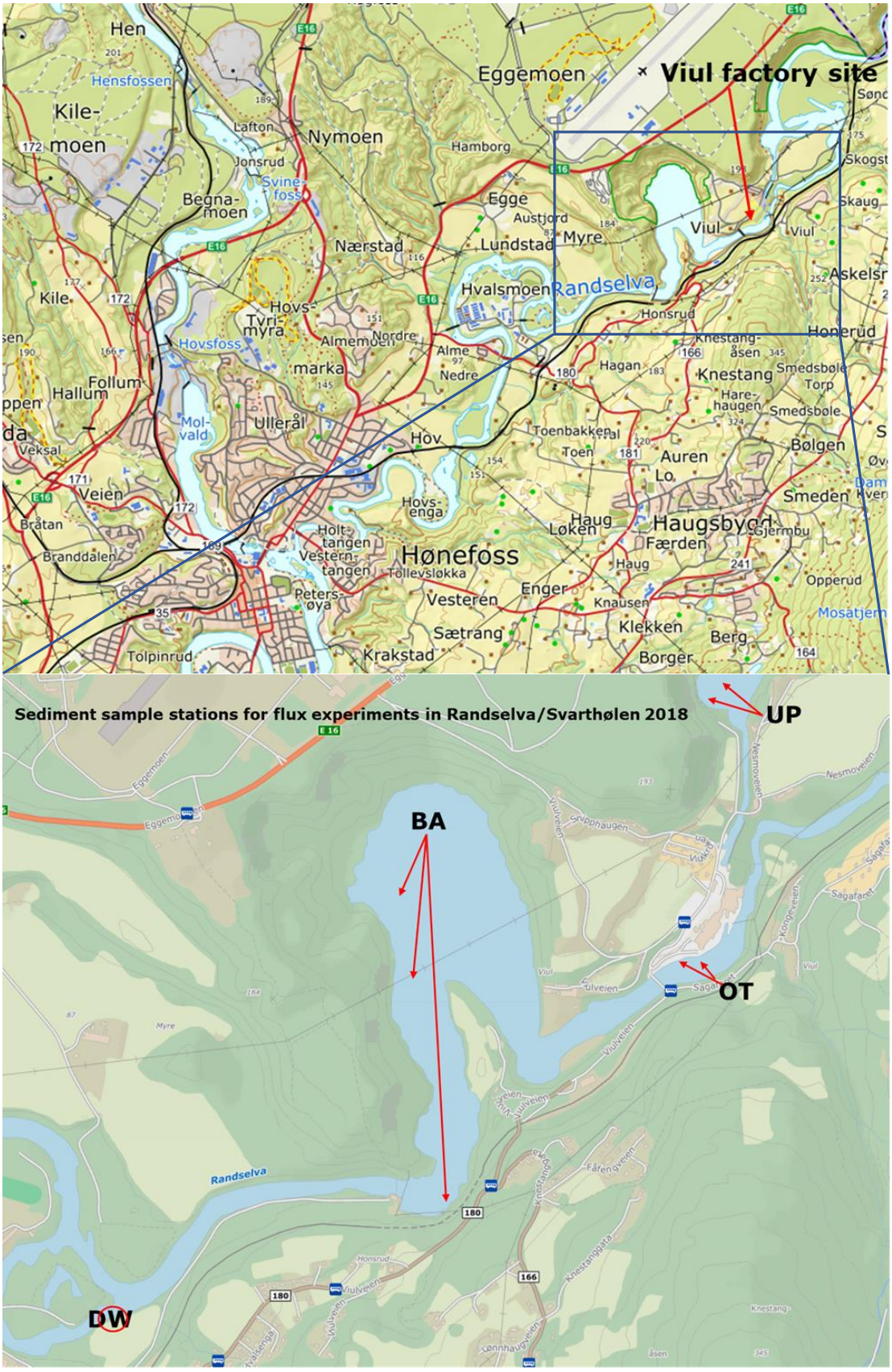


Figure 2. Sampling stations at Viul. The samples from Up, Ot and Ba were a mixture of 2-4 samples collected at the point of the arrows.

2.2 Experimental set-up

The experiment was performed in a light and temperature regulated laboratory at NIVA Research Facility Solbergstrand (isotope laboratory). The room temperature and supply water were kept at about 10^{o}C and the light was switched off during night (20:00-08:00).

The set-up included 13 glass aquaria measuring 15x20x22 cm (BWD) continuously supplied with a flow of freshwater from a header tank via a 20-channel Watson-Marlow® peristaltic pump (Figure 3). Tubing materials were marprene across the pump head and 2 mm (ID) PVC on either side of the pump. The header tank was continuously supplied from a large tank with fresh water sampled from the stream flowing by the research station. Analyses of samples drawn from the tank water three times during the experiment showed concentrations below detection limits of all compounds except for PFOA which showed consistent presence of 0.7-3.2 ng L^{-1}.

In addition to three replicate aquaria[1] for each of the four sampling locations one aquarium was set up with control sediment from Lake Årungen (Con). The reason for including this treatment was that this was the only known site in our area at which clams for the bioaccumulation experiment were available mid-winter. Lake sediments without any trace of PFAS may be hard to find and not required for validation of the results on fluxes and bioaccumulation. Both Up and Con treatments should be considered natural reference locations with relatively low contaminant levels and no direct impact of discharges from the factory site.

The experiment was set up on 15.01.2019 (day 0). The sediment in each transport box was thoroughly mixed using an electric stirring device. Aliquots of 2 kg wet sediment was placed in an even layer in the bottom of each aquarium. Sediment sticking to the walls was carefully washed off with water from the header tank and allowed to settle on the sediment surface in each aquarium. The aquaria where then slowly filled up with water delivered via the peristaltic pump set at 20 rpm (rotations per minute) which corresponded to a nominal flow of 2.8 ml min^{-1}. This was later adjusted to 10 rpm and further down to 5 rpm based on design sampling and analyses of the overlying water during the first few weeks (Table 2).

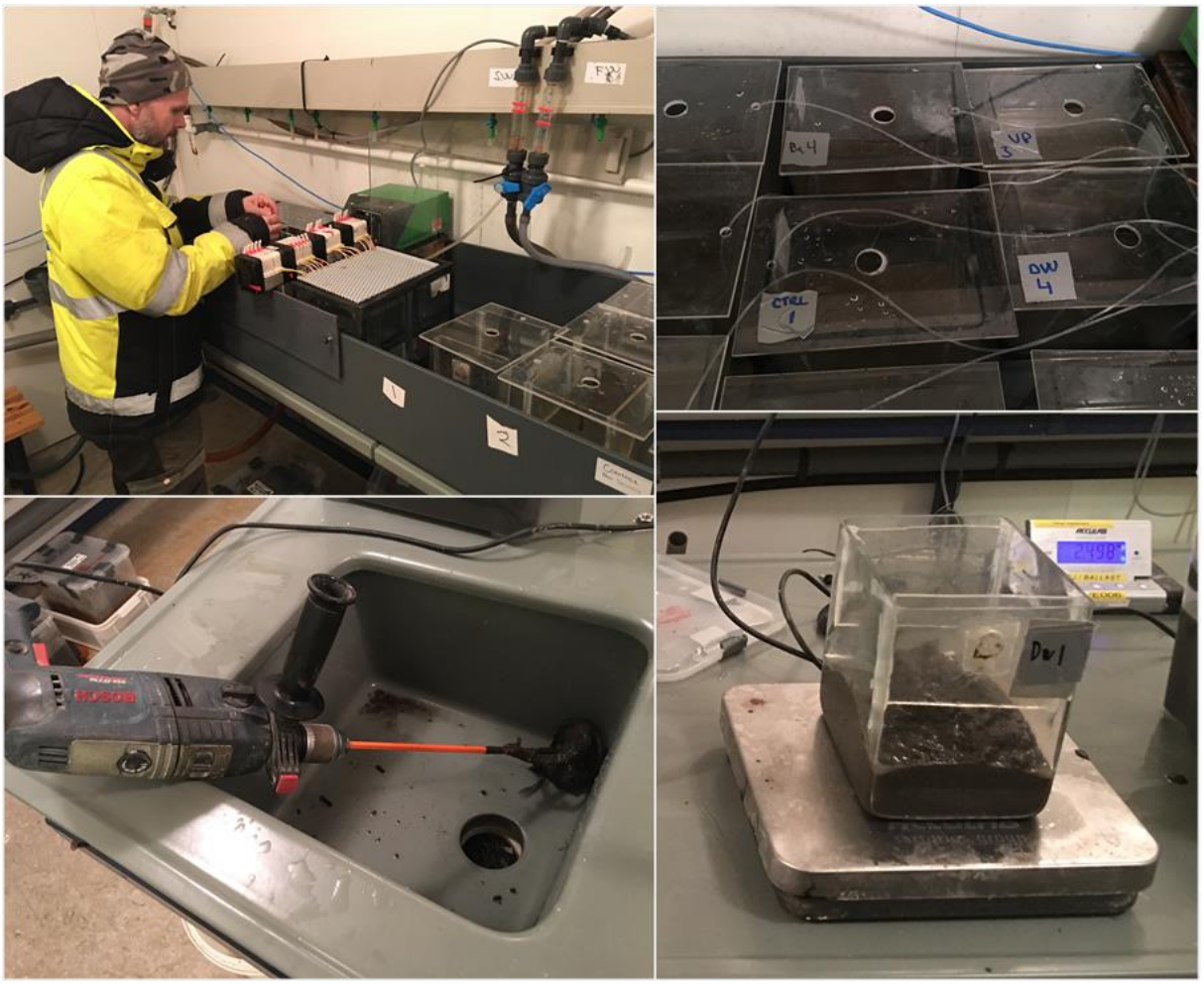
Aeration was applied using air stones placed 2 cm above the sediment inside the open end of a vertical tube. Air bubbles inside the tube generated a circulation system with water entering at the bottom of the tube and leaving horizontally at the surface. Thus, the aeration system served a dual purpose of maintaining oxygen supply for benthic respiration and stirring to avoid stratification of the water column. The air supply was adjusted to avoid resuspension of sediment particles.

2.1 Organisms

The experiment was initiated mid-winter when availability of experimental animals from wild populations are limited. In joint considerations with the contractor it was agreed to go for a wild population of clams (*Anodonta anatine*) from Lake Årungen in Akershus and an oligochaete (*Tubifex tubifex*) sold commercially as live food for aquarium fish. The oligochaete is a subsurface deposit feeder, about 1 cm long with diameter about 0.1 cm. They were delivered in 10 ml plastic bags filled

[1] Fig. 3 shows the real set-up with four replicate aquaria for each treatment. The fourth aquarium was included as a spare replicate in case of loss of aquaria due to breakage or other unforeseen events. No data from the spare aquaria have been used in this report.

with water and yeast and about 4 g of live organisms in each bag. Two bags were distributed to each aquarium on day 52 by emptying the bags in the overlying water. The worms made their way into the sediments and after a few hours they were no longer seen in the water or on the sediment surface. Simultaneously, two randomly selected bags – zero samples - with Tubifex worms were frozen and



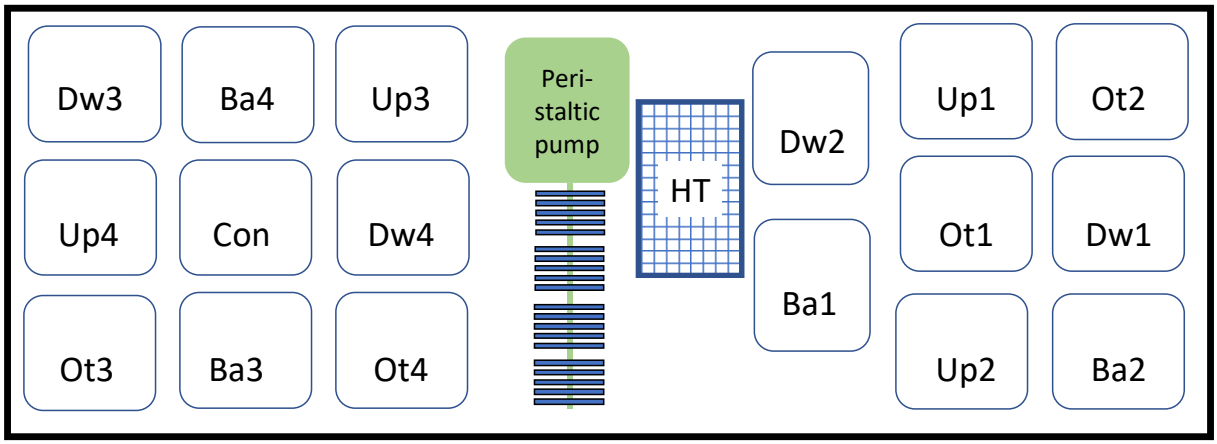


Figure 3. Experimental set-up. Schematic design in lower frame. Note that aquaria Up4, Ot4, Ba4 and Dw4 were spare replicates (not sampled).

Figure 4. Clam (*Anodonta anatine*) in depuration beaker. The particles had all been excreted during the depuration period.

Table 2. Time schedule of experimental work and sampling between the 15th of January until 9th of April 2019 at Solbergstrand research station. X = done. Numbers are pump speed (rate of water exchange) or number of samples

Day relative to 15.01.2019	-11	0	3	20	30	38	52	77	78	79
Preparation of laboratory set-up	x--	x								
Addition of sediment		x								
Flow rate (rpm)		On 20	-10-	-10-	--5--	-----	-----	-----	-off	
Aeration			on--	-----	-----	-----	-----	-----	-off	
Sediment samples from transport boxes and Årungen control		5								
Design sampling and analyses of water from Dw1, Ba1 and HT				3--	--x					
Water samples, 13 aquaria + HT						14	14	14		
Addition of organisms, zero samples of clam + worms							2			
Sampling of sediments, 13 aquaria								13		
Extraction of pore water, 13 aquaria								13		
Overnight depuration and sampling, 13 clams + 5 worms									x--	18

stored till the end of the experiment. The clams were added on the same day. One to each aquarium. One randomly selected clam – zero sample - was placed in freezer until the end of the experiment. The clams positioned themselves almost completely buried in the sediment with the siphon sticking up a few mm above the sediment surface.

2.2 Sampling

Flux measurements are determined by the concentration difference between the in- and out-let water to the aquaria. This again, depends on the water exchange rate. Therefore, in order to optimize water exchange rate, initial (design) analyses of water from selected sampling points were performed during the first month of the experiment (section 3.1). At the same time analyses were performed on sediment sampled from the transport boxes and Lake Årungen control. These samples confirmed high concentrations of PFAS at station Ot and not detectable concentrations at station Con and Up (Table 5, Table 6). In the water, several PFAS compounds were detectable in aquaria with sediments from Dw and Ba (Ot not sampled). These concentrations are steady state concentrations determined by the rate of leakage from the sediment to the overlying water and the rate of exchange of the overlying water. To improve the detection limit of the flux of PFAS, the pump rate was reduced to 5 rpm after the result of the design samples were made available from the laboratory.

After the initial analyses were completed, water samples were drawn with a siphon from header tank (HT) and the center of each aquarium on days 32, 52 and 77. After water sampling on day 77, the remaining water was sifted off and 50 ml wet sediment was transferred to centrifuge tubes and centrifuged for 20 min at 6000 rpm and 5^{o}C in a Sorval® centrifuge. The clear supernatant was removed with pipette to 15 ml vials, carefully avoiding any particles coming off the wall of the centrifuge tubes.

Organisms were added after water sampling on day 52 and retrieved after sediment sampling on day 77. Four weeks is a typical exposure time frequently applied in bioaccumulation tests (Ruus et al., 2005). The mussel was transferred to a depuration beaker with ca 500 ml of the set-up source water. The worms were separated from the sediments by washing through sifts with pore size down to 0.5 mm. The material was then transferred to white trays and the worms picked up one by one with a pincer and placed in separate depuration beakers (ca 250 ml set-up source water). All organisms were left over night in the depuration beakers. Length, width and thickness of the mussels were measured using a digital slide caliper, before opening with scalpel and drip-dried ca 15 min over white paper. Free water on body surface of worms were removed on a blotter. Wet weight of soft tissues of the mussels (one individual from each aquarium) and worms (pooled from each treatment) were determined before transfer to burnt glass vials and placed in freezer.

Between sampling and chemical analyses, water, pore water and sediment samples were stored in dark cold chamber at 4^{o}C. Biota samples were stored in a dark freezer at -20^{o}C.

Size of the mussels were calculated as square volume (length x width x thickness) and a condition index calculated as the weight of soft tissue divided by the weight of the hard shell. The condition index varied from 0.90 in Con to 1.19 in Up (Figure 5). The total weight of the worms collected from each treatment at the termination of the experiment, ranged from 0.15 g (wet weight) in Ot to 2.18 in Dw (Table 3). To some extent, the weight of each sample was inversely related to sampling effort and thus to the survival of worms in each treatment. Most effort was spent in obtaining sufficient amount of sample from Ba and Ot and the number of worms survived was clearly smallest in Ot.

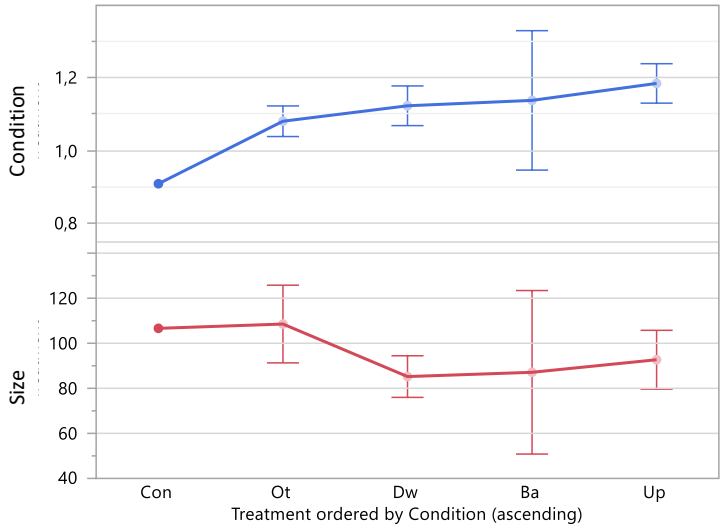


Figure 5. Size calculated as cm^3 square volume and condition (weight of soft tissue/weight of hard shell) of the mussel *Anodonta anatine* determined after four weeks exposure in sediments from the Viul site (Up, Dw, Ba, Ow) and Lake Årungen (Con).

Table 3. Wet weight of the oligochaete worms (*Tubifex tubifex*) retrieved after 4 weeks exposure in sediments from the Viul site (Up, Dw, Ba, Ow) and Lake Årungen (Con) compared to the amount added on day zero. The day zero sample was never exposed to any sediments.

