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Consumption of freshwater fish: A variable but significant risk factor for PFOS exposure

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ABSTRACT

PFOS, PFOA, PFNA and PFHxS are the PFAS substances that currently contribute most to human exposure, and in 2020 the European Food Safety Authority (EFSA) presented a draft opinion on a tolerable intake of 8 ng/kg/week for the sum of these four substances (equaling 0.42 μg/kg if expressed as an annual dose). Diet is usually the dominating exposure pathway, and in particular the intake of PFOS has been shown to be strongly related to the consumption of fish and seafood. Those who eat freshwater fish may be especially at risk since freshwater and its biota typically display higher PFOS concentrations than marine systems. In this study, we estimated the range in PFOS intake among average Swedish "normal" and "high" consumers of freshwater fish. By average we mean persons of average weight who eat average-sized portions. The "normal consumers" were assumed to eat freshwater fish 3 times per year, and the "high consumers" once a week. Under these assumptions, the yearly tolerable intake for "normal" and "high" consumers is reached when the PFOS concentrations in fish equals 59 and 3.4 μg per kg fish meat. For this study, PFOS concentrations in the muscle tissue of edible-sized perch, pike and pikeperch were retrieved from three different Swedish datasets, covering both rural and urban regions and a total of 78 different inland waters. Mean PFOS concentrations in fish from these sites varied from 0.3 to 750 μg/kg. From the available data, the annual min-max dietary PFOS intake for male "normal consumers" was found to be in the range 0.0021–5.4 μg/kg/yr for the evaluated scenarios, with median values of 0.02–0.16 μg/kg/yr. For male "high consumers", the total intake range was estimated to be 0.04–93 μg/kg/yr, with median values being 0.27–1.6 μg/kg/yr. For women, the exposure estimates were slightly lower, about 79% of the exposure in men. Despite highly variable PFOS concentrations in fish from different sites, we conclude that the three most commonly consumed freshwater species in Sweden constitute an important source for the total annual intake even for people who eat this kind of fish only a few times per year. The analyses of PFOA, PFNA and PFHxS showed values which were all below detection limit, and their contribution to the total PFAS intake via freshwater fish consumption is negligible in comparison to PFOS.

1. Introduction

The group of chemicals that is commonly referred to as PFASs, or per- and polyfluorinated alkyl substances, contains a large number of industrially produced compounds. They all contain stable covalent carbon-fluorine bonds as well as both hydrophilic and hydrophobic parts. This results in molecules that are extremely persistent, e.g. resistant to high temperatures, and with grease, oil, and water repellant properties. These technically useful characteristics have made PFASs widely used in a variety of industrial and consumer products (Wang et al., 2017). At present, perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorooctane sulfonate (PFOS) and perfluorohexane sulfonic acid (PFHxS) are the PFAS substances that contribute most to human exposure (EFSA, 2020). They have a similar potential for biological uptake, are distributed and accumulated in target organs in a similar way, share toxicokinetic properties, and have the potential to induce the same detrimental effects in mammals (EFSA, 2020). Numerous studies from the past two decades show how these

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chemicals can now be found in basically all types of environmental compartments; in sediments, surface- and groundwater, soil and biota (Houde et al. 2006, 2011; Lau et al., 2007). Although several manufacturers have acted to phase out many long-chained PFASs over the past two decades, the recent review by Land et al. (2018) concludes that concentrations in the environment are not decreasing. Human body burdens of PFOS and PFOA seem to be declining however, at least in Europe and North America, according to the same review. In Sweden, it has recently been suggested that also PFNA and PFHxS are impacting humans to a lessening degree (Miaz et al., 2020; Nyberg et al., 2018). The declining human body burdens, when indicated, are thought to be due mainly due to the removal of certain PFASs from consumer products, such as food packaging materials, where they have previously been used (Hu et al., 2018; Schaider et al., 2017; Susmann et al., 2019).

People may be exposed to PFOA, PFNA, PFOS and PFHxS through a range of different exposure pathways, but diet often dominates (Fromme et al., 2009; Haug et al., 2011; Vestergren and Cousins, 2009). In February 2020, the European Food Safety Authority (EFSA) announced that its CONTAM Panel had presented a revised draft opinion on health-based tolerable intake levels for the sum of these four PFASs. The proposed limit is 8 ng/kg body weight/week for this sum, which means a dramatic lowering of the previous EFSA CONTAM tolerable intakes, which were 150 ng/kg bodyweight and *day* for PFOS and 1500 ng/kg/*day* for PFOA. The critical effect in humans –which is the endpoint motivating the revision – is a decreased immune system response to vaccination. Although the critical effect used in the risk assessment is based on studies in children, exceedance of the tolerable intake is regarded as a health concern for the entire human population, irrespective of age and gender.