Treatment	g w.w.
Con	1.63
Up	1.38
Dw	2.18
Ba	0.78
Ot	0.15
Zero (1 bag)	3.86

2.3 Analytical methods

In this work we have analysed 31 different congeners (Table 4) including nine carboxylic acids (PFCAs), five sulfonic acids (PFSAs), three sulfonamides (PFOSAs), two sulfonamidoethanols (PFOSEs), five fluorotelomeresulfonates (FTSAs) and three sulfonamide acetic acids (PFOSAAs). Limits of quantification (LOQ) in the sample matrixes, water (and pore water), sediment and biota are given in Table 4. Seven compounds were not detected above LOQ in any of the samples. These compounds are omitted throughout the remaining of this report.

2.3.1 Sample preparation

An aliquot of about 2 g of homogenized biota, or 5 g of wet sediment, was spiked with 6 ng each of the mass-labelled internal standards. The sample was extracted with 7 mL of acetonitrile for 30 min in an ultrasonic bath. Following centrifugation, the supernatant extract was removed, and the

Table 4. Analyzed compounds, compound groups and limits of quantification (LOQ). Items in grey italic were below LOQ in all samples and omitted from further comments in this report. Compound groups are separated by broken lines and group sums of concentrations are denoted ∑PFCAs etc.

			LOQ in:	Water	Sediment	Biota
Short name	n	Chemical name		ng/L	ng/g	ng/g
PFHxA	C6	perfluoro-n-hexanoic acid		0.5	0.5	0.5
PFHpA	C7	perfluoro-n-heptanoic acid		0.5	0.5	0.5
PFOA	C8	perfluoro-n-octanoic acid		0.5	0.5	0.5
PFNA	C9	perfluoro-n-nonanoic acid		0.4	0.4	0.4
PFDA	C10	perfluoro-n-decanoic acid		0.4	0.4	0.4
PFUnDA	C11	perfluoro-n-undecanoic acid		0.4	0.4	0.4
PFDoDA	C12	perfluoro-n-dodecanoic acid		0.4	0.4	0.4
PFTrDA	C13	perfluoro-n-tridecanoic acid		0.4	0.4	0.4
PFTeDA	C14	perfluoro-n-tetradecanoic acid		0.4	0.4	0.4
∑PFCAs		Sum carboxylates				
PFBS	C4	perfluoro-1-butanesulfonic acid		0.1	0.1	0.1
PFPeS	C5	perfluoro-1-pentanesulfonic acid		0.1	0.1	0.1
PFHxS	C6	perfluoro-1-hexanesulfonic acid		0.1	0.1	0.1
PFHpS	C7	perfluoro-1-heptanesulfonic acid		0.1	0.1	0.1
PFOS	C8	perfluoro-1-octanesulfonic acid		0.1	0.1	0.1
∑PFSAs		Sum sulfonates				
PFNS	*C9*	*perfluoro-1-nonanesulfonate*		*0.1*	*0.1*	*0.1*
PFDS	*C10*	*perfluoro-1-decanesulfonate*		*0.1*	*0.1*	*0.1*
PFDoS	*C12*	*perfluoro-1-dodecansulfonate*		*0.2*	*0.2*	*0.2*
8-ClPFOS	*C8*	*8Cl-perfluoro-1-octanesulfonate*		*0.2*	*0.2*	*0.2*
PFOSA	C8	perfluoro-1-octanesulfonamide		0.1	0.1	0.1
N-MeFOSA	*C8*	*N-methylperfluoro-1-octanesulfonamide*		*0.2*	*0.2*	*0.2*
N-EtFOSA	C8	N-ethylperfluoro-1-octanesulfonamide		0.2	0.2	0.2
∑PFOSAs		Sum sulfonamides				
N-MeFOSE	C8	2(N-methylperfluoro-1-octanesulfonamido)-ethanol		2	2	2
N-EtFOSE	C8	2(N-ethylperfluoro-1-octanesulfonamido)-ethanol		2	2	2
∑PFOSEs		Sum sulfonamido-ethanols				
4:2 FTS	*C4*	*1H,2H-perfluorohexan sulfonate (4:2)*		*0.3*	*0.3*	*0.3*
6:2FTS	*C6*	*1H,2H-perfluorooctane sulfonate (6:2)*		*0.3*	*0.3*	*0.3*
8:2 FTS	C8	1H,2H-perfluorodecan sulfonic acid (8:2)		0.3	0.3	0.3
10:2 FTS	C10	1H,2H-perfluorododecan sulfonic acid (10:2)		0.3	0.3	0.3
12:2 FTS	C12	1H,2H-perfluorododecan sulfonic acid (12:2)?		0.3	0.3	0.3
∑FTSAs		Sum fluorotelomer sulfonic acids				
FOSAA	C8	perfluoro-1-octansulfonamidoacetic acid		0.3	0.3	0.3
N-MeFOSAA	C8	2(Nmethylperfluoro-1-octansulfonamido)acetic acid		0.3	0.3	0.3
N-EtFOSAA	C8	2(N-ethylperfluoro-1-octansulfonamido)acetic acid		0.3	0.3	0.3
∑PFOSAAs		Sum sulfonamido-acetates				

extraction was repeated with another 5 mL of acetonitrile. The combined acetonitrile extract underwent dispersive clean-up with graphitized carbon and acetic acid. A volume of 0.4 mL of the cleaned-up extract was added to 0.2 mL of aqueous ammonium acetate. The final extract was centrifuged before a clear supernatant was transferred to an autoinjector vial.

Internal standards were added to the water sample (0.4 L) before extraction using a MeOH activated (200mg) HLB solid phase extraction cartridge (Waters). The analytes were eluted of the HLB with MeOH. The MeOH extract was evaporated under nitrogen and resolved in 60+40 MeCN and 5.2 mM ammonium acetate (aq).

2.3.2 Instrumental analysis

An Acquity Ultra Performance HPLC system (Waters) was used to inject aliquots of 7 µl extract onto a Waters Acquity BEH C8 reversed phase column (100 x 2.1 mm, 1,8 µm particles. The target compounds were separated at a flow rate of 0.5 ml/min using acetonitrile (A) and 5.2 mM NH4OAc in water (B). The following binary gradient was applied: 0-1.5 min, 12% of A; 1.5-11 min, linear change to 99% of A; 11-13 min, 99% of A.

The Xevo G2-S Q-ToF-HRMS instrument (Waters) was employed in negative ion electrospray ionization (ESI(-) mode. Mass spectra were registered in full scan mode (mass range m/z 150-1100). The following optimized parameters were applied: Capillary voltage, 0.7 kV; desolvation temperature, 500 °C; source temperature, 120 °C; nitrogen desolvation gas flow, 800 L/h. Quantitative analysis was performed employing extracted mass chromatograms from full scan recording using the m/z (typical mass tolerance of 0.03 Da) for the different analytes.

2.4 Calculations

Data handling and statistical analyses were performed using JMP®v13 statistical software from SAS-institute.

The fluxes (F) were calculated based on the following equation:

$$F = (C_i - C_o) \times Q/A$$

where

C_i = concentration in header tank,
C_o = concentration in aquarium water,
Q = flow rate measured as weight of water collected for 30 minutes
A = sediment area in aquarium.

A more comprehensive description of flux calculations can be found in Schaanning et al. (2008).

3 Results

3.1 Preparatory analysis

The residence time of the water in the aquaria will determine the concentration of PFAS in the aquarium water. A short residence time is good for the environmental condition in the aquaria, but too short will increase the uncertainty in the concentration difference between in- and outlet water. To optimize the pump rate a test period was run before sampling for design analyses of the water.

Also, preparatory analyses to confirm PFAS compounds present in the sediments collected were done. The results of the preparatory analyses are shown in Table 5 and Table 6. These data are, however, not further used in this report.

Table 5. Preparatory analyses of sediment samples collected from Lake Årungen (Con) and Randselva (Up, Dw, Ba and Ot). Samples were drawn 23.01.2019, before set-up of the experiment. Station locations in Randselva are shown on map in Figure 2.

Sediment	Con	Up	Dw	Ba	Ot
	ng/g	ng/g	ng/g	ng/g	ng/g
PFOS	<0.1	<0.1	0.4	3.2	37
PFOSA	<0.1	<0.1	<0.1	8.9	26
etFOSAA	<0.1	<0.1	0.2	175	341
10:2 FTS	<0.1	<0.1	<0.1	134	>1000
12:2 FTS	<0.1	<0.1	<0.1	233	>1000
SAmPAP	<0.1	<0.1	0.3	118	119
PFDA	<0.1	<0.1	<0.1	2.7	432

Table 6. Preparatory analyses of water samples collected from the source water (header tank) and two selected aquaria (Dw1 and Ba1) 06.02.2019 at pump rate 10 rpm.

Water	Dw1	Ba1	Header tank (HT)
	ng/L	ng/L	ng/L
PFOS	1.49	0.18	0.25
PFOSA	0.05	0.22	<0.05
etFOSAA	0.45	1.79	<0.1
10:2 FTS	<0.1	0.98	<0.1
12:2 FTS	<0.1	0.75	<0.1
SAmPAP	<0.05	0.09	<0.05
PFDA	0.51	<0.1	<0.1

3.2 Fluxes and concentrations in sediment and pore water

All groups of PFAS were present at relatively high concentrations in sediments collected outside the factory site (Ot) and in Svarthølen basin (Ba) (Figure 6-Figure 9). At the station downstream of the hydropower dam (Dw), concentrations were lower, but residues of a few compounds were detectable. These are important as they may indicate more refractory components of the PFAS originating from the old factory. Only a few PFCAs (mainly PFOA) were present above LOQ in sediments from the upstream station (Up) and Lake Årungen (Con). Below, the observations of fluxes and concentrations in sediments and pore water are reported for each of the five groups specified in Table 4 and presented in detail in Figure 7 - Figure 9.

3.2.1 Fluorotelomere sulfonates (FTSAs)

The fluorotelomere sulfonates (FTSAs) were the most abundant compounds in river sediments and pore waters collected outside the factory site (Ot) and in the Svarthølen basin (Ba) (Figure 6), but below LOQ in aquaria with sediments from the upstream location (Up) and Lake Årungen (Con). Downstream of the Viul hydropower dam (Dw) FTSAs were only detectable in the pore water.

Outside the factory site (Ot), the concentration in sediments was 1000-2000 ng/g of each of the three quantifiable compounds (8:2 FTS, 10:2 FTS and 12:2 FTS), but in the pore water the concentrations differed according to carbon chain length with 3000 ng/L 8:2 FTS, 1000 ng/L 10:2 FTS and 200 ng/L 12:2 FTS (Figure 9, top and middle). The flux measurements (Figure 9, bottom) confirmed this difference by a 13x faster release of C8 FTS than C12 FTS to the overlying water.

In the Svarthølen basin (Ba), the internal distribution of the three FTSA-compounds was almost opposite with similar concentrations in the pore water (60-100 ng/L) and similar release rates (4-15 ng/m2/h), but lower concentration of 8:2 FTS (20 ng/g) than 10:2FTS (300 ng/g) and 12:2FTS (500 ng/g). It appeared that rapid loss of the short-chain compounds had reduced the concentrations in the solid phase to a level at which the three compounds were leached to the pore water and overlying water at similar rates.

Further downstream (Dw) the apparently same pattern of transformation had resulted in non-detectable concentration of the C8 FTS (<0.3 ng/g) in the solid sediment, and much reduced but still detectable concentrations of C-10 (1.5 ng/g) and C-12 FTS (3 ng/g). In the pore water, all three compounds were below detection limits, but the fluxes showed that all three compounds were still leached to the overlying water at about the same rates as at the stations closer to the source. This indicated that these compounds are released to the overlying water at rates which are not proportional to the sediment concentration and that relatively small residuals in the sediments may have an important impact on the quality of the overlying waterbody. The explanation to this somewhat unexpected result is most likely to be found in different sediment quality between the sampling stations.

3.2.2 Sulfonamido-ethanols (PFOSEs)

The sulfonamide ethanol N-EtFOSE was present in high concentrations in sediments from Svarthølen (100 ng/g at Ba) and outside the factory site (1000 ng/g at Ot). Correspondingly high concentrations were found in the pore water (20 and 130 ng/L), but a flux of this compound was detected from Svarthølen sediments only (Figure 8).

The concentrations of N-MeFOSE were low in the sediments (4 ng/g at Ba, 20 ng/g at Ot) and not detectable in pore water and fluxes.

PFOSEs were not detected in sediments, pore water or fluxes from any of the other locations (Dw, Con, Up).

Thus, 8:2FTS, 10:2FTS, 12:2FTS and N-EtFOSE were the most abundant PFAS-compounds near the closed down factory site with sediment concentrations at Ot ranging from 983 to 2374 ng/g.

3.2.3 Carboxylates (PFCAs)

PFOA was detected in the sediments from all sites reflecting the ubiquitous occurence of this compound. Only the sediments from outside the old factory site (Ot) showed significantly elevated concentrations compared to the control sediments from Lake Årungen (Con) and the upstream location (Up) (Figure 7). In the pore water from Lake Årungen, PFOA and three other PFCAs were determined at concentrations from 2 to 65 ng/L, compared to none detected at the upstream location (Up) and only one (PFUnDA) detected at the location downstream the hydropower dam (Dw). Thus, the presence of PFOA and other PFCAs at the two locations downstream the factory site (Ba and Dw), cannot for certain be assigned to discharges from the old factory site.

The highest fluxes of PFCAs were determined in aquaria with sediments collected outside the factory site and in Svarthølen basin (0,03-40 ng/m^2/h in Ot and <0,01-10 ng/m^2/h in Ba). At these two stations, the fluxes showed a similar decrease with increasing carbon chain lengths as was observed for the FTSAs. In the aquaria with sediments collected downstream the hydropower dam (Dw), fluxes were relatively uniform (0.5-4 ng/m^2/h) for all nine congeners determined in this group (Figure 7). This showed that even though hardly detectable in sediments and pore water, leakage of PFCAs to the overlying water may have an impact on the quality of the overlying water.

3.2.4 Sulfonates (PFSAs)

Of the sulfonates, only PFOS was detected in sediments collected near the old factory site (45 ng/g at Ot and 5 ng/g at Ba) (Figure 8). Further downstream at Dw, PFOS (and PFHxS) was detected in the sediments, but at very low concentrations (0.4 ng/g). In the pore water, PFOS was detected in all aquaria, decreasing from 80 ng/L close to the old factory site (Ot) to 1.5 ng/L downstream the hydropower dam (Dw). PFOS was also detected in pore water extracted from Lake Årungen sediments (3 ng/L in Con) and upstream the factory site (0.5 ng/L in Up). The fluxes of PFOS were ≤0.2 ng/m^2/L from sediments collected at the upstream location (Up) and Lake Årungen (Con), and 10x higher from sediments collected outside and downstream of the old factory site (Ot, Ba and Dw). Thus, PFOS leaked to the overlying water at similarly elevated rates from all sediments potentially affected by the source at the old factory site.

3.2.5 Sulfonamides (PFOSAs)

The sulfonamides PFOSA and N-EtFOSA were abundant (10-12 ng/L) in sediments collected outside the old factory site (Ot) and in Svarthølen basin (Ba), but below or near LOQ of 0,5 ng/L in sediments collected downstream the hydropower dam (Dw) and in control sediments from Lake Årungen (Con) and upstream the factory site (Up) (Figure 8). A similar pattern of distribution was found for pore water and fluxes. Thus, concentrations in pore water were 2-30 ng/L in Ot and Ba and below or near LOQ in Dw, Up and Con. Fluxes of PFOSA were 1-7 ng/m^2/h in Ot and Ba and ≤0.2 ng/m^2/h in the other aquaria. A small flux of 0.1 ng/m^2/h N-EtFOSA was detected in aquaria with sediments from Svarthølen basin (Ba) only.

3.2.6 Sulfonamido-acetates (PFOSAAs)

All three sulfonamide-acetates (FOSAA, meFOSAA and etFOSAA) were present in the sediments from outside the factory site (Ot) and in Svarthølen basin (Ba) (Figure 9). etFOSAA was the most abundant of these compounds in the sediments as well as in pore water and fluxes. Downstream the hydropower dam (Dw) these compounds were hardly detectable in sediments and pore water, but fluxes were measured of both me- and et-FOSAA. Control sediments collected upstream the factory site and in Lake Årungen provided no detectable levels of PFOSAAs neither in sediments, pore water or fluxes (Figure 9).

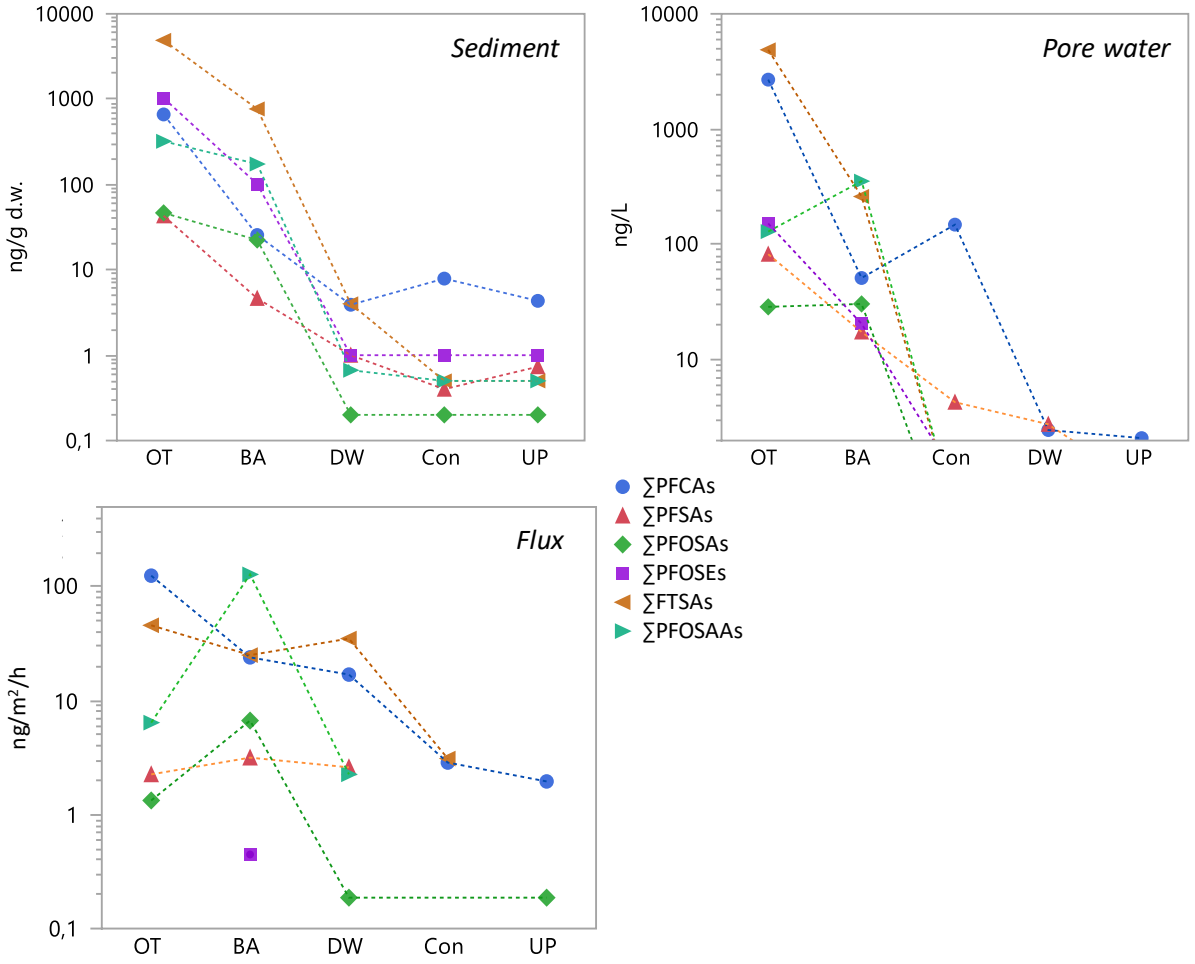


Figure 6. Group sums of perfluorinated compounds (PFAS) in sediment (top left), pore water (top right) and flux from sediment to overlying water (bottom) in aquaria with sediments collected at the factory site (Ot), Svarthølen basin (Ba), downstream the hydropower dam (Dw) and in control (Con) sediments collected upstream the factory site and in Lake Årungen. ∑PFCAs = sum of carboxylates, ∑PFSAs=sum sulfonates, ∑PFOSAs=sum sulfonamides, ∑PFOSEs = sum sulfonamido-ethanols, ∑FTSAs = sum fluorotelomer sulfonates, ∑PFOSAAs=sum sulfonamide-acetates. Contributing compounds within each group are given in Table 4 and figures below.