With regards to the dietary intake, we know that fish and seafood are key contributors, in particular of PFOS (EFSA, 2018; Gebbink et al., 2015; Haug et al., 2010; Noorlander et al., 2011; Vestergren et al., 2012). This conclusion is drawn from previous dietary exposure assessments that are based on mean concentrations in commercial food; fish products as well as other items. PFOS concentrations in commercial fish are then typically below 3 μg/kg (EFSA, 2018; Haug et al., 2010; Noorlander et al., 2011; Vestergren et al., 2012). Commercial fish, however, typically originates from marine fishing grounds. And marine fish species usually contain significantly lower concentrations of various PFAS substances than freshwater fish (Berger et al., 2009; EFSA, 2020; Yamada et al., 2014b). Whereas dilution acts to keep concentrations low in the marine environment, the proximity to high-emission point sources may render severely contaminated hotspots in freshwater systems. In Lake Halmsjön, near the largest airport in Sweden (Arlanda), for example, PFOS concentrations of 792 μg/kg have been found in perch muscle (Norström et al., 2015). If a woman of average weight (69 kg) were to eat fish with this PFOS content, already 27 g per year, or about 20% of a normal-sized fish portion (130 g), would be enough to reach the maximum annual intake (and then leave no room for exposure from other dietary sources and to the other three PFASs).

In general, the health benefits associated with fish consumption are assessed to outweigh the risks related to the potential exposure to certain environmental pollutants (Domingo et al., 2007; Mozaffarian and Rimm, 2006; Rodriguez-Hernandez et al., 2016). However, most studies that assess these risks are based on analyses of marine fish species, and PFAS substances are not among the commonly addressed contaminants (Baldwin et al., 2016; Blocksom et al., 2010; Hinck et al., 2009; Maruya et al., 2012). From a food safety perspective, PFASs may therefore be considered contaminants of emerging interest, even though the use of certain PFAS has been restricted. The study by Rose et al. (2015), which surveyed the occurrence of a wide range of contaminants in fish from 23 freshwater sites (rivers, lakes and canals) across the UK, showed that PFOS was the most prevalent compound, detected in every fish sample tested – with concentrations ranging from 2 to 153 μg/kg.

To sum up, exposure assessments that use contaminant concentrations in commercially bought fish may underestimate the intake of PFAS via fish consumption for people who consume a significant amount of freshwater fish, for example recreational fishermen and their relatives. Therefore, the remainder of this article will focus on the role of freshwater fish consumption for the dietary intake of PFOS in particular. PFNA, PFOA and PFHxS are also analysed, but these are usually found primarily in other categories of food and are expected to have much lower concentrations in freshwater fish than PFOS (EFSA, 2020).

Using three different sets of data from Sweden, the aims of the study presented here were to investigate:

1) PFOS, PFNA, PFOA and PFHxS concentrations in edible parts of Swedish freshwater fish (fish muscle) and its variability;
2) what levels of exposure, in particular of PFOS, that can be expected among people who consume Swedish freshwater fish on a regular basis.

2. Materials and methods

2.1. Datasets for characterisation of PFOS, PFOA, PFNA and PFHxS in freshwater fish

To retrieve PFAS concentrations in edible fish meat for utilisation in the exposure assessments, we evaluated three Swedish datasets. The first was compiled for this study. The other two were originally collected for the purpose of environmental monitoring rather than for this research project. The advantage of including all three datasets is the large amount of data it provides: a total of 78 sites are included. The disadvantage is that there is a lack of consistency when it comes to sample preparation and analysis, making comparisons between datasets difficult.

We have focused on concentrations in fish muscle from the three most commonly consumed freshwater species in the country: pike (*Esox lucius*), perch (*Perca fluviatilis*) and pikeperch (*Sander lucioperca*). What the sampling sites for all three datasets have in common is that i) it is relatively easy to catch one or more of the three fish species there, and ii) they are accessible for recreational fishing. Most of the sites are not directly affected by known point sources. A complete list of all sampling sites is provided in the Supplementary Material (Table S1).

The first dataset represents a semi-rural region, exemplified by the county of Kronoberg in southeastern Sweden; "Semi-Rural Kronoberg". This was the only dataset for which sampling and PFAS analyses were performed specifically for the present study. A total of 65 fishes (20 perch, 20 pike, 25 pikeperch) from 6 lakes were caught and individually analysed. The lakes were not located in or near any of the larger cities in the region, and were not in direct proximity to known point sources. Fish of normal consumption size (adult life-stage) were caught between November 3, 2017 and October 23, 2018 using either angling equipment (i.e. rod, reel and artificial bait), fyke-nets or purse seining. The chemical analyses are described below. They were performed at the same lab and using the same methods as applied to the second dataset.

The second dataset represents an urban region. It is from the capital of Sweden, "Urban Stockholm", and was provided by the Stockholm City Environmental Office. This dataset covers 15 sites, with annual analyses of perch from the time period 2015–2018. These samples had already been analysed as composite samples, with material from 15 individual adult (edible-sized) fishes in each sample.

And finally, the third dataset covers the rest of "Sweden". This data was retrieved from the Water Information System Sweden (WISS), which is an open access database for the monitoring of Swedish water quality, administered by the County Administrative Boards. Due to a partial overlap between dataset 3 and the first two datasets, the overlapping sites were removed from the third dataset. The remaining data covered 57 sites, where we selected data from the years 2013–2018. All three fish species are represented in the Sweden dataset, but perch dominates (>80%). As for the second dataset, the data here also comes from analyses of pooled samples (based on 10 or more adult (edible-sized) fishes per sample). Since the WISS database does not provide any details on the laboratory procedures, we can only assume that there may be differences with respect to this.

2.2. Chemical analyses of fish from "Semi-rural Kronoberg" and "Urban Stockholm"

The analyses of all samples for dataset 1 and 2 were conducted at the Swedish Environmental Research Institute (IVL). Homogenised fish muscle samples (approximately 1 g) were spiked with 10 ng of an isotope labelled internal standard mix of $^{13}C_4$ sodium perfluorooctane sulfonate ($^{13}C_4$-PFOS) and $^{13}C_4$ sodium perfluorooctanoic acid ($^{13}C_4$-PFOA). The spiked extracts were equilibrated at room temperature for 30 min before extraction with 2 × 5 mL acetonitrile in ultrasonic bath for 30 min. The extracts were combined and concentrated to 1 mL using a gentle N_2-stream followed by a clean-up step using acidified graphitized carbon (50 μL glacial acetic acid in 25 mg Supelclean ENVI-Carb 120/400). After centrifugation (10 000 rpm, 10 min) the supernatants were further concentrated to 0.5 mL, whereafter 0.5 mL of ammonium acetate solution (4 mM) was added. The extracts were kept in a freezer (−20 °C) overnight to facilitate the precipitation of substances that react with the added ammonium acetate (metals, proteins etc), and that may otherwise affect the sensitivity of the PFAS analysis. Before analysis, the samples were brought up to room temperature and after centrifugation (10 000 rpm for 10 min) the supernatants were collected in LC vials, followed by the addition of 50 μL of 3,5-bis(trifluoromethyl)phenyl acetic acid (3,5-BTPA) as injection standard before the analyses. The PFAS analyses were performed by employing high performance liquid chromatography (Shimadzu LC-20 series) coupled to tandem mass spectrometry (API 4000™ system, AB SCIEX) in negative electron spray ionisation (-ESI). The quantification of PFASs was carried out using Analyst software (Analyst 1.63, AB SCIEX). The detection limits were 0.2 μg/kg fresh weight for PFOS and PFOA, 0.1 μg/kg for PFNA, and 0.02 μg/kg for PFHxS.

2.3. Characterisation of exposure scenarios

The exposure calculations of this study consider four different exposure scenarios, accounting for two different kinds of fishing patterns and two levels of freshwater fish consumption.