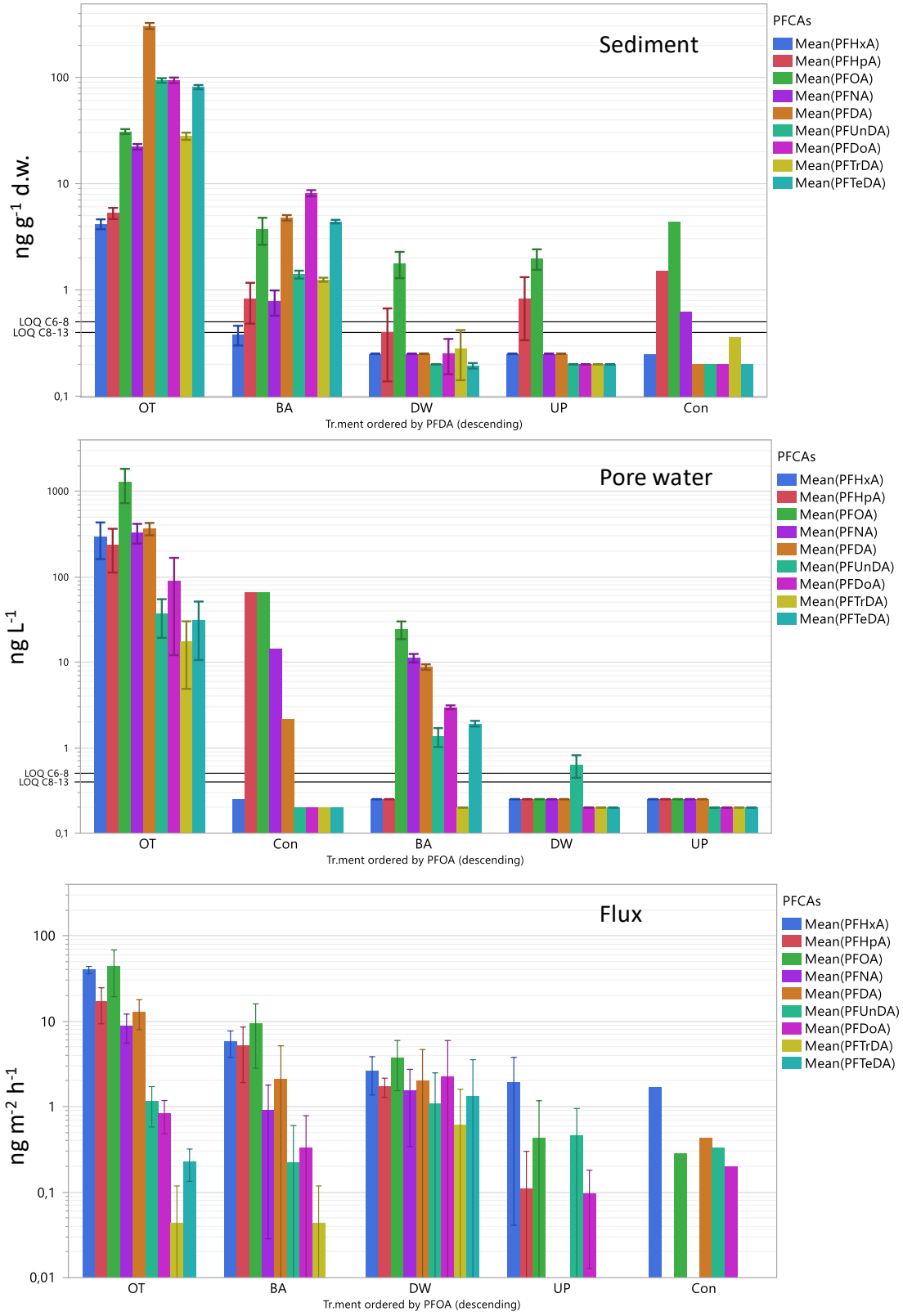


Figure 7. Carboxylates (PFCAs) in sediment, pore water and flux from sediment to water. NB: treatments (stations) ordered along x-axes by descending values of most abundant congener means that the order is not necessarily the same for sediment, pore water and flux. Error bars represent +/- one standard deviation.

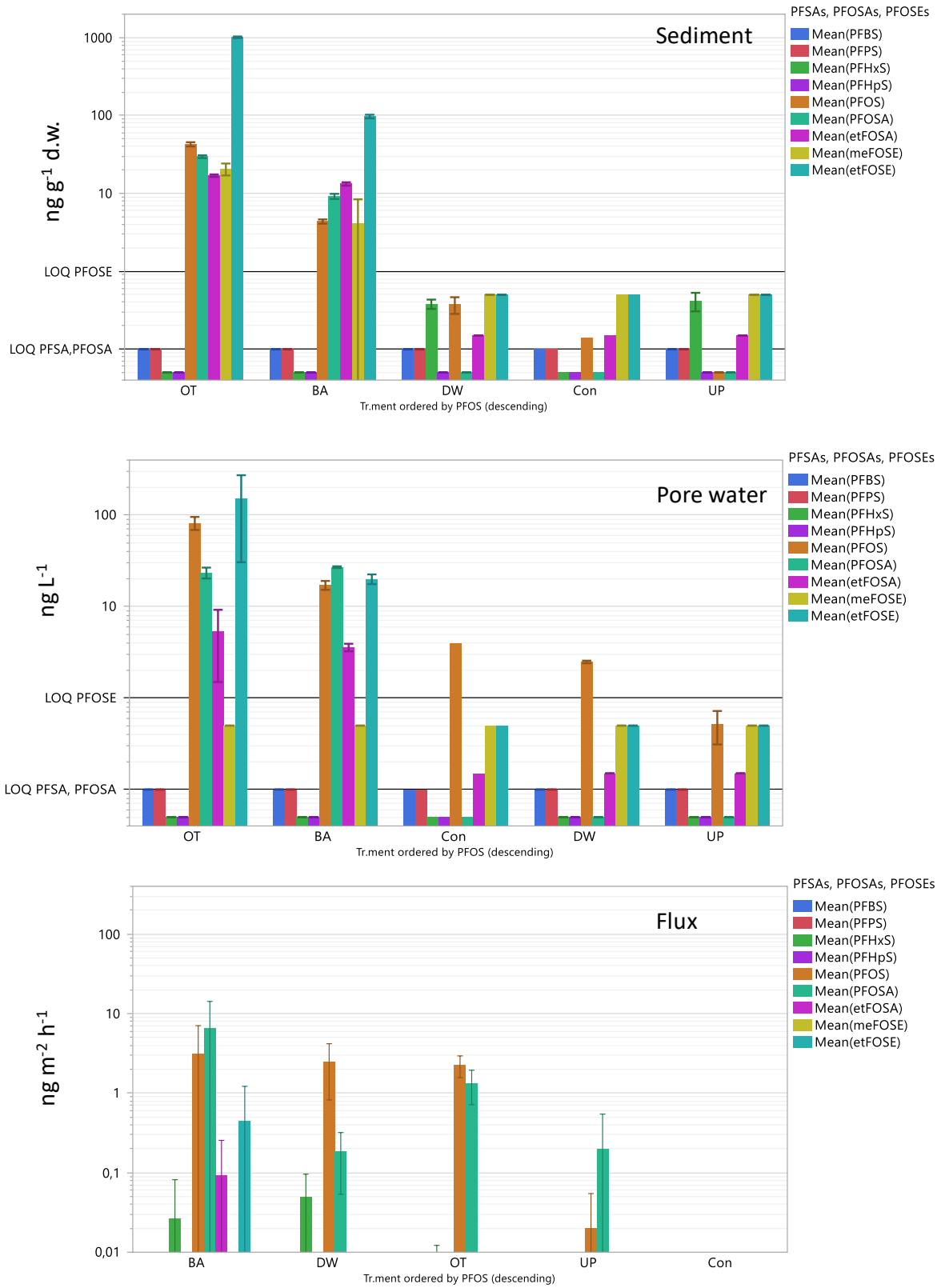


Figure 8. Sulfonates (PFSAs), sulfonamides (PFOSAs) and sulfonamide ethanols (PFOSEs) in sediment, water and flux from sediment to water. NB: treatments (=stations) ordered along x-axes by descending values of most abundant congener means that the order is not necessarily the same for sediment, pore water and flux. Error bars represent +/- one standard deviation.

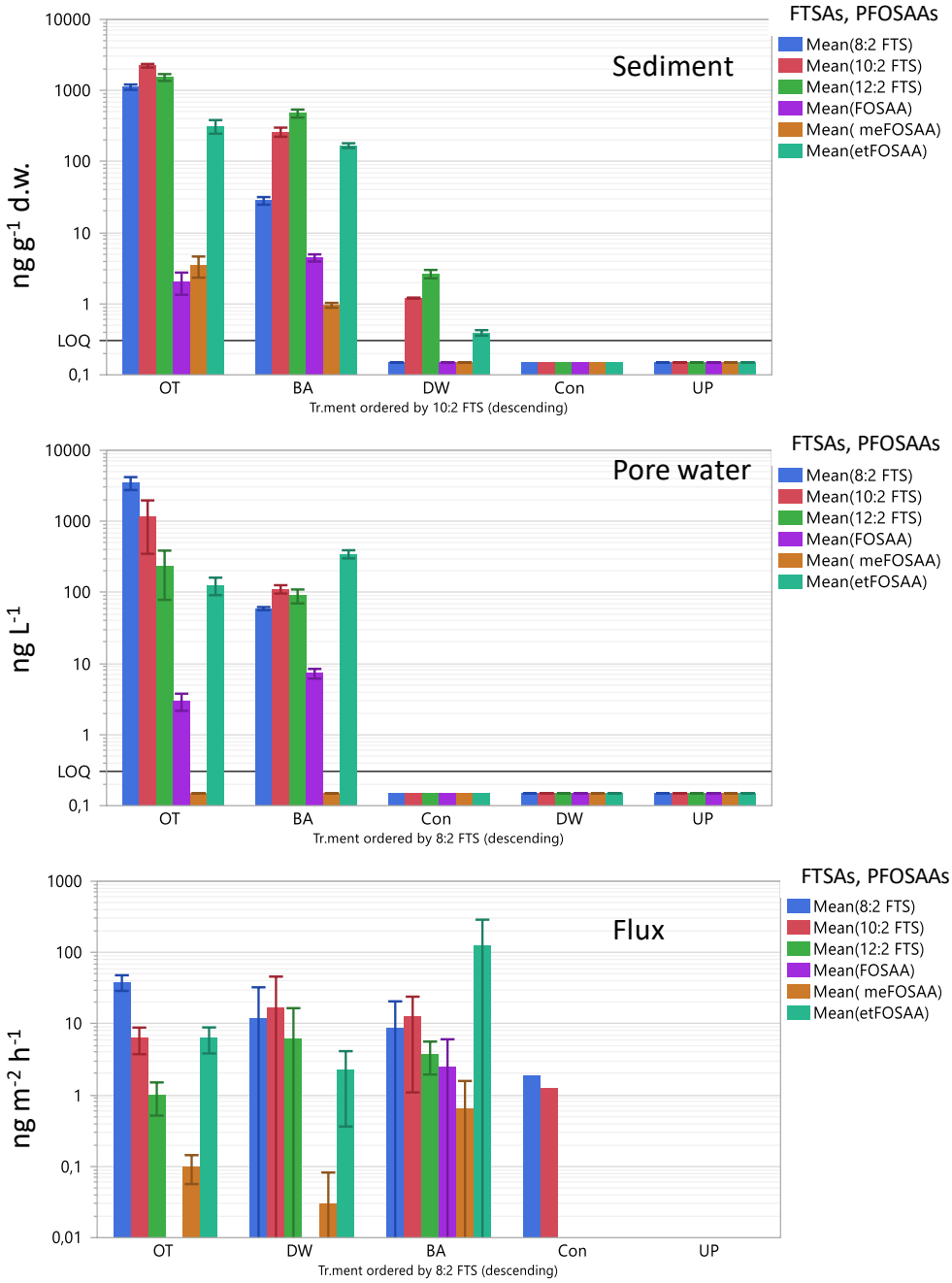


Figure 9. Fluorotelomer sulfonates (FTSAs) and sulfonamide acetates (PFOSAAs) in sediment, water and flux from sediment to water. NB: treatments (=stations) ordered along x-axes by descending values of most abundant congener means that the order is not necessarily the same for sediment, pore water and flux. Error bars represent +/- one standard deviation.

3.3 Biota

In general, the worms (*Tubifex tubifex*) had accumulated much more of the PFAS than the clams (*Anodonta anatine*) (Figure 10). The difference was about 2 orders of magnitude and most likely a result of the different biology of the two organisms. Thus, the shell protects the clam from direct exposure of soft tissues to the contaminants present in sediment and pore water. The worm, however, is in direct contact with sediment and pore water and with a very large body surface:volume ratio (long and thin). Also feeding by filtering overlying water via its' siphon protruding a few mm above the sediment surface, pose less exposure via food intake than the worm ingesting the whole sediment.

In sediments and pore water, the concentrations were generally higher in the sample collected outside the factory site (Ot) than in the sample collected in Svarthølen basin (Ba) (Figure 6), but in the organisms this relationship was opposite with the highest tissue concentrations most often occurring in the organisms exposed in sediments from Svarthølen basin (Ba) (Figure 10). This was probably a result of better environmental conditions in Ba, both with respect to lower contaminant levels of petrogenic hydrocarbons and PFAS and better food availability. The latter may be expected from increased microbiological and biological production based on biodegradation of pulp and paper waste accumulated in the Svarthølen basin sediments during the epoch of paper production at Viul.

3.3.1 Fluorotelomere sulfonates (FTSAs)

As in sediments and pore water, fluorotelomere sulfonates (FTSAs) were the most abundant PFAS in both organisms (Figure 10).

After 4 weeks exposure in sediments collected at the various stations, the worms (*Tubifex tubifex*) had acquired tissue concentrations of 3 238 ng/g ww (wet weight) in Svarthølen (Ba), 479 ng/g outside factory site (Ot) and 51 ng/g downstream hydropower dam (Dw). 10:2 and 12:2 FTS exceeded LOQ also after exposure in sediments from Lake Årungen (Con) and the upstream location (Up). In the zero sample not exposed to any sediments, a low concentration at about LOQ of 0.3 ng/g 12:2 FTS could be quantified (Figure 11).

In the sediments collected outside the factory site (Ot) the three FTSAs (8:2, 10:2 and 12:2 FTS) were the three most abundant PFAS-congeners and the 10:2 FTS was the one most abundant (Figure 9). Downstream the factory site (Ba and Dw) the sediment concentration of all three FTSAs decreased, but 8:2 and 10:2 FTS decreased more strongly than 12:2 FTS. Thus, the short chain C8-FTS appears to be lost more rapidly from the sediments than the longer chain C10- and C12-FTS. This was further elucidated by the C12:C10 ratio, which increased from 0.7 at Ot, to 1.8 at Ba and 2.2 at Dw. This downstream transition appeared upsized in the worms to an increase of the C12:C10 ratio of 0.1 at Ot to 1.0 at Ba and 4.6 at Dw. A similar downstream increase of the C12:C10 FTS ratio was found in all matrixes (Figure 13).

An interesting feature in Figure 13 was the similarity of the pattern of worm and sediment ratios, whereas the pattern in the clams were more like the pattern in fluxes. Apparently, this confirmed that the worms accumulate PFAS primarily from the sediments, whereas the clams accumulate PFAS primarily from the overlying water in which the concentration of PFAS will be proportional to the fluxes. This is true for the experimental set-up, but in natural systems the water concentrations will of course be influenced by a more complex set of factors.

3.3.2 Sulfonamido-ethanols (PFOSEs)

Next to the fluorotelomer sulfonates (FTSA), N-EtFOSE was present in the highest tissue concentrations both in worms (510 ng/g w.w.) and clams (11 ng/g w.w.) and higher in organisms exposed in sediments from Svarthølen (Ba) than those exposed in sediments from outside the factory site (Ot) (Figure 11, Figure 12). Low concentrations of N-EtFOSE was found in the worms exposed in sediments collected downstream the hydropower dam (Dw), but below LOQ in control sediments not affected by the discharges from the old factory (Up, Con and zero).

N-MeFOSE was below LOQ in both organisms from all aquaria.

3.3.3 Carboxylates (PFCAs)

Some carboxylate compounds were present in sediments and fluxes from all treatments including those not possibly affected by discharges from the old factory site (Con and Up) and the same was found in tissues of both organisms, even the non-exposed zero samples. In the worms these compounds were an order of magnitude higher in Ot and Ba than in the other aquaria, but in the mussels the highest concentrations of PFCAs were found for PFOA, PFHpA and PFHxA in worms exposed to sediments from Lake Årungen (Con) (Figure 10). These compounds were also detected in sediments, pore water and fluxes in aquaria with sediments from Lake Årungen. This confirmed that this control sediment was contaminated with short-chain C6-C10 carboxylic acids.

3.3.4 Sulfonates (PFSAs)

Sulfonates were below LOQ in the clams (Figure 12) from all aquaria. In the worms (Figure 11), PFOS were quantified in all samples ranging from the highest concentration of 43 ng/g w.w. in worms exposed in sediments from Svarthølen (Ba) to the lowest concentration of 1.1 ng/g w.w. in worms exposed to sediments collected upstream of the factory site (Up). Intermediate concentrations of PFOS was determined in non-exposed worms (zero) and in worms exposed in control sediments from Lake Årungen. This again demonstrates the ubiquitous occurrence of PFOS.