Regarding the fishing pattern, recreational fishermen can either fish in the same lake the entire time (we call this "**immobile fishing**") or in different lakes ("**mobile fishing**"). Regarding the fish consumption levels, we assume that "**high consumers**" eat freshwater fish once a week, and that "**normal consumers**" do so three times a year. The "high consumer" scenario represents consumption of freshwater fish at the 99th percentile in the food consumption survey for adults in Sweden (2010), and the "normal consumer" scenario mirrors the average consumption in the same study population (Amcoff, 2014; SFA, 2012). Combined with the fish portion sizes mentioned in the section about Exposure assessment below, this gives an annual fish consumption for our hypothetical "normal consumers" of 390 g for women and 600 g for men, equalling 1.1 g and 1.6 g fish per day, respectively. For the "high consumers", the same calculation gives 6800 g/year (19 g/day) for women, and 10 000 g/year (28 g/day) for men.

The characterisation of our "high" and "normal" fish consumption levels may deserve some further motivation, or perspective. Although data on the consumption of self-caught fish is limited, the reviewed literature suggests that these estimates are reasonable. In the latest national survey of food consumption, from which we have derived our data on fish portion sizes and body weight as mentioned above, less than 1% of the 1797 participants indicated that they eat perch, pike and pike-perch at least once a week. About 45% indicate that they consume this type of fish 1–3 times a year. However, recreational fishermen may well differ from the general population in how much freshwater fish they eat, and the available literature implies that a non-negligible proportion eat their catch regularly. For example, a study among French anglers revealed that as much as 30% ate self-caught fish more than 6 times per year (Denys et al., 2014). Among recreational fishermen in North America, Kosatsky et al. (1999) reported that the population in their study ate on average 0.92 meals of game fish per weak, and average daily intakes have been reported to range from 16 to 21 g (Abdelouahab et al., 2008; Harris and Jones, 2008). That is not far from the "high consumers" in our study, with an estimated 19 g of fish per day for women and 28 g/day for men. Yet another study (Philibert et al., 2006) showed that about a quarter of the recreational fishers surveyed in their study consumed more than 66 g of locally caught freshwater fish per day. So in conclusion, it seems plausible to assume that we have not insignificant subpopulations of recreational fishermen who eat freshwater fish at least once a week. In addition, while not addressed in this study, the recent increase in migrants to Sweden – as well as many other European countries - may include subpopulations where fishing and consumption of freshwater fish is a cultural norm.

2.4. Exposure assessment

As described above, this study characterises variability in exposure – covering the scale from high to low intakes –for four different scenarios by considering two different fishing patterns ("mobile"/immobile") and two levels of fish consumption ("normal"/"high"). Exposure simulations were made separately for each scenario, and also for each of the three datasets. Calculations were further made separately for adult men and women. Children were not included, however, because of the scarcity of data on their fish consumption.

In short, the contaminant intakes associated with the consumption of freshwater fish were simulated for 1000 hypothetical persons for each scenario. Since freshwater fish is typically consumed only a few times per year by Swedish consumers (Borthwick et al., 2019), we chose to calculate and evaluate the intake on a yearly basis rather than per day or week. To do so, we firstly calculated the contaminant intake for each separate fish meal (AIM; μg per kg bodyweight). The intakes from all meals eaten over a year were then added to give the annual exposure figure for a certain simulated person. The AIM was calculated as:

$$AIM = Rig^{*}Cf/BW$$

where Rig is the average fish portion size (kg), Cf represents the concentration of the critical PFAS substances in the consumed fish meat (μg/kg) and BW is the human body weight (kg). Body weights and fish portion sizes were characterised by the same mean values in all simulations. Data on these variables were retrieved from the population of women and men included in the latest national food survey conducted by the Swedish Food Agency (SFA). The average body weights here were 69 kg for women and 84 kg for men (SFA, 2012). Average portion sizes of freshwater fish were 130 g for women and 200 g for men (Amcoff, 2014).

The fish contaminant concentration (Cf) was for each simulation randomly selected from the evaluated datasets. Fig. 1 illustrates the principle of this selection, using "normal consumers" as the example. In a bootstrapping procedure, a fishing site is first drawn at random from those sites available in the target dataset. Then, one of the available fish samples from that particular site is drawn. For immobile fishermen only one random site is picked, from which three samples are drawn for the calculation of annual exposure. For mobile fishermen, three sites are selected. The same site could be selected multiple times. The procedure was repeated 1000 times for each scenario, generating 1000 unique figures of annual exposures for hypothetical "normal consumers". Combined, the results give a range in exposure that covers the scale from low to high intakes for "normal consumers". The same principle for picking fishing site(s) was then applied to "high consumers", but the number of meals per year was increased to 52.