3.3.5 Sulfonamides (PFOSAs)

The concentrations of etFOSA was below LOQ in most of the biota-samples. In clams etFOSA was detected at 0.35-1.1 ng/g in two aquaria with sediments from Svarthølen (Ba). In worms 13 ng/g and 0.46 ng/g etFOSA were detected in aquaria with sediments from Svarthølen (Ba) and outside factory site (Ot), respectively.

PFOSA was present in worms exposed in sediments from Svarthølen (144 ng/g in Ba), outside the factory site (12.5 ng/g in Ot) and downstream the hydropower basin (2.4 ng/g in Dw) and in clams from the same sites (4.34 ng/g in Ba, 1.53 ng/g in Ot and 0.1 ng/g in Dw).

3.3.6 Sulfonamido-acetates (PFOSAAs)

MeFOSAA and FOSAA was not detected in any biota sample.

EtFOSAA was present in clams exposed in sediments collected in Svarthølen (12.6 ng/g w.w. at Ba) and outside factory site (1.4 ng/g at Ot) and in worms exposed in sediments from the same locations (147.8 ng/g at Ba and 11.6 ng/g at Ot). A low concentration of etFOSAA was also detected in worms exposed to sediments collected downstream the hydropower dam (0.4 ng/g at Dw).

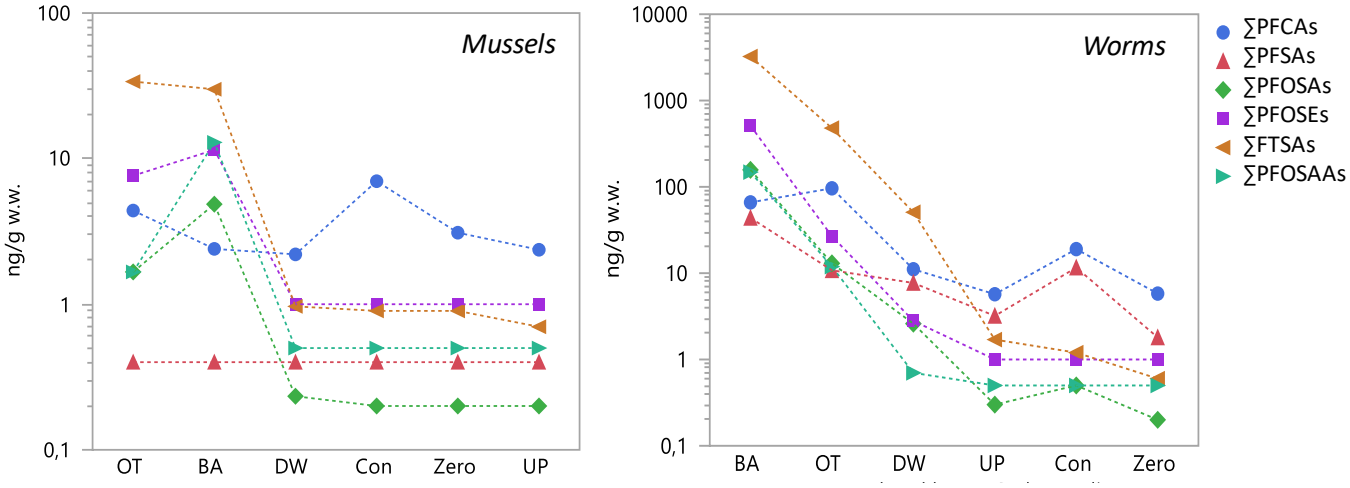


Figure 10. Group sums of perfluorinated compounds in mussels (*Anodonta anatine*) and worms (*Tubifex tubifex)* after 4 weeks exposure in aquaria with sediments collected at the factory site (Ot), Svarthølen basin (Ba), downstream the hydropower dam (Dw) and in control sediments collected upstream the factory site and in Lake Årungen. ∑PFCAs = sum of carboxylates, ∑PFSAs=sum sulfonates, ∑PFOSAs=sum sulfonamides, ∑PFOSEs = sum sulfonamido-ethanols, ∑FTSAs = sum fluorotelomer sulfonates, ∑PFOSAAs=sum sulfonamide-acetates. Contributing compounds within each group are given in Table 4 and figures below.

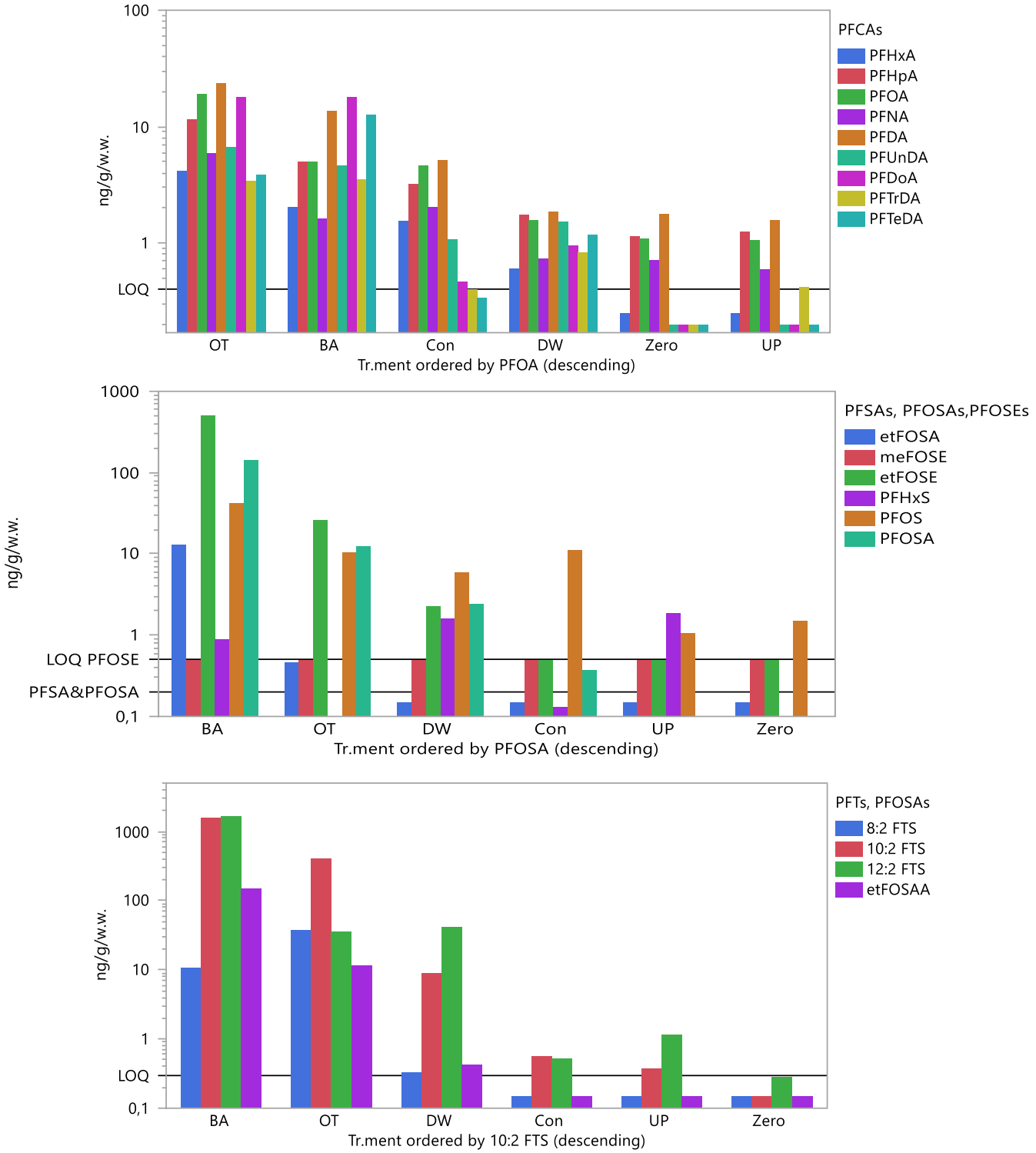


Figure 11. PFASs detected in worms (*Tubifex* tubifex) after 4 weeks exposure in aquaria with sediments collected at the factory site (Ot), Svarthølen basin (Ba), downstream the hydropower dam (Dw) and in control sediments collected upstream the factory site(Up) and in Lake Årungen (Con). Zero is non-exposed worms. Compounds below LOQ in all samples are not shown.

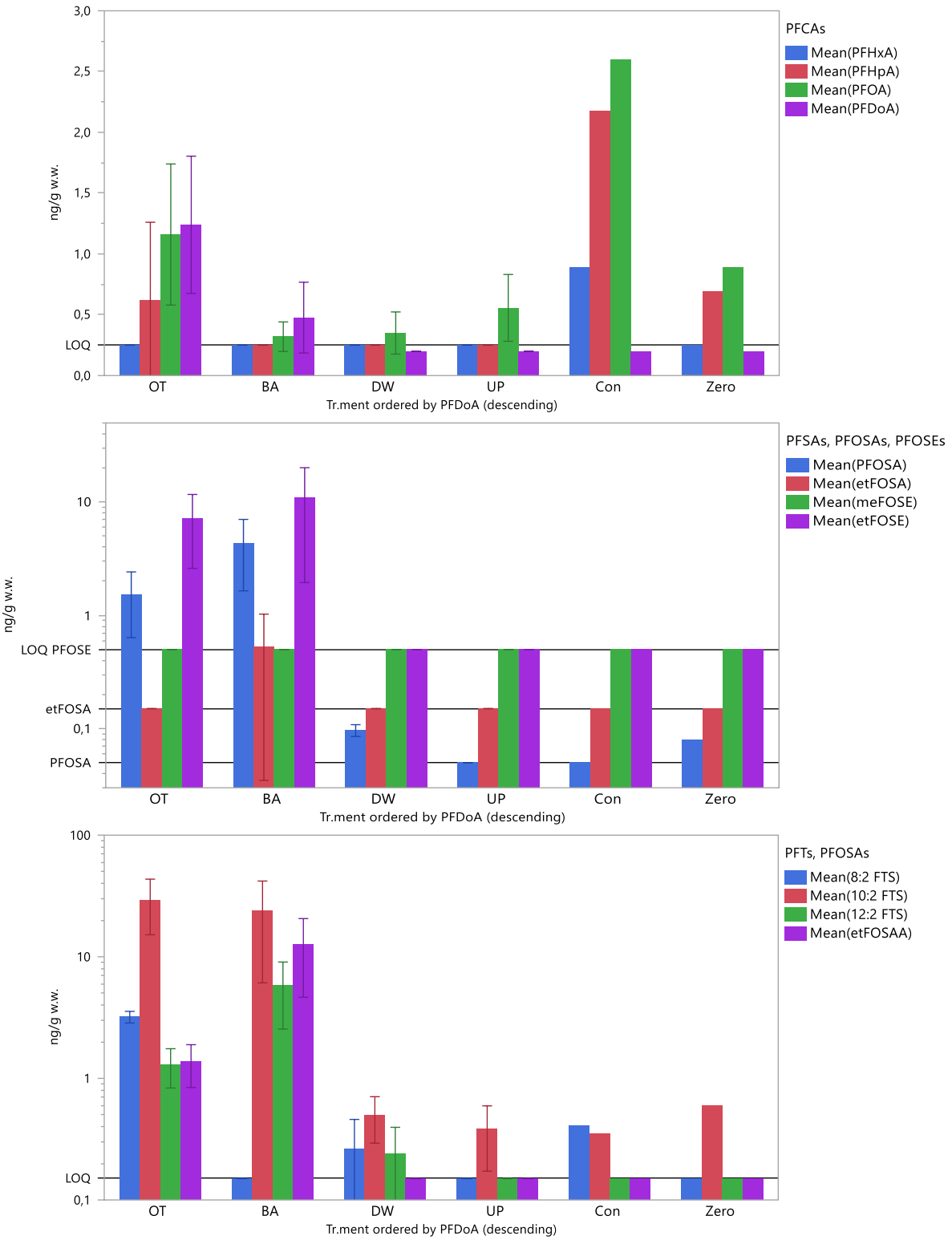


Figure 12. PFAS in clams (*Anodonta anatine*) after 4 weeks exposure in aquaria with sediments collected at the factory site (Ot), Svarthølen basin (Ba), downstream the hydropower dam (Dw) and in control sediments collected upstream the factory site (Up) and in Lake Årungen (Con). Zero is a

non-exposed clam sampled on day zero. Compounds below LOQ in all samples are not shown. Error bars represent +/- one standard deviation.

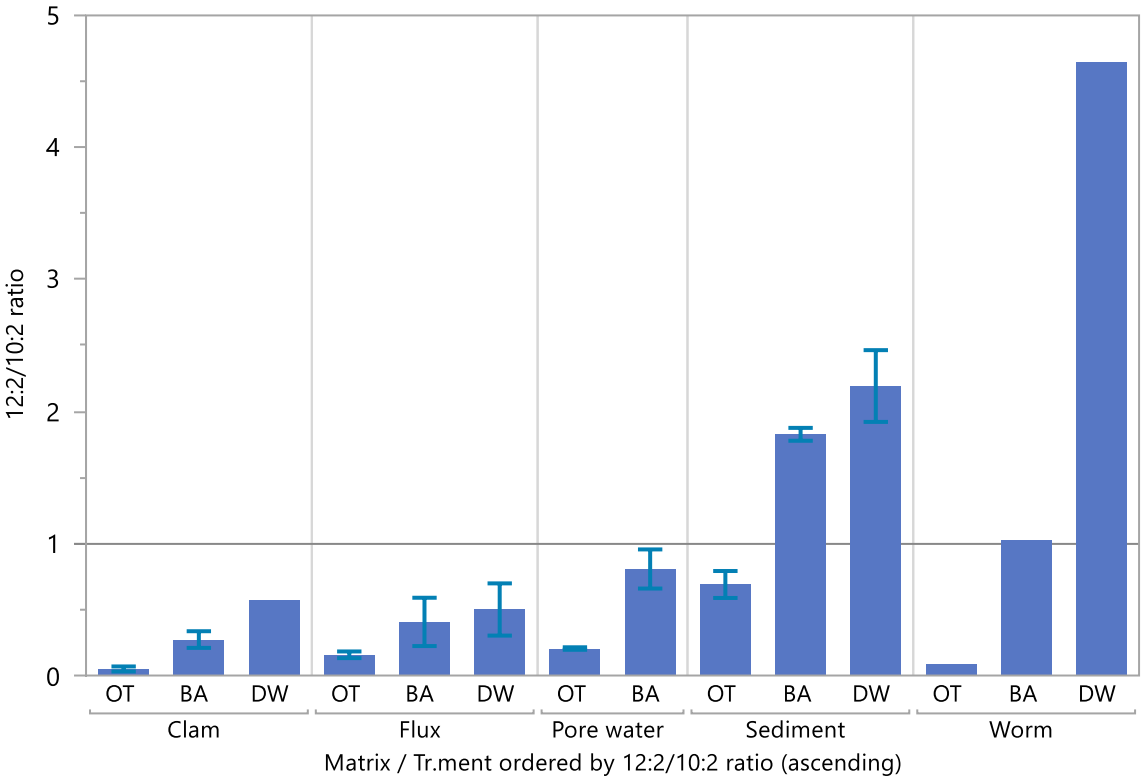


Figure 13. Ratio between 12:2 FTS and 10:2 FTS in the various samples from aquaria with sediments collected at increasing distance from the assumed source, i.e. the old factory site (Ot), Svarthølen (Ba) and downstream the hydropower dam (Dw).

4 Discussion

4.1 Sediment-pore water partition coefficients

Partition coefficients (K_d) were calculated by dividing the concentrations determined in sediments (ng/kg) with the concentration determined in the pore water (ng/L). A low K_d indicates that the compound is loosely associated with the sediment and will tend to yield higher concentrations in pore water and higher fluxes from sediment to water. The coefficients were calculated for all components present at concentrations higher than LOQ in both matrixes. All calculated K_d-values are shown in Figure 14 and Figure 15.

The logK_d ranged 1.2-2.3 for PFOA and 2.0-2.9 for PFOS. This compared reasonably well with the range of 1.3-4.5 for PFOA and 2.4-4.4 for PFOS determined in sorption experiments reviewed by Zareitalabad et al. (2013). The Kd's calculated for Dw and Con in the samples which provided detectable concentrations in both media, were within same order of magnitude as those determined at Ot and Ba (Figure 14). This was consistent with the universal nature of the partition coefficients.

Figure 15 shows a positive correlation between K_d and molecular weight with a correlation coefficient R^2 of 0.6. Thus, the larger PFAS molecules tend to be more firmly associated with the sediments than the smaller PFAS molecules. Different K_ds for the same compound may result from different sediment quality at one station compared to another. Thus, sediments with high abundancies of organic carbon is generally known to bind polar substances more strongly than sediments in which organic carbon is less abundant.

For PFOS the logK_ds decreased from 2.6 at Ot (outside the old factory site), via 2.4 in the Svarthølen basin (Ba) to 2.2 at Dw (downstream the dam). Thus, the solubility of this compound appeared to increase with increasing distance from the source. This is reasonably explained by high concentrations of petrogenic carbon outside the old factory site (Ot) and abundant remnants of pulp and paper in Svarthølen.

PFOS and five other compounds (etFOSAA, PFDA, PFOSA, PFUnDA) had significantly higher K_ds at Ot than Ba. Only two compounds (PFOA and PFTrDA) had higher K_ds at Ba than Ot. Thus, it appears that the petrogenic hydrocarbons present in the sediments at Ot may retain PFAS equally or more efficient than the pulp and paper remnants at Ba.

No K_ds could be determined in aquaria with sediments from the upstream location (Up), and only three K_ds (PFHpA, PFOA and PFNA) could be determined in aquaria with sediments from Lake Årungen. Both PFOA and PFNA were more strongly bound to the sediments from the Svarthølen basin (Ba) than to the sediments from Lake Årungen (Figure 14).

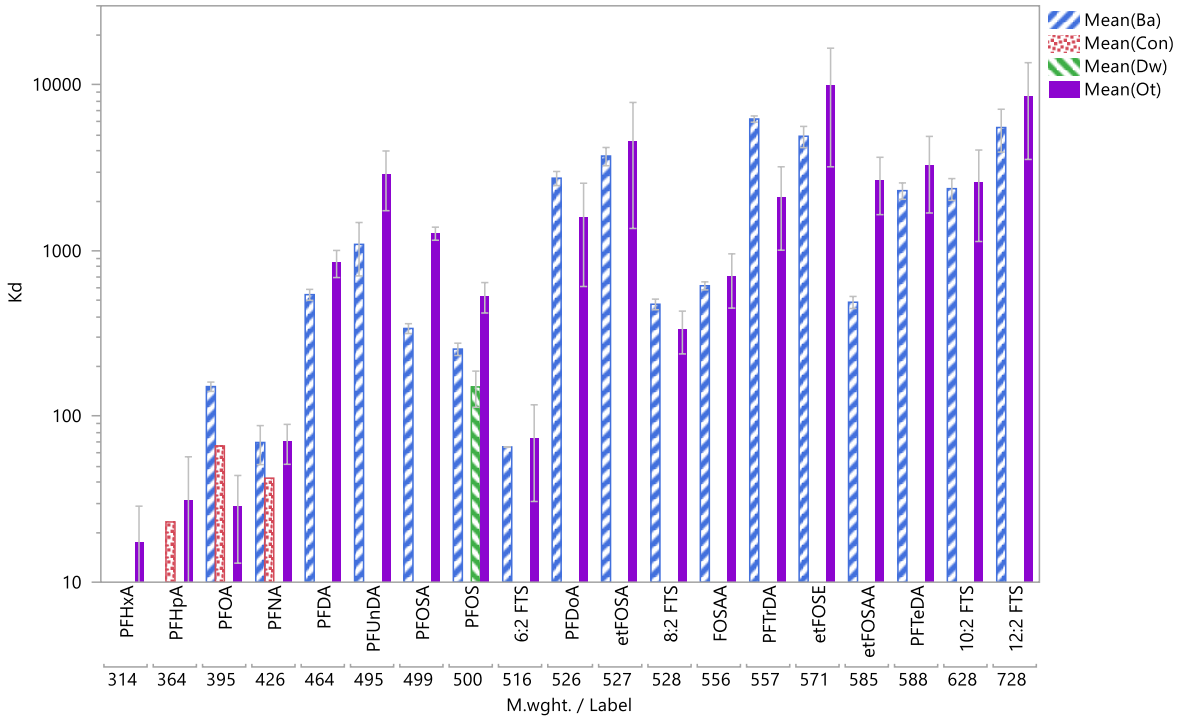


Figure 14. Sediment pore water partition coefficients (Kd) calculated for all aquaria in which both sediment and porewater concentrations were above LOQ. Vertical bars represent ± one standard deviation (N=3).

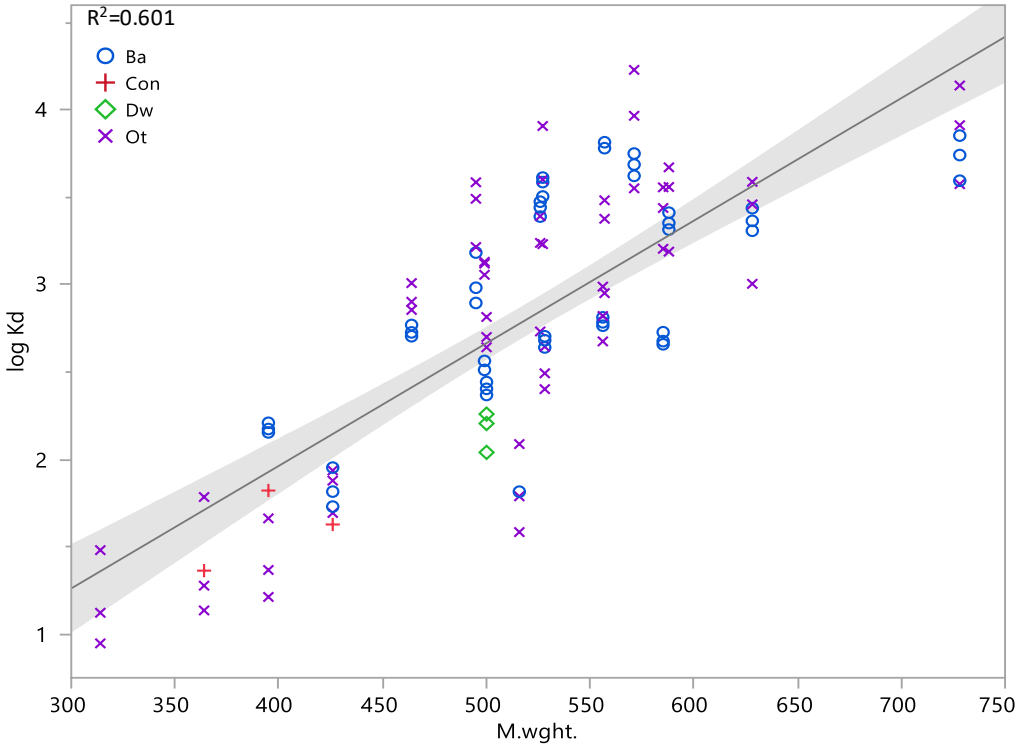


Figure 15. Sediment pore water partition coefficients (**log**K_d) for PFAS as function of molecular weight. basin (Ba). A similar downstream change in solubility may apply to other PFAS compounds not detectable at Dw.

4.2 Bioaccumulation

Biota Sediment Accumulation Factors (BSAFs) were calculated as the ratio between the concentration of PFAS in worms (ng/g w.w.) and the concentration in sediment (ng/g d.w.). BSAF is a simple way to present the tendency of a compound present in the sediment to accumulate in an organism. Similarly, Biota Concentration Factors (BCFs) were calculated as the ratio between concentration in clams and aquarium water. Because the clams will accumulate most of the contaminants from the aquarium water through the siphon, BCF is more relevant than BSAF for this organism. While BSAF has no unit (g/g), BCF has the unit L/kg.

Bioaccumulation factors were calculated for all aquaria with sediments from the three sites in Randselva outside the closed down factory site (Ot), and downstream at Ba and Dw. If one of the two concentrations were below LOQ, the bioaccumulation factor was not calculated. The BCFs calculated for clams varied between 1.4 and 4.2 and were positively correlated ($p=0.0003$, $R^2 = 0.26$) with the molecular weight (Figure 16, lower diagram). Labadie and Chevreuil (2010) determined BCF for carboxylates and sulfonates in various tissues of European chub (*L. cephalus*). For liver and muscle, they found logBCFs between 1 and 6 and a clear increase with increasing carbon chain length.

At Ba and Dw, the BSAFs calculated for the worms (Figure 16, upper diagram), did not provide a positive correlation ($p=0.32$, $R^2=0.04$). The absolute values and lack of trends observed at Ba and Dw were, however, consistent with the BSAFs determined in European chub in Orge river, in which logBSAF for PFCAs and PFSAs ranged from -1.3 to +1.5 and no increase with increasing carbon chain length (Labadie and Chevreuil, 2011).

At Ot the BSAFs provided a significant negative correlation ($p=0.0003$, $R^2 = 0.59$). Further analyses of the BSAFs was done by comparing the mean BSAFs from all aquaria using the Tukey Kramer HSD (honestly significant difference) test, which is a conservative test for data with different sample size. This test (Figure 17) showed that the BSAFs in Ot were significantly lower than the BSAFs in the other aquaria.

The sediments from Ot were visibly and olfactorily different from the other sediments by an abundancy of small strips of plastic films and a smell of petrogenic substances (like diesel). This is due to oilspills at the factory site in the 1970s (Rambøll 2018). Indicative analyses of the plastic strips revealed high concentrations of PFAS in this matrix. One possible explanation to the anomalous BSAFs at Ot might therefore be that the worms avoided, or was prevented from, ingestion of the plastic strips. Also, as indicated by the low recovery (Table 3), appetite and the general food ingestion may have been reduced due to the nuisance of the petrogenic hydrocarbons. In addition to reduced uptake via ingested food items, contaminants may accumulate in the organisms by diffusion of dissolved or loosely bound contaminants across body membranes. This pathway may have been less significant for the high molecular weight PFAS compounds with low bioavailability due to association with petrogenic hydrocarbons.

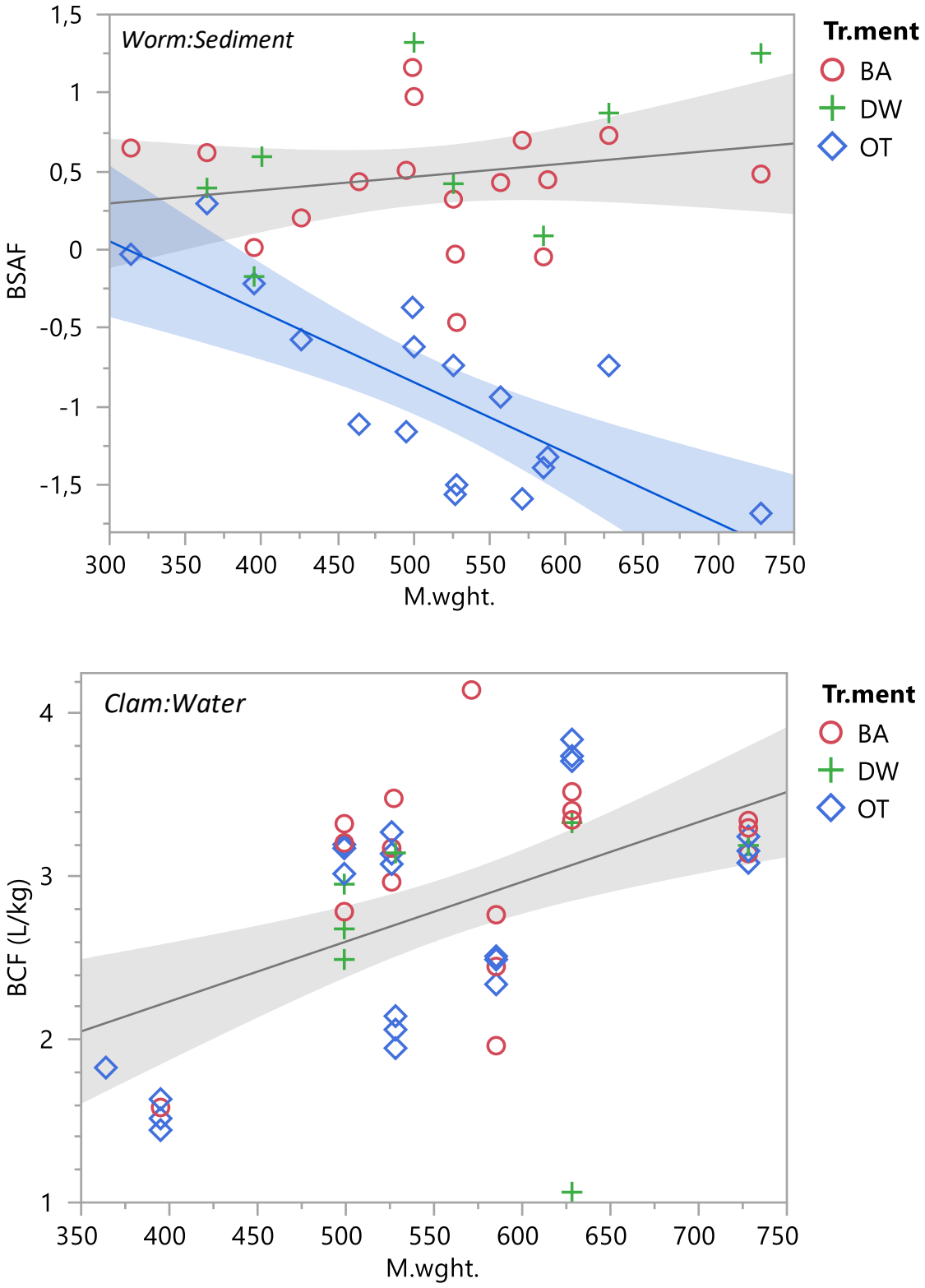
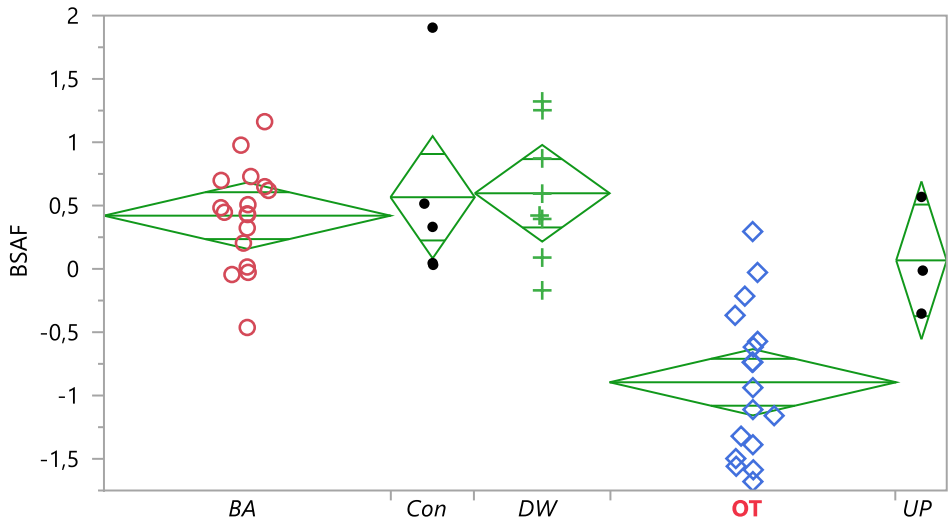


Figure 16. Bioaccumulation factors BSAF for the worm:sediment ratio (top) and BCF for the clam:water ratio (bottom). Linear regression lines and confidence intervals were calculated from all clam:water-ratios, but for the worm:sediment-ratio regression analyses were done separately for the Ot-aquaria (blue) and the BA- and DW-aquaria (grey) (see text).



Level		Mean
DW	A	0,597
Con	A	0,566
BA	A	0,420
UP	A	0,067
OT	B	-0,895

Figure 17. Comparison of all BSAFs calculated for the worms in all aquaria (Tukey Kramer HSD test). The table gives the mean BSAF in each treatment/station and the letter B shows that the BSAFs in Ot were significantly lower than the BSAF in all other treatments (letter A). The diamonds show mean values and 95% confidence interval. The width of the diamond is proportional to sample size.

4.3 Regulatory classification, PFOS and PFOA

Norwegian guidelines (Veileder 2:2018) provide Environmental Quality Standards (EQS) for two PFAS compounds, PFOS and PFOA (Table 1). Because these are derived from exclusively man-made substances, class I "background" does not exist. Upper boundaries for class III are high compared to all concentrations determined in the present study. Hence our observations comply with class II "no toxic effects" or class III "chronic effects from longterm exposure". As shown in Figure 18, PFOS exceeded the threshold value of 2.3 ng/g d.w. in sediments collected outside and in the basin downstream the old old factory site (Ot and Ba). Water samples for flux measurements were collected three times from each aquarium and several of the samples collected from the same aquaria and the aquaria with sediments collected further downstream (Dw) had acquired concentrations exceeding the threshold value of 0.65 ng/L indicating risk of "toxic effects from longterm exposure". The extent to which the overlying water at the natural sites will be contaminated with PFOS, will depend on the residence time of the water and will most likely be confined to areas with relatively stagnant water.

The pore water may be considered as a highly stagnant water, and all samples from Randselva downstream the factory site as well as the pore water extracted from Lake Årungen sediments (Con) were contaminated to class III "toxic effects from long-term exposure" (Figure 18). The classification of water and porewater provided here is indicative for the level of contamination. Valid classification must be done based on concentrations measured in samples collected in Randselva and Lake Årungen, not in the experimental set-up in which the rates of water renewal is different.

The clams did not accumulate PFOS or PFOA to concentrations exceeding environmental quality standards, but the oligochaetes accumulated PFOS to concentrations exceeding the environmental quality standard as defined for marine and freshwater fish, but it is unclear if this can be applied for oligochaetes.

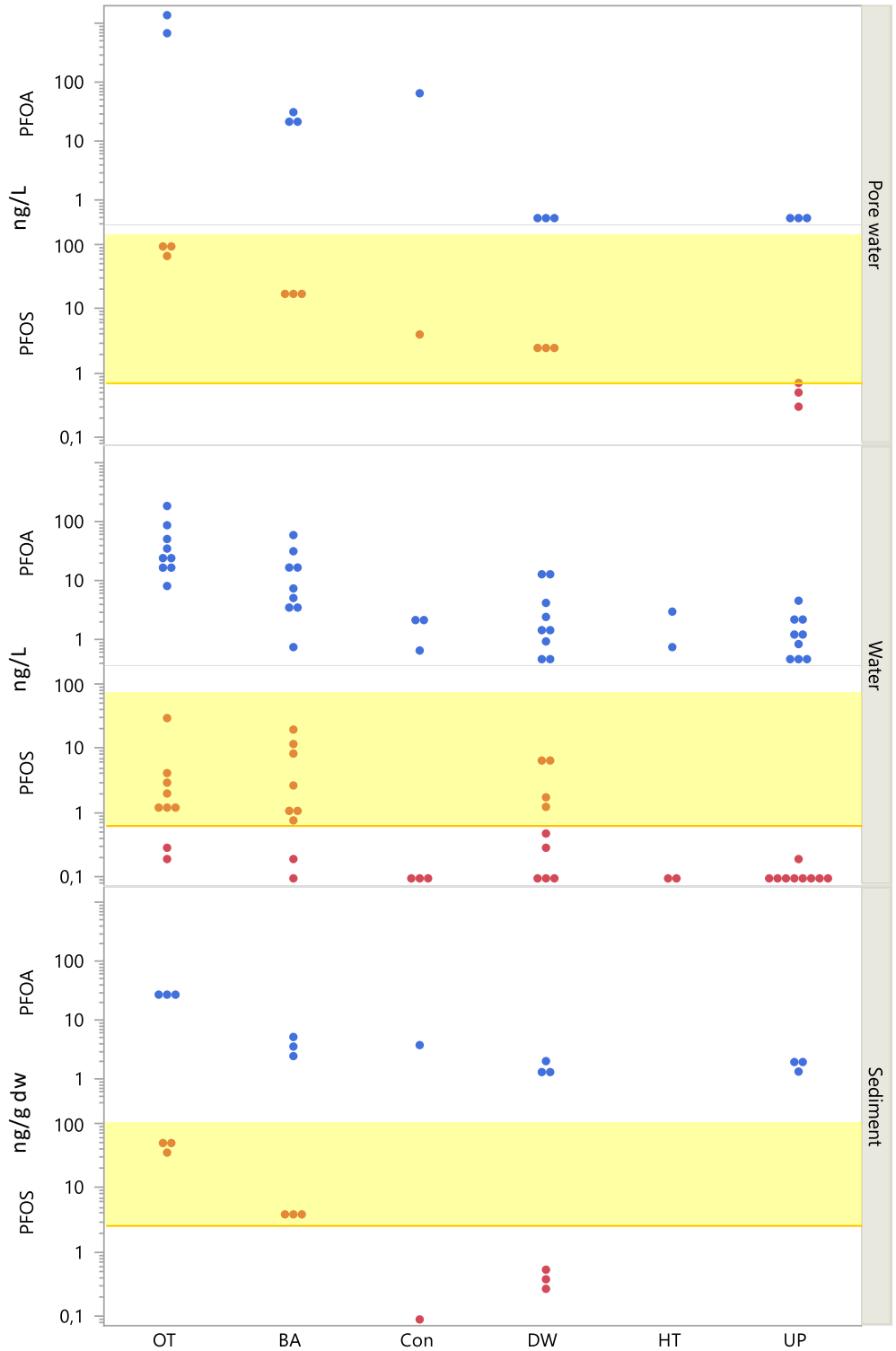


Figure 18. Concentrations of PFOS (red dots) and PFOA (blue dots) in sediment, pore water and overlying water compared to EQS-values given in Norwegian guidelines (Veileder 02:2018). HT is the source water used for continuous renewal of the water in the aquaria. Data within yellow rectangles are classified as class III and may provide "chronic effects from longterm exposure". Class I "background" does not exist for these compounds and no data exceeded the upper boundary for class III. Thus, all data outside yellow rectangles are classified as class II "no toxic effects". Porewater and overlying water are classified using EQS-values defined for freshwater.