An important detail to recall is that data on PFOS concentrations in individual fishes was only available for "Semi-Rural Kronoberg",

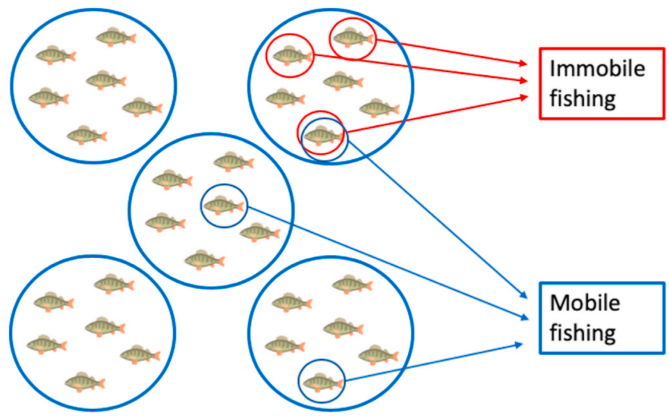

Fig. 1. Principle of how fishing sites and PFOS concentrations in fish from these are randomly picked in a bootstrapping procedure to generate PFOS concentration indata for the exposure calculations.

whereas the other two datasets were based on composite samples. The available mean concentrations from "Urban Stockholm" and "Sweden" were, however, dealt with in the same way as the concentrations from Kronoberg (a random fishing site was first picked, after which a sample was drawn), even though this discrepancy obviously contributes with uncertainty when the results from the three datasets are compared.

3. Results and discussion

3.1. PFOS (+PFOA, PFNA and PFHxS) concentrations in Swedish freshwater consumption fish

The analyses of the "Semi-Rural Kronoberg" fishes, as well as the "Urban Stockholm" material, revealed detectable PFOS concentrations in all the samples. The remaining compounds were consistently below detection limit (0.2 μg/kg for PFOA, 0.1 μg/kg for PFNA, and 0.02 μg/kg for PFHxS). For our results to be comparable with the EFSA Lower Bound (LB) approach (EFSA, 2020), we too assigned a value of zero to all samples below detection limit, in other words, for all the analyses of PFOA, PFNA and PFHxS. This means that our exposure assessment, and comparisons with the EFSA tolerable intake, is based solely on PFOS.

Fig. 2 shows the location of all the fishing sites included in the study. The color of each point indicates the mean PFOS concentration found in fish muscle from that site. More specific data for each point is given in Table S1 in the supplementary material. For the "Sweden" dataset, the mean concentrations from the 57 different sites ranged from 0.3 to 750 μg/kg. The median of these site mean values was 5 μg/kg and the mean of the means was 27 μg/kg. The distribution is thus skewed to the right, with half of the sites displaying mean PFOS concentrations in fish muscle <5 μg/kg. For the 15 sites of the "Urban Stockholm" dataset, mean concentrations ranged from 2.9 to 44 μg/kg, with a median and mean for all sites of 8.9 and 12.6 μg/kg, respectively. And finally, the "Semi-Rural Kronoberg" sites had mean PFOS concentrations between 0.9 and 13.0 μg/kg, with the median and means being 3.8 μg/kg and 4.5 μg/kg, respectively. When the results for the samples from the "Semi-Rural Kronoberg" material, where the fishes were analysed individually, were compared with the pooled samples from "Sweden" and "Urban