4.4 PFOS and PFOS precursors

4.4.1 Degradation of SAmPAP esters

N-EtFOSE is a biodegradable compound and a precursor to PFOS which is considered a toxic substance, persistent in the environment, and accumulates and magnifies in living organisms. Hence, it is on the EU-list of priority hazardous substances (EU-directive 2013).

N-EtFOSE is derived from hydrolytic cleavage of the SAmPAP diester which was produced in large volumes for use in food contact paper and packaging in North America between 1974 and 2002 (Benskin et al., 2012). SAmPAP ester was analyzed in the sediment samples, but not quantified due to the lack of standard solution. Still, the SAmPAP peaks were identified clear and strong in the chromatograms and twice as large in sediments from Ot as in sediments from Ba. Even at Dw small peaks of about 1/1000 of the size as Ba and Ot were clearly recognizable, whereas no traces were found in sediments upstream the old factory site (Up). If SAmPAP has been discharged from the factory at Viul, N-EtFOSE and its early degradation products should be expected to be abundant in the sediment outside the factory site (Ot) and in the downstream basin (Ba).

The aerobic pathway of degradation from N-EtFOSE to PFOS (Benskin et al., 2012) involves several intermediate compounds analyzed in this study: N-EtFOSE → N-EtFOSAA → N-EtFOSA → FOSAA → PFOSA → PFOS. The whole group is here referred to as FOSAMs (N-alkyl-substituted perfluorooctane sulfonamides) and are all based on the PFOS C8 molecule ($C_8F_{17}SO_3^-$) with various amino-groups attached.

4.4.2 Occurrence of PFOS precursors

The FOSAMs were highly abundant in all matrixes in aquaria with sediments from Ot and Ba (Figure 19). At Dw the FOSAMs were much less abundant, but in most matrixes more abundant than in samples not impacted by discharges from the old factory (Up, Con and zero). As shown in (Figure 20) the products near the end of the degradation chain (PFOS and PFOSA) were the dominant compounds at these stations. This indicated that the sediments from Ot and Ba were closer to the source with predominant presence of the early degradation products (EtFOSE and EtFOSAA). These two compounds were still present (but less dominant) at Dw, and below LOQ in all matrixes not possibly affected by discharges from the old factory site (i.e. Con, Up, zero). In these samples, PFOSA and PFOS were the only compounds left of the degradation series (Figure 8, Figure 11). This is as expected for samples collected remote from any direct source of SAmPAP esters.

Because of the dam, the water flows slowly through the Ba site which therefore acts as a sedimentation basin. Probably, contaminants have been deposited in relatively thick layers under partially anoxic conditions which will contribute to reduce rates of degradation. This is very different from the situation at Dw located further down the river in a presumably much more dynamic environment with more favorable conditions for aerobic degradation. In aquaria with sediments from this location, N-EtFOSE was present above LOQ in the worms (Figure 11) , but not in any other matrix (i.e. sediments, pore water, flux and clams). Besides indicating a significant potential for bioaccumulation of this compound in sediment dwelling species, the observations of N-etFOSE in worms and N-EtFOSAA in several matrixes (sediment, fluxes and worms) in the Dw-aquaria (Figure 20), showed that residual FOSAMs from the closed down factory site were present in the sediments collected at all three stations in Randselva outside and downstream from the old factory site.

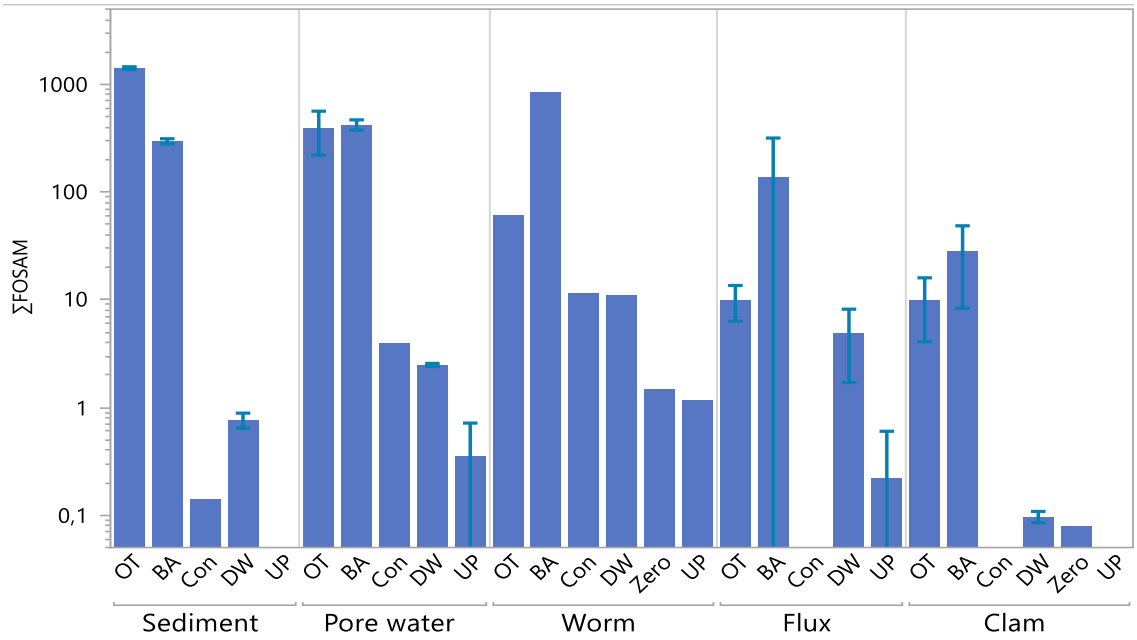


Figure 19. Sum of PFOS and PFOS precursors in the various matrixes determined in each aquarium. Note different units on y-axis: ng/g for sediment, ng/L for pore water, ng/g w.w. for biota and ng/m²/h for flux. Error bars show +/- one standard deviation.

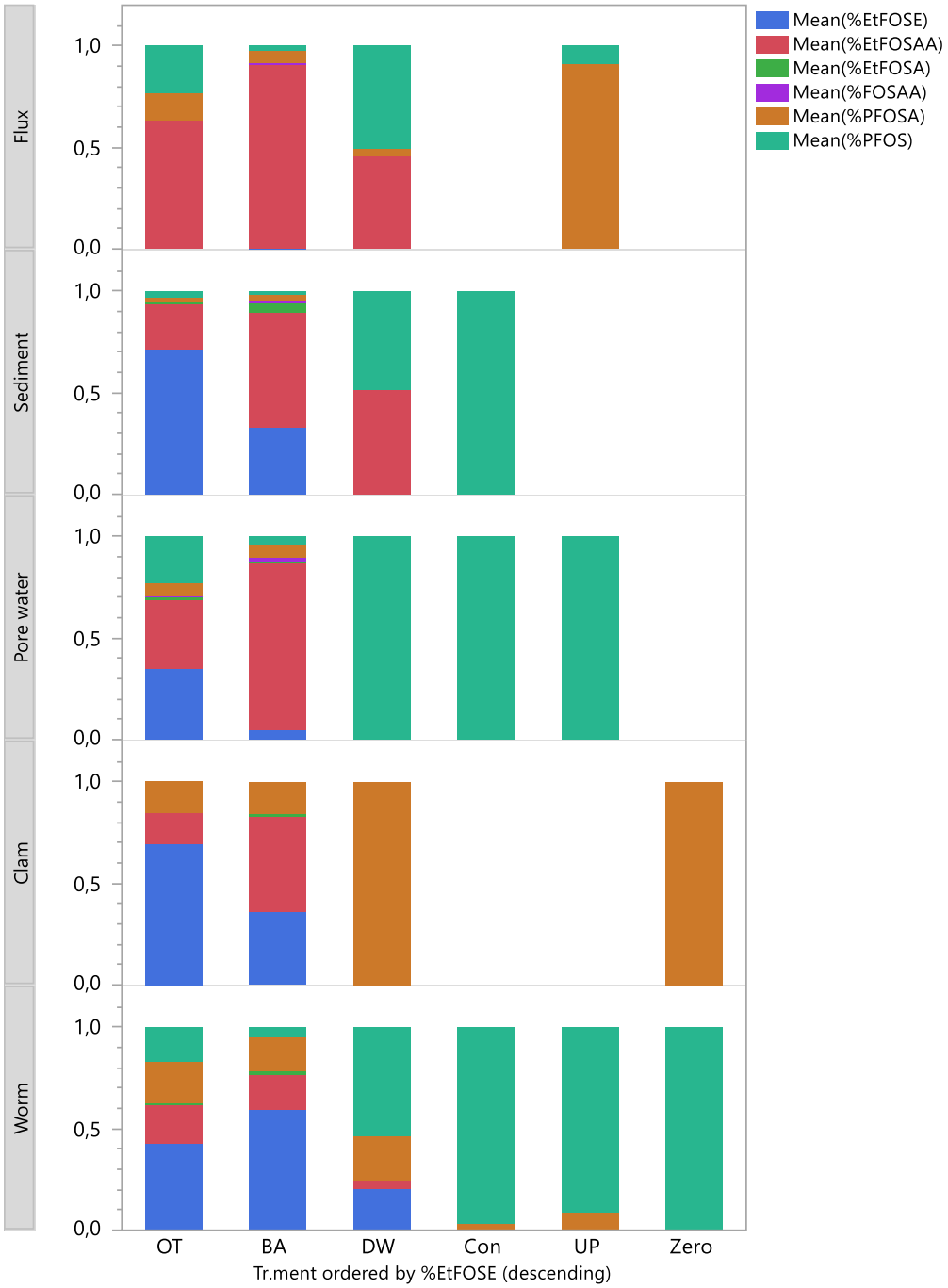


Figure 20. Relative composition of PFOS and PFOS precursors at each station and matrix. Concentrations ≤LOQ set to zero.

4.5 FTSAs and PFCAs

In sediments influence by the old factory site, the six C8 FOSAM compounds accounted for a major fraction of the 17 sulphonate-, sulfonamide-, sulfonamidoethanol- and sulfonamidoacetate compounds analyzed, i.e. 98% at Ot and Ba and 67% at Dw. Compared to the total concentration of all 31 PFAS congeneres determined, the FOSAMs accounted for not more than 10-27% of the

sediment concentration. The remaining compounds were mostly fluorotelomers (FTSAs) (50-70%) and carboxylates (PFCAs) (2-35%) and some minor contributions from PFHxS in sediment and worms from Dw and Up (Figure 21).

In aquaria with sediments from Ba, the bulk of PFAS compounds was accounted for by FOSAMs and fluorotelomers (FTSAs) in all matrixes. PFCAs were primarily observed in fluxes (21%) and pore water (7%) from these aquaria. In aquaria with sediments from Ot and Dw, FOSAMs and fluorotelomers (FTSAs) dominated the pool of PFASs in sediments and biota, whereas the relative contribution from PFCAs were larger in pore water and fluxes. This tendency towards higher contribution of the carboxylates (PFCAs) in pore water and fluxes was consistent with the K_ds which were generally higher for FTSAs and FOSAMs than for PFCAs (Figure 14, Figure 15). Furthermore, the five PFCAs with the lowest molecular weights accounted for most of the total abundance of PFCAs in pore water and fluxes (Figure 7).

The shift from 8:2 towards 10:2 and 12:2 FTS at successively larger distance from the old factory site (Ot) both in sediment, pore water, clams and worms (Figure 9, Figure 11 and Figure 12) was also consistent with the lower K_d of the shorter C8 compound. The difference in K_ds was further substantiated by the fluxes of 8:2 FTS which were relatively similar (10-40 ng/m2/h) at the three stations despite sediment concentrations ranging from more than 1000 ng/g at Ot to below LOQ in Dw (Figure 9). Thus, 8:2 FTS appeared to be lost from the sediments relatively independent of concentrations in sediment and pore water. This must be related to differences in sediment quality with the higher sorption capacities in the sediments rich in organic carbon from pulp and paper at Ba and hydrocarbons at Ot. Possibly also the plastic fibers present in the sediments at Ot may have contributed to stronger binding of 8:2 FTS.

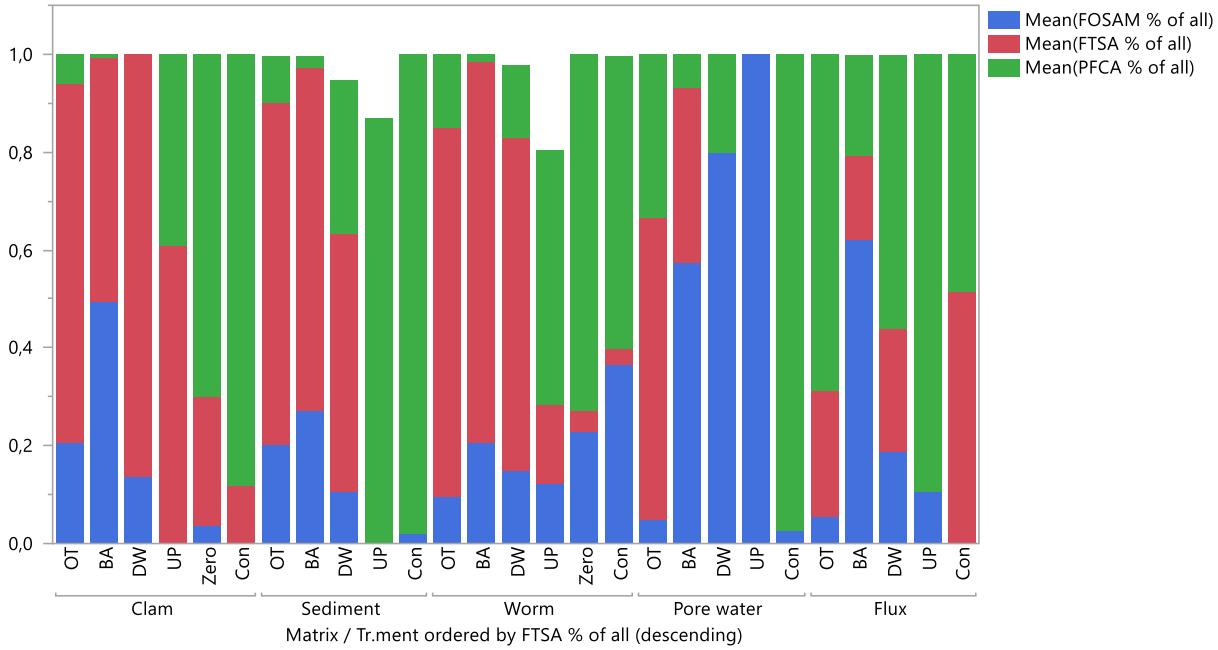


Figure 21. Relative contribution of carboxylates (PFCAs), fluorotelomers (FTSAs) and PFOS precursors (FOSAMs) to all 31 compounds determined in **all** matrixes and samples. Deviations from 1.0 is mostly accounted for by the sulfonic acid PFHxS.

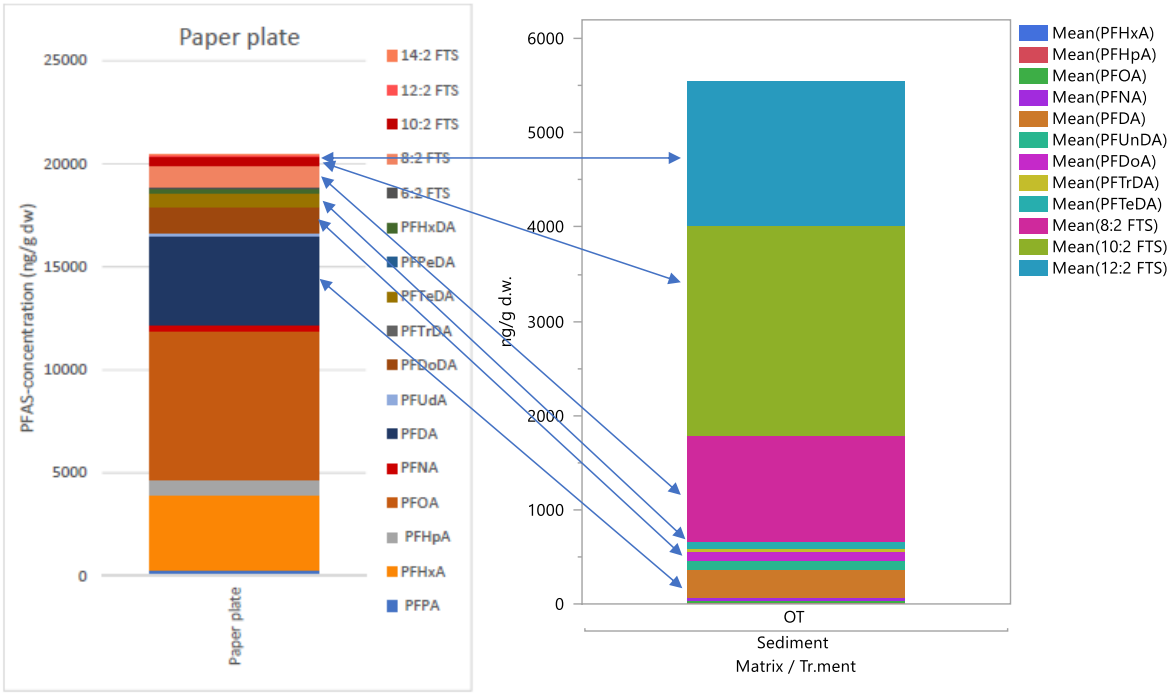


Figure 22. Comparison of PFCAs and FTSAs found in paper plate, claimed to have been produced at the factory in 2007, and in sediments at Ot in 2018. Arrows connect bands representing same compound. From bottom to top these are: PFDA, PFDoDA, PFTeDA, 8:2FTS, 10:2 FTS and 12:2 FTS. Paper plate diagram copied from Grønning et al., 2019.