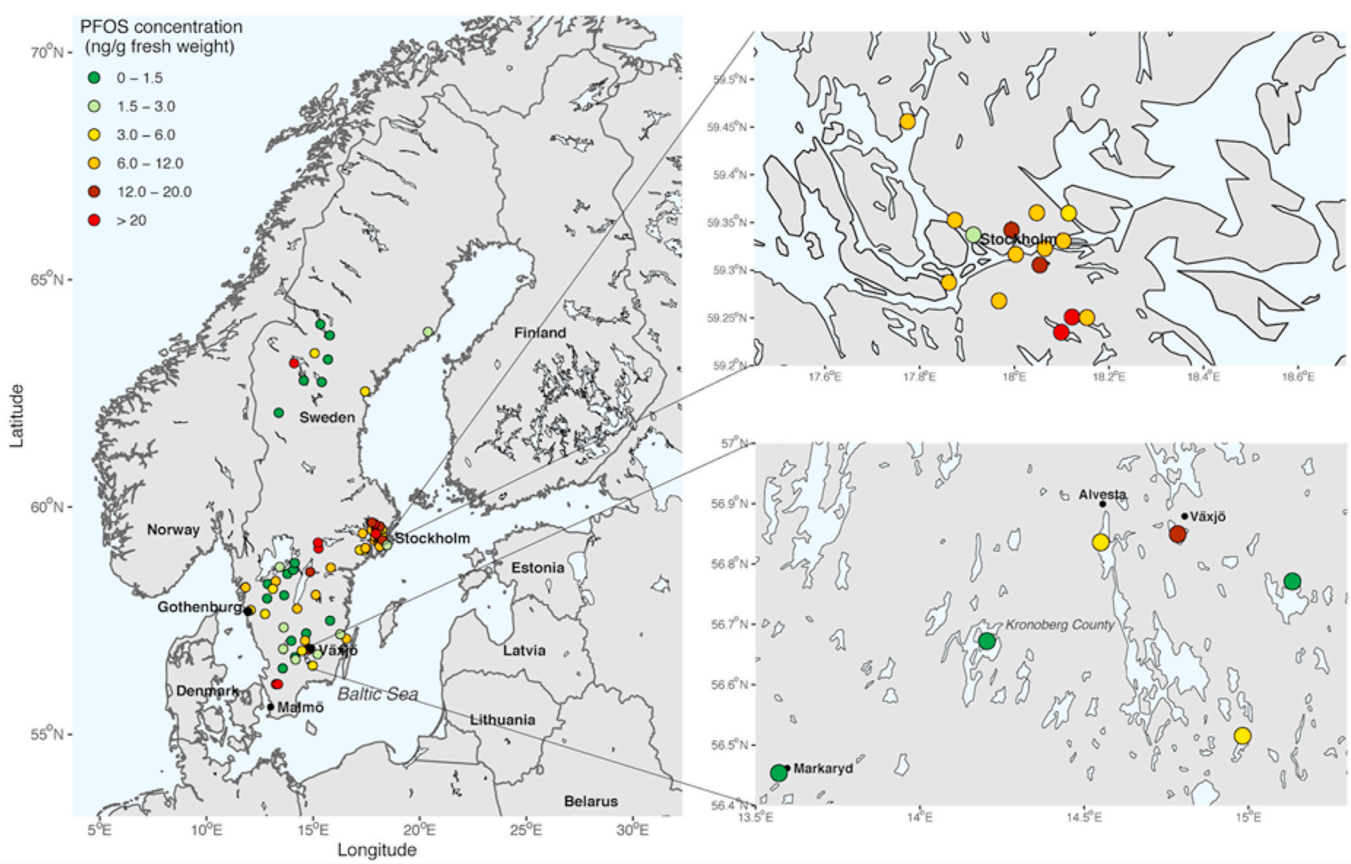

Fig. 2. Maps showing the location of all fishing sites from which data was retrieved, with mean PFOS concentrations in fish muscle from each site color-indexed. The figure shows the 57 sites in the Sweden dataset (left subfigure), 15 sites of the Urban Stockholm dataset (upper right corner) and 6 sites in Semi-Rural Kronoberg (lower right corner). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Stockholm", we observe – as expected – that the range of site mean concentrations derived from pooling fishes can be markedly lower than the range revealed when analyses are made on individual fishes. This information is relevant when we want to assess exposure levels for people who eat freshwater fish only sporadically (and may have the misfortune of consuming a fish with a particularly high PFOS concentration). In the Kronoberg lake with the mean value of 13.0 μg/kg, for example, one of the ten analysed fishes (a perch) contained 22 μg PFOS per kg muscle tissue, i.e. almost twice the mean value (Table S1). The Kronoberg dataset also implies that the largest source of variation in fish PFOS concentrations is fishing location, and not which fish species were caught (Se Supplementary Fig. S1). Thus, human PFOS exposure related to eating "freshwater fish" in this study is assessed without any adjustments for fish species.

All three datasets evaluated in the present study confirm a high variability in concentrations of PFOS in Swedish freshwater fish. In addition, these concentrations are generally higher than those found in European commercial fish, which most commonly originates from marine waters. Pooled fish product samples from grocery stores in the Netherlands (Noorlander et al., 2011), Sweden (Vestergren et al., 2012) and Norway (Haug et al., 2010) showed PFOS concentrations between 0.06 and 1.3 μg/kg. In the EFSA compilation from 2018 (EFSA, 2018), a mean range of PFOS between 2.2. and 2.8 μg/kg was reported for commercial fish meat in the EU. In the updated EFSA report from 2020 (EFSA, 2020), concentrations are provided for 10 different kinds of commercially marketed fish; including one strictly freshwater species (carp) while the remainder were marine (herring, sardine, anchovy, salmon + trout, mackerel, tuna, cod + whiting, halibut and eel). The mean PFOS concentrations in the marine species were between 0.16 and 9.2 μg/kg, while the mean concentration in freshwater carp was 14.1 μg/kg. The same conclusion about higher PFOS concentrations in fish from freshwater systems was drawn by Berger et al. (2009), who compared concentrations in fish muscle from Lake Vättern in Sweden (where mean concentrations in perch were 11.3 μg/kg) and the Baltic Sea (mean concentration 2.1 μg/kg). Another study that convincingly showed the same difference was conducted by Yamada et al. (2014b), who analysed 94 composite samples of the most commonly consumed marine species in France (with the resulting mean value being 0.58 μg/kg) and 387 composite samples covering the 16 most consumed freshwater species from 18 different sites (where the mean value was nearly 80 times higher, 45.2 μg/kg).

3.2. PFOS exposure among freshwater fish consumers

Under the applied assumptions, the maximum dietary intake proposed by EFSA (0.42 μg per kg bodyweight and year) is reached for male "normal consumers" when the consumed fish contains on average 59 μg PFOS per kg fish meat. For male "high consumers", a mean concentration of 3.4 μg PFOS per kg consumed fish is enough to reach the annual maximum intake. Already a straightforward comparison with the above presented PFOS concentrations in fish thus implies that we will have consumers with PFOS intakes exceeding the proposed tolerable intake.

A summary of the estimated PFOS exposure figures from the present study, calculated from the three datasets jointly, is provided in Table 1. Generally, men eat larger portions in relation to their bodyweight than women (200 g fish per portion/84 kg bodyweight in comparison to 130 g/69 kg). Since only average bodyweights and fish portion sizes were used in the calculations, the resulting PFOS intakes per kg body weight are consistently higher (1.26 times) for men than for women. The following discussion will therefore focus mainly on men, for which the exposure estimates are the highest. The results of the four evaluated scenarios are further displayed in Fig. 3, which shows cumulative exposure curves for each dataset individually. As seen in the two left subfigures, the annual PFOS exposure for average male "normal consumers", resulting from 3 meals of freshwater fish, varies from 0.006 to 3.6 μg/kg/yr (median 0.05–0.16) for mobile fishers and 0.002–5.4 μg/kg/yr (median 0.02–0.07) for immobile fishers. It may be worth repeating that we by average here mean someone of average bodyweight who eats average-sized fish portions. The variability in exposure is thus solely the result of the variability in fish PFOS concentrations. As seen, almost all "normal consumer scenarios" are below the EFSA guidance value for the sum of PFOS, PFOA, PFNA and PFHxS, which expressed as an annual dose equals 0.42 μg/kg/yr. This value was exceeded at the 92nd and 96th percentiles when the exposure was estimated from the "Sweden" dataset (mobile and immobile fishers, respectively), and at the 99th percentile when using the "Urban Stockholm" dataset. Note that the curves in Fig. 3 are truncated at the 95th percentile.

For the high consumer scenario, illustrated by the two right subfigures of Fig. 3, the annual PFOS exposure ranges from 0.13 to 22 μg/kg/yr (median 0.27–1.6) for mobile fishers and 0.04–93 μg/kg/yr (median 0.45–1.3) for immobile fishers, depending on which dataset is used. All scenarios show an exceedance of the EFSA guidance value of 0.42 μg/kg/yr at some point. For the "Urban Stockholm" dataset under a mobile fishing scenario, this happens already at the first percentile - implying that basically anyone who would fish in these urban waters and eat this kind of fish once a week would reach an annual PFOS dose above the tolerable intake level. In "Semi-rural Kronoberg", however, the level of 0.42 μg/kg/yr is not reached until the 50th (immobile) or 95th (mobile) percentile. These contrasting results clearly point to the significance of where the fish is caught, but nevertheless also indicate the potential of freshwater fish for the dietary PFOS intake. Previous studies on PFOS exposure from freshwater fish consumption have reported similar, or even higher figures for their mean