Based on the literature, the fluorotelomer sulfonates (FTSAs) appears to have come into use as a substitute for SAmPAPs after the ban in 2002 (Wang et al., 2010, Benskin et al., 2013). A dated sediment core from Tyrifjorden has shown a shift from FOSAMs to FTSAs in the material deposited between 2000 and 2005 (Grønning et al., 2019). This suitably explains the abundance of FTSAs at the Viul site, but not necessarily the PFCAs.

It has been proposed (Wang et al., 2005, 2011) that the FTSAs with X fluorinated carbon atoms (X = 4, 6, 8…14) and 2 non-fluorinated may degrade by splitting off the sulfonate and one or both of the adjacent carbon atoms leaving an (X+1) or X carbon acetic acid. Thus, the 6:2 FTS may be a precursor heptanoic and hexanoic acids (PFHpA, PFHxA), the 8:2 FTS may be precursor to the nonanoic and octanoic acid (PFNA and PFOA) and so on (Wang et al., 2005, 2011). However, the PFCAs may have other sources as well and analyses of a paper plate claimed to have been produced at the factory at Viul in 2007 (Grønning et al., 2019) showed that the PFAS was exclusively composed of 10-12 different PFCAs and 3-5 FTSAs including significant amounts of the C6, C8, C10 and C12 carbonates and the 8:2, 10:2 and 12:2 FTS. These same components were found in the sediments outside the factory site in 2018, but the composition had shifted from predominantly short chain PFCAs in the paper plate to predominantly long chain FTSAs in the sediments (Figure 22). The generally increasing dominance of the long chain FTSAs from Ot to Ba and Dw both in sediment and biota (Figure 23) shows that this shift can be recognized both in time (increasing time since discharge) and space (increasing distance from discharge point). A similar shift from short to longer chain PFCAs was not found (Figure 24).

FTSAs are still present in the sediments and leaking to the overlying water at all three stations outside and downstream the old factory (Figure 6, Figure 9). Also, the PFCAs were found in the sediments and leakage to the overlying water was still significant for all nine compounds at the three downstream stations.

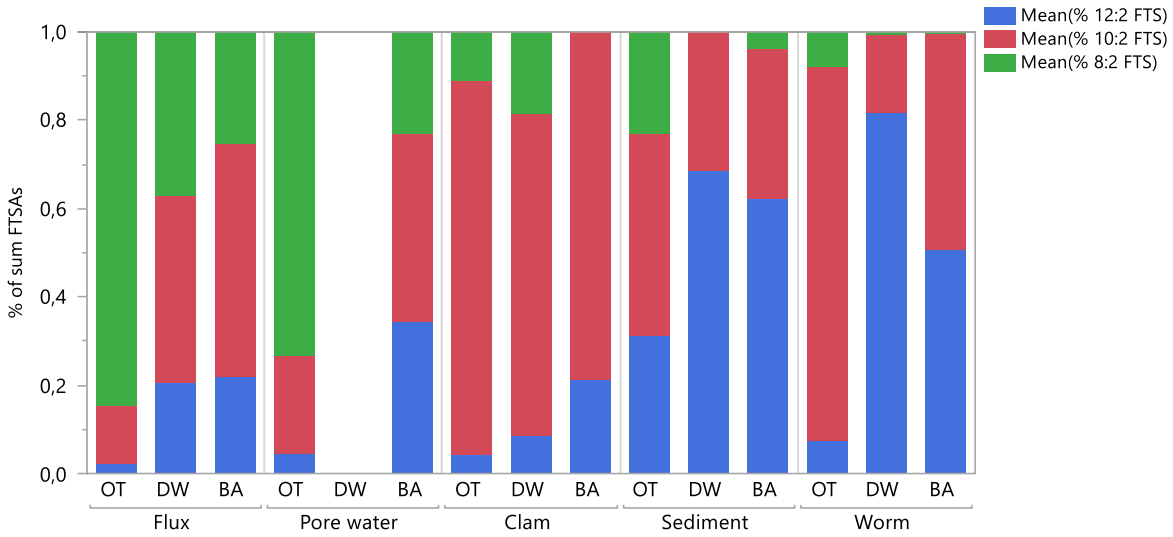


Figure 23. Relative contribution of the three FTSA compounds (% of ∑FTSA) detected in various matrixes analysed in aquaria with sediments collected in Randselva outside the old factory (Ot) and the two downstream locations (Ba and Dw).

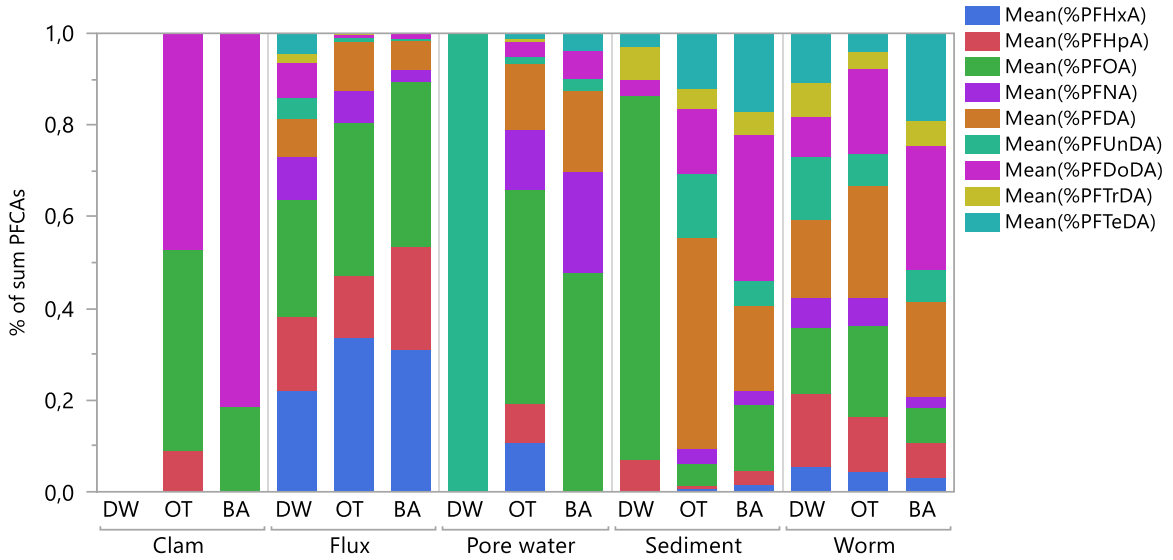


Figure 24. Relative contribution of PFCAs (% of ∑PFCA) in various matrixes analyzed in aquaria with sediments collected in Randselva outside the old factory (Ot) and the two downstream locations (Ba and Dw).

4.6 Natural depletion of PFAS by loss to overlying water

If the sediment is considered a source of PFAS to the overlying and downstream waters, the fluxes measured can be used to estimate the time expected before the sediment reservoir is exhausted. If we consider an average sediment thickness of 10 cm there will be about 50 kg dry sediment per m^2 at the sampling locations Ba and Ot. If the concentration of PFAS determined here are representative for the entire layer we can estimate the amount of PFAS present below each m^2 of the sediment. Divided by the flux, this will give an estimated time required for depletion of the reservoir of the PFAS in the sediments. [2]

Depletion times were calculated for 89 cases with measured sediment concentration > LOQ and flux > 0. The depletion times varied from less than one year for the lighter carboxylates (e.g. 0.5±0.1 yr. for PFHxA) to 10800±6700 yr. for 12:2 FTS at Ot (Figure 25). The depletion times were generally higher at Ot (grand mean = 1200 yr., n=46) than Ba (grand mean = 120 yr., N=43). At both stations, the fluorotelomers came out with the clearly highest depletion times of 2300 yr. for 10:2 FTS and 5800 yr. for 12:2 FTS (median values, both stations).

Flux of N-etFOSE was only detected in one aquarium from Ba. This sample gave an estimated time of depletion of 415 years. Biodegradation is one of the factors which may alter the depletion time. Thus, the degradation products of etFOSE yielded depletion times of 170 yr. for etFOSAA, 271 yr. for etFOSA, 17 yr. for FOSAA, 85 yr. for PFOSA and 72 yr. for PFOS. For this series it appears that biodegradation will enhance site remediation by converting the precursor to products with shorter depletion times. If biodegradation yields an increase in sediment concentration the flux is likely to increase as well and the total effect on depletion time is difficult to predict. It appears likely, however, that biodegradation to compounds with lower molecular weight and higher K_ds are likely to enhance natural site remediation by increased loss to the overlying water.

Remediation by loss to the overlying water will not remove PFAS from the biogeochemical cycle. For further assessment of total environmental effects, bioavailability and toxicity to downstream pelagic organisms will be an important issue. In this respect, the clams tested in this experiment may be considered a pelagic organism accumulating PFAS mostly from the overlying water in each aquarium.

Recent analyses of sediment cores sampled in Svarthølen basin (Helland, pers.com.) showed that most of the PFAS was present within the 0-10 cm layer, but with large vertical variations and typical maxima 2-5 cm below the sediment-water interface. Core maxima and mean concentrations within 0-10 cm are shown for selected compounds in Table 7 together with the concentrations determined in the mixed sediments in the Ba aquaria. The table also shows the depletion times estimated assuming the mean 0-10 cm core concentrations and the fluxes determined in Ba. This comparison is questionable as there tend to be a correlation between sediment concentration and flux, and the fluxes are likely to correspond better with the surface concentrations than with core mean concentrations. The table does however, illustrate that the estimated depletion times rests heavily on the assumptions involved.

[2] In this simplified calculation we assume that the flux remains constant until the reservoir is empty. A more accurate model might consider that the flux is likely to decrease as the reservoir becomes smaller. Therefore the times to depletion calculated here are most likely significantly underestimated.

Table 7. Comparison of depletion times estimated from sediment concentrations and fluxes determined in aquaria with sediments from station Ba and depletion times estimated assuming sediment concentrations recently measured in sediment cores from the basin (Helland, pers. com.).

	Sediment concentration (ng g^{-1} d.w.)			Depletion times (years)		
	Station Ba	Core max.	Core 0-10 cm	Station Ba	Core max	Core 0-10 cm
PFOSA	9	37	16,6	24	99	44
N-EtFOSE	97	250	97	415	349	415
10:2 FTS	262	117	54,1	142	63	29
12:2 FTS	478	262	74,3	823	451	127

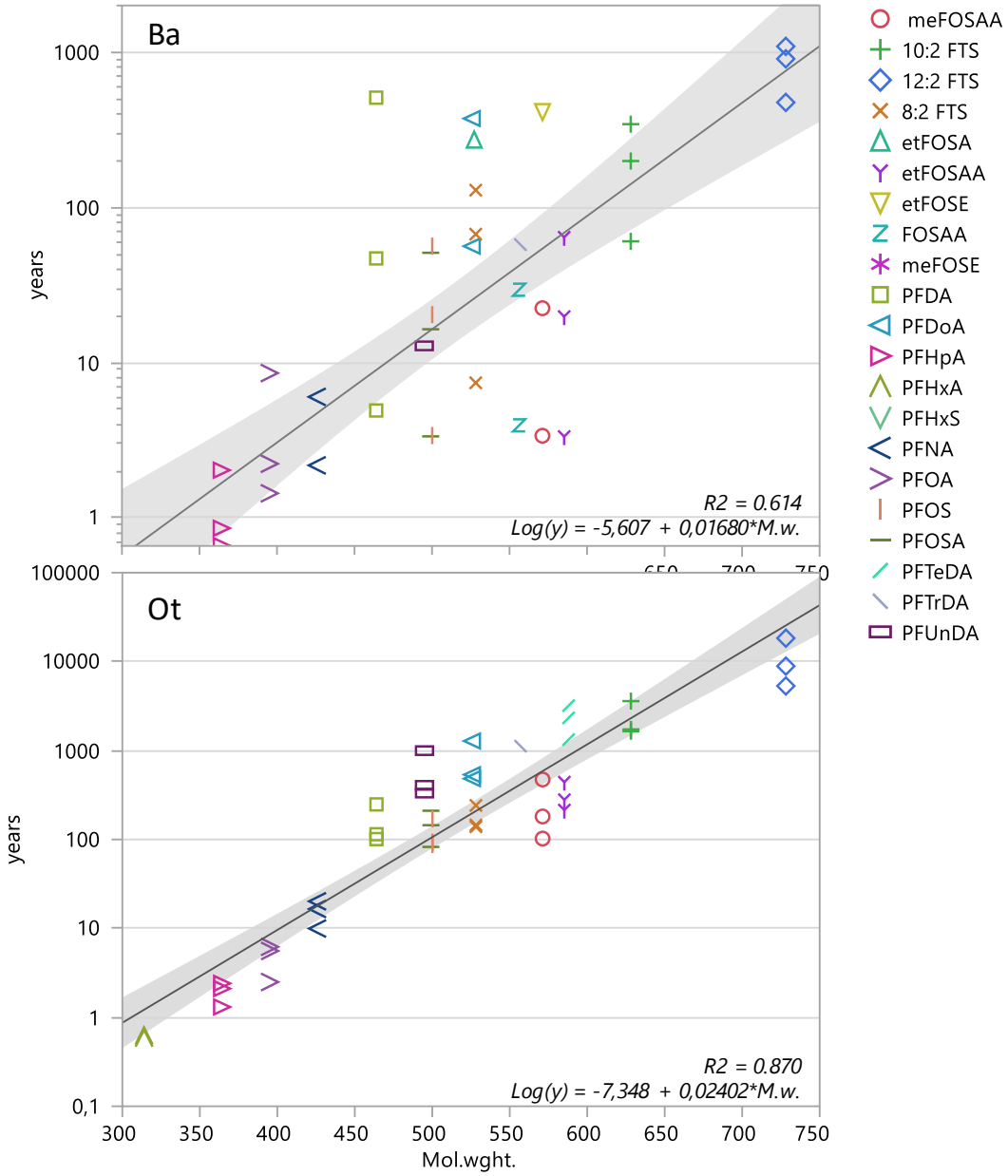


Figure 25. Estimated time for depletion of a load of PFAS corresponding to 10 cm layer thickness with concentrations and fluxes determined in aquaria with sediments from stations Ba (upper diagram) and Ot (lower diagram) in Randselva.

5 Conclusions

- All matrixes (sediment, pore water, fluxes, worms and clams) determined in aquaria with sediments collected in Randselva outside the closed down factory (Ot) and in the downstream Svarthølen basin (Ba) showed high abundances of perfluorinated compounds (PFAS).
- Classification in accordance with Norwegian guidelines for freshwater and sediments provided class III "risk of toxic effects from longterm exposure" for PFOS in aquaria with sediments collected in Randselva at all three sites downstream the old factory (Ot, Ba and Dw) and Lake Årungen (Con).
- Three main groups could account for 80-100% of the PFAS-compounds. These were the FTSAs (fluorotelomeres), the FOSAMs (PFOS and PFOS precursors) and the PFCAs (fluorinated carboxylic acids).
    - FTSAs and FOSAMs were most abundant in sediments and biota in Randselva downstream the factory site (Ot, Ba and Dw).
    - Relative to total PFAS, PFCAs were most abundant in unexposed samples of the biota (Zero) and in aquaria with sediments from control locations in Randselva (Up) and Lake Årungen (Con).
    - Shortchain PFCAs with low K_ds were more frequently detectable as fluxes from sediment to overlying water than compounds with higher molecular weights and higher K_ds.
- Sediment-pore water partitioning coefficients (K_ds) were positively correlated with molecular size and within the range of K_ds determined in laboratory experiments cited in the literature.
- Biota:water Concentration Factors (BCFs) calculated for the clams *(Anodonta anatine)* were positively correlated with molecular weight.
- Biota:Sediment Accumulation Factors (BSAFs) calculated for the worms (*Tubifix tubifix*) showed no correlation with molecular weight at Ba and Dw and a negative correlation at Ot.
- The negative correlation was related to disturbance of feeding behavior and low porewater partitioning due to the presence of petrogenic hydrocarbons.
- Six SAmPAP degradation products were abundant in all matrixes in aquaria with sediments from the three stations in Randselva downstream the old factory. A shift from predominantly early degradation products (EtFOSE and EtFOSAA) near the factory site towards more of the end products (PFOS and PFOSA) at the most distant location (Dw) was observed in all matrixes.
- The fluorotelomers and carboxylates determined in a paper plate claimed to have been produced at the factory in 2007 were all detected in the sediments at Ot and Ba, but with a shift in relative concentrations from predominantly PFCAs in the plate to FTSAs in the sediments.
- Depletion times based on estimated sediment reservoir and current fluxes to the overlying water were positively correlated with increasing molecular size and ranged from less than one year for short-chain carboxylates to 5800 years for the 12:2 FTS.

6 References

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Appendix A. Chemical analyses.

Table A1. Analyses of PFAS in aquarium water. Concentrations < LOQ are given as 0.5*LOQ. LOQs are given in Table 4.

Aq.	Matrix	Date	Unit	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoA	PFTrDA	PFTeDA	PFBS	PFPS	PFHxS	PFHpS	PFOS	8Cl-PFOS	PFNS	PFDS	PFDoS	PFOSA	meFOSA	etFOSA	meFOSE	etFOSE	4:2 FTS	6:2 FTS	8:2 FTS	10:2 FTS	12:2 FTS	FOSAA	meFOSA	etFOSAA
HT	Water	22.02.19	ng/L	3,4	2,2	3,2	0,7	0,25	0,2	0,2	0,2	0,2	0,1	0,1	0,05	0,1	0,1	0,1	0,1	0,1	0,1	0,05	0,15	0,15	0,5	0,5	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP-1	Water	22.02.19	ng/L	2,34	3,18	2,21	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP-2	Water	22.02.19	ng/L	2,22	1,41	4,85	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP-3	Water	22.02.19	ng/L	0,61	1,15	1,24	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,75	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW-1	Water	22.02.19	ng/L	1,96	3,83	2,60	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW-2	Water	22.02.19	ng/L	1,66	1,57	1,65	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	1,30	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW-3	Water	22.02.19	ng/L	1,60	0,97	1,50	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,30	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
OT-1	Water	22.02.19	ng/L	21,30	14,30	17,68	2,15	2,42	0,86	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,31	0,10	0,10	0,10	0,10	0,35	0,15	0,15	0,50	0,50	0,15	0,73	4,14	2,84	0,88	0,15	0,15	0,80
OT-2	Water	22.02.19	ng/L	13,84	5,49	8,74	1,19	1,12	0,30	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,24	0,10	0,10	0,10	0,10	0,11	0,15	0,15	0,50	0,50	0,15	0,65	3,74	1,29	0,15	0,15	0,15	0,81
OT-3	Water	22.02.19	ng/L	19,97	7,53	17,94	4,52	6,65	0,72	0,20	0,20	0,20	0,10	0,10	0,05	0,10	1,33	0,10	0,10	0,10	0,10	0,72	0,15	0,15	0,50	0,50	0,15	1,78	3,46	2,76	0,15	0,15	0,15	4,36
BA-1	Water	22.02.19	ng/L	8,13	11,99	17,88	1,75	0,90	0,20	0,20	0,20	0,20	0,10	0,10	0,27	0,10	1,07	0,10	0,10	0,10	0,10	2,84	0,15	0,15	0,50	0,50	0,15	0,69	1,05	1,73	0,65	0,51	0,15	14,65
BA-2	Water	22.02.19	ng/L	8,54	8,63	17,91	2,47	7,10	1,19	1,07	0,20	0,20	0,10	0,10	0,11	0,10	8,81	0,10	0,10	0,10	0,10	17,47	0,15	0,57	0,50	1,89	0,15	1,09	25,72	20,33	3,56	7,62	2,08	332,97
BA-3	Water	22.02.19	ng/L	3,00	2,91	4,42	0,57	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,22	0,10	0,10	0,10	0,10	0,63	0,15	0,15	0,50	0,50	0,15	0,15	0,63	1,33	0,40	0,15	0,15	5,82
con	Water	22.02.19	ng/L	2,27	2,22	2,21	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
HT	Water	21.03.19	ng/L	0,25	0,25	0,72	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,10	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP-1	Water	21.03.19	ng/L	4,11	0,25	0,86	0,25	0,25	1,09	0,10	0,20	0,20	0,10	0,10	0,05	0,02	0,07	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP-2	Water	21.03.19	ng/L	8,93	0,25	2,47	0,25	0,25	0,20	0,55	0,20	0,20	0,10	0,10	0,05	0,10	0,12	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP-3	Water	21.03.19	ng/L	3,43	0,25	1,26	0,25	0,25	2,41	0,52	0,20	0,20	0,10	0,10	0,05	0,10	0,12	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW-1	Water	21.03.19	ng/L	3,97	1,65	4,53	1,25	0,52	0,40	0,26	0,20	0,10	0,10	0,10	0,15	0,10	1,52	0,10	0,10	0,10	0,10	0,20	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	1,75
DW-2	Water	21.03.19	ng/L	9,29	5,81	12,80	6,37	2,21	1,29	0,70	0,41	0,45	0,10	0,10	0,36	0,10	7,09	0,10	0,10	0,10	0,10	0,59	0,15	0,15	0,50	0,50	0,15	0,27	0,76	0,71	0,51	0,15	0,15	3,93
DW-3	Water	21.03.19	ng/L	6,88	5,89	14,68	4,73	11,61	6,26	14,86	4,15	8,97	0,10	0,10	0,29	0,10	6,64	0,10	0,10	0,10	0,10	0,78	0,15	0,15	0,50	0,50	0,15	1,98	80,46	113,75	41,09	0,02	0,36	9,93
DW-3	*Water*	*21.03.19*	*ng/L*	*6,88*	*5,89*	*14,68*	*4,73*	*11,61*	*6,26*	*14,86*	*4,15*	*8,97*	*0,10*	*0,10*	*0,29*	*0,10*	*6,64*	*0,10*	*0,10*	*0,10*	*0,10*	*0,78*	*0,15*	*0,15*	*0,50*	*0,50*	*0,15*	*1,98*	*80,46*	*113,75*	*41,09*	*0,02*	*0,36*	*9,93*
OT-2	Water	21.03.19	ng/L	54,32	21,93	46,73	7,72	4,57	0,60	0,52	0,20	0,20	0,10	0,10	0,05	0,10	1,25	0,10	0,10	0,10	0,10	0,61	0,15	0,15	0,50	0,50	0,15	3,37	15,74	2,29	0,44	0,15	0,15	3,12
OT-3	Water	21.03.19	ng/L	47,70	14,60	27,59	5,25	6,77	0,99	1,79	0,49	0,95	0,10	0,10	0,05	0,10	1,31	0,10	0,10	0,10	0,10	0,35	0,15	0,15	0,50	0,50	0,15	2,43	30,06	10,55	3,15	0,15	0,15	4,58
BA-1	*Water*	*21.03.19*	*ng/L*	*32,84*	*26,97*	*63,45*	*6,28*	*8,11*	*1,70*	*6,16*	*1,30*	*4,39*	*0,10*	*0,10*	*0,27*	*0,10*	*11,27*	*0,10*	*0,10*	*0,10*	*0,10*	*35,18*	*0,15*	*6,87*	*0,50*	*53,90*	*0,15*	*6,62*	*75,25*	*253,72*	*151,64*	*1,36*	*1,83*	*351,06*
BA-2	*Water*	*21.03.19*	*ng/L*	*19,09*	*11,67*	*33,82*	*5,87*	*13,94*	*2,22*	*3,20*	*0,20*	*0,37*	*0,10*	*0,10*	*0,11*	*0,10*	*20,75*	*0,10*	*0,10*	*0,10*	*0,10*	*42,59*	*0,15*	*3,51*	*0,50*	*20,80*	*0,15*	*1,99*	*50,18*	*86,97*	*8,24*	*7,45*	*5,83*	*786,67*
BA-3	Water	21.03.19	ng/L	6,46	2,82	3,62	0,53	0,51	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,84	0,10	0,10	0,10	0,10	1,45	0,15	0,15	0,50	0,50	0,15	0,32	1,95	4,82	2,52	0,15	0,15	24,47
con	Water	21.03.19	ng/L	3,35	0,25	0,67	0,25	0,25	0,95	0,47	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP 1	Water	08.04.19	ng/L	0,79	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP 2	Water	08.04.19	ng/L	1,60	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP 3	Water	08.04.19	ng/L	0,44	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,18	0,10	0,10	0,10	0,10	0,75	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW 1	Water	08.04.19	ng/L	1,05	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW 2	Water	08.04.19	ng/L	1,62	0,25	1,04	0,46	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,51	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW 3	Water	08.04.19	ng/L	1,15	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,12	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
OT 1	Water	08.04.19	ng/L	46,59	26,10	93,24	17,61	23,84	2,04	1,77	0,20	0,51	0,10	0,10	0,05	0,10	4,10	0,10	0,10	0,10	0,10	2,82	0,15	0,15	0,50	0,50	0,15	8,44	65,35	9,00	1,05	0,06	0,34	10,89
OT 2	Water	08.04.19	ng/L	26,91	8,11	23,20	7,37	11,88	0,91	0,85	0,20	0,53	0,10	0,10	0,05	0,10	2,11	0,10	0,10	0,10	0,10	1,36	0,15	0,15	0,50	0,50	0,15	2,04	43,59	4,42	0,94	0,03	0,26	4,30
OT 3	Water	08.04.19	ng/L	16,70	5,39	19,22	7,72	19,24	1,69	0,92	0,20	0,20	0,10	0,10	0,05	0,10	3,06	0,10	0,10	0,10	0,10	1,66	0,15	0,15	0,50	0,50	0,15	1,92	60,25	4,79	0,44	0,03	0,43	9,61
Ba 1	Water	08.04.19	ng/L	3,82	2,09	3,89	0,51	0,51	0,20	0,39	0,40	0,20	0,10	0,10	0,05	0,10	0,98	0,10	0,10	0,10	0,10	2,48	0,15	0,15	0,50	0,50	0,15	0,37	3,23	11,34	4,73	1,21	0,52	56,62
Ba 2	Water	08.04.19	ng/L	5,11	2,40	6,09	0,98	1,83	0,20	0,60	0,20	0,20	0,10	0,10	0,05	0,10	2,82	0,10	0,10	0,10	0,10	5,68	0,15	0,15	0,50	1,12	0,15	0,37	8,27	18,21	5,43	2,53	0,78	137,31
Ba 3	Water	08.04.19	ng/L	2,42	0,77	0,80	0,12	0,10	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,10	0,08	0,10	0,10	0,10	0,10	0,28	0,15	0,15	0,50	0,50	0,15	0,15	0,30	2,55	2,42	0,15	0,15	1,89
Con	Water	08.04.19	ng/L	2,02	0,42	2,41	0,63	1,22	0,20	0,38	0,20	0,20	0,10	0,10	0,05	0,10	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	4,33	2,95	0,15	0,15	0,15	0,15
HT	Water	08.04.19	ng/L	0,25	0,54	0,80	0,25	0,25	0,20	0,20	0,20	0,20	0,00	0,10	0,17	0,10	0,12	0,10	0,00	0,15	0,15	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15

Table A2. Analyses of PFASs in sediment, pore water (PW) and biota. Concentrations < LOQ are given as 0.5*LOQ. LOQs are given in Table 4.

Aq.	Matrix	Date	Unit	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoA	PFTrDA	PFTeDA	PFBS	PFPS	PFHxS	PFHpS	PFOS	8Cl-PFOS	PFNS	PFDS	PFDoS	PFOSA	meFOSA	etFOSA	meFOSE	etFOSE	4:2 FTS	6:2 FTS	8:2 FTS	10:2 FTS	12:2 FTS	FOSAA	meFOSA	etFOSAA
UP 1	Sed.	08.04.19	ng/g	0,25	1,30	2,39	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,50	0,05	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP 2	Sed.	08.04.19	ng/g	0,25	0,86	2,01	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,46	0,05	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP 3	Sed.	08.04.19	ng/g	0,25	0,32	1,53	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,29	0,05	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW 1	Sed.	08.04.19	ng/g	0,25	0,71	2,35	0,25	0,25	0,20	0,36	0,20	0,20	0,10	0,10	0,41	0,05	0,28	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	1,20	2,32	0,15	0,15	0,35
DW 2	Sed.	08.04.19	ng/g	0,25	0,25	1,56	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,32	0,05	0,38	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	1,18	2,56	0,15	0,15	0,41
DW 3	Sed.	08.04.19	ng/g	0,25	0,25	1,44	0,25	0,25	0,20	0,20	0,44	0,18	0,10	0,10	0,41	0,05	0,46	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	1,22	3,02	0,15	0,15	0,41
OT 1	Sed.	08.04.19	ng/g	4,45	5,87	31,41	22,04	303,44	97,28	97,52	29,65	81,63	0,10	0,10	0,05	0,05	43,65	0,10	0,10	2,06	0,10	29,18	0,15	16,66	22,81	1016,04	0,15	8,85	1175,16	2220,84	1698,30	2,32	4,13	284,50
OT 2	Sed.	08.04.19	ng/g	4,39	5,34	32,15	23,63	324,46	94,30	96,38	28,72	84,07	0,10	0,10	0,05	0,05	44,75	0,10	0,10	1,90	0,10	30,95	0,15	16,40	22,47	1029,59	0,15	9,24	1176,28	2110,49	1541,90	2,56	4,14	267,50
OT 3	Sed.	08.04.19	ng/g	3,65	4,61	29,00	20,94	284,25	88,26	87,13	25,50	76,74	0,10	0,10	0,05	0,05	39,72	0,10	0,10	1,63	0,10	28,66	0,15	17,60	16,40	983,55	0,15	7,72	1008,72	2373,88	1361,10	1,24	2,15	393,80
Ba 1	Sed.	08.04.19	ng/g	0,46	1,21	4,85	1,02	5,01	1,45	8,51	1,31	4,53	0,10	0,10	0,05	0,05	4,49	0,10	0,10	0,10	0,10	9,93	0,15	13,86	0,61	102,08	0,15	0,47	31,17	296,47	538,20	4,90	0,98	163,80
Ba 2	Sed.	08.04.19	ng/g	0,38	0,70	3,50	0,67	4,83	1,48	8,37	1,22	4,44	0,10	0,10	0,05	0,05	4,53	0,10	0,10	0,10	0,10	8,98	0,15	13,29	2,85	97,35	0,15	0,46	29,11	269,69	481,40	4,53	1,01	182,90
Ba 3	Sed.	08.04.19	ng/g	0,30	0,56	2,77	0,65	4,46	1,26	7,50	1,21	4,20	0,10	0,10	0,05	0,05	4,04	0,10	0,10	0,10	0,10	8,57	0,15	12,54	8,88	91,94	0,15	0,43	24,41	220,37	414,40	3,88	0,87	158,40
Con	Sed.	08.04.19	ng/g	0,25	1,51	4,35	0,62	0,20	0,20	0,20	0,36	0,20	0,10	0,10	0,05	0,05	0,14	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP-1	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,78	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,56	0,15	0,15	0,15	0,15
UP-2	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,64	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,44	0,15	0,15	0,15	0,15
UP-3	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW-1	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,09	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,33	0,15	0,15	0,15	0,15
DW-2	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,55	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,11	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,49	0,73	0,42	0,15	0,15
DW-3	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,09	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,44	0,15	0,15	0,15	0,15
OT-1	Clam	08.04.19	ng/g w.w.	0,25	1,36	1,82	0,25	0,25	0,20	1,84	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	2,49	0,15	0,15	0,50	11,80	0,15	0,15	3,08	41,15	1,18	0,15	0,15	1,90
OT-2	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,73	0,25	0,25	0,20	0,72	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,73	0,15	0,15	0,50	2,72	0,15	0,15	2,93	13,63	0,90	0,15	0,15	0,85
OT-3	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,93	0,25	0,25	0,20	1,16	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	1,36	0,15	0,15	0,50	6,91	0,15	0,15	3,60	33,19	1,80	0,15	0,15	1,35
BA-1	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,25	0,25	0,25	0,20	0,45	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	4,29	0,15	0,15	0,50	8,45	0,15	0,15	0,15	21,70	5,95	0,15	0,15	10,02
BA-2	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,46	0,25	0,25	0,20	0,78	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	7,06	0,15	1,09	0,50	21,14	0,15	0,15	0,15	43,00	8,97	0,15	0,15	21,58
BA-3	Clam	08.04.19	ng/g w.w.	0,25	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	1,67	0,15	0,35	0,50	3,52	0,15	0,15	0,15	7,38	2,47	0,15	0,15	6,27
Con	Clam	08.04.19	ng/g w.w.	0,89	2,18	2,60	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,41	0,35	0,15	0,15	0,15	0,15
Zero	Clam	21.03.19	ng/g w.w.	0,25	0,69	0,89	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,05	0,10	0,10	0,10	0,10	0,08	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,60	0,15	0,15	0,15	0,15
UP-1	Worm	08.04.19	ng/g w.w.	0,25	1,26	1,06	0,60	1,57	0,20	0,20	0,42	0,20	0,10	0,10	1,85	0,05	1,06	0,10	0,10	0,10	0,10	0,10	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,37	1,14	0,15	0,15	0,15
DW-1	Worm	08.04.19	ng/g w.w.	0,61	1,76	1,59	0,74	1,86	1,54	0,95	0,83	1,19	0,10	0,10	1,61	0,05	5,88	0,10	0,10	0,10	0,10	2,44	0,15	0,15	0,50	2,27	0,15	0,15	0,33	8,94	41,52	0,15	0,15	0,43
OT-1	Worm	08.04.19	ng/g w.w.	4,17	11,57	19,13	5,90	23,50	6,74	17,88	3,42	3,90	0,10	0,10	0,05	0,05	10,53	0,10	0,10	0,26	0,10	12,54	0,15	0,46	0,50	26,32	0,15	0,15	37,35	406,40	35,52	0,15	0,15	11,63
BA-1	Worm	08.04.19	ng/g w.w.	2,05	5,04	5,02	1,63	13,63	4,66	17,91	3,52	12,68	0,10	0,10	0,90	0,05	42,58	0,10	0,10	0,61	0,10	144,32	0,15	13,01	0,50	510,12	0,15	0,15	10,74	1590,17	1636,90	0,15	0,15	147,83
Con	Worm	08.04.19	ng/g w.w.	1,56	3,24	4,68	2,03	5,20	1,08	0,47	0,40	0,34	0,10	0,10	0,13	0,05	11,25	0,10	0,10	0,50	0,10	0,37	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,56	0,52	0,15	0,15	0,15
Zero	Worm	21.03.19	ng/g w.w.	0,25	1,14	1,10	0,72	1,79	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	1,49	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,28	0,15	0,15	0,15
UP 1	PW	08.04.19	ng/L	0,25	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,73	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP 2	PW	08.04.19	ng/L	0,25	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,50	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
UP 3	PW	08.04.19	ng/L	0,25	0,25	0,25	0,25	0,25	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	0,32	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW 1	PW	08.04.19	ng/L	0,25	0,25	0,25	0,25	0,25	0,49	0,20	0,20	0,20	0,10	0,10	0,05	0,05	2,53	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW 2	PW	08.04.19	ng/L	0,25	0,25	0,25	0,25	0,25	0,84	0,20	0,20	0,20	0,10	0,10	0,05	0,05	2,39	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
DW 3	PW	08.04.19	ng/L	0,25	0,25	0,25	0,25	0,25	0,56	0,20	0,20	0,20	0,10	0,10	0,05	0,05	2,53	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15
OT 1	PW	08.04.19	ng/L	147,20	96,39	683,31	255,00	297,11	25,22	39,55	9,75	17,38	0,10	0,10	0,05	0,05	66,78	0,10	0,10	0,10	0,10	21,58	0,15	2,06	0,50	59,90	0,15	72,27	2661,32	572,56	123,02	2,38	0,15	103,54
OT 2	PW	08.04.19	ng/L	331,62	282,07	1380,55	312,55	406,42	57,41	178,53	32,10	54,38	0,10	0,10	0,05	0,05	89,15	0,10	0,10	0,10	0,10	27,18	0,15	9,61	0,50	289,10	0,15	150,22	3781,97	2085,80	409,62	3,87	0,15	166,69
OT 3	PW	08.04.19	ng/L	411,93	337,33	1777,03	424,13	396,43	28,45	50,25	10,70	21,17	0,10	0,10	0,05	0,05	90,63	0,10	0,10	0,10	0,10	21,69	0,15	4,41	0,50	106,15	0,15	201,13	3995,46	823,55	166,38	2,62	0,15	109,16
Ba 1	PW	08.04.19	ng/L	0,25	0,25	29,95	11,32	8,53	1,50	3,07	0,20	1,75	0,10	0,10	0,05	0,05	16,20	0,10	0,10	0,10	0,10	27,13	0,15	3,59	0,50	20,96	0,15	7,15	61,51	127,85	97,51	8,05	0,15	359,35
Ba 2	PW	08.04.19	ng/L	0,25	0,25	24,51	12,45	9,54	0,97	2,80	0,20	1,97	0,10	0,10	0,05	0,05	19,36	0,10	0,10	0,10	0,10	27,48	0,15	3,24	0,50	17,29	0,15	7,00	60,53	98,36	67,47	7,77	0,15	384,83
Ba 3	PW	08.04.19	ng/L	0,25	0,25	18,55	9,89	8,36	1,60	3,06	0,20	2,03	0,10	0,10	0,05	0,05	15,93	0,10	0,10	0,10	0,10	26,35	0,15	3,92	0,50	21,90	0,15	6,53	55,82	108,24	105,23	5,96	0,15	296,36
Con	PW	08.04.19	ng/L	0,25	65,55	65,52	14,54	2,17	0,20	0,20	0,20	0,20	0,10	0,10	0,05	0,05	3,99	0,10	0,10	0,10	0,10	0,05	0,15	0,15	0,50	0,50	0,15	0,15	0,15	0,15	0,15	0,15	0,15	0,15

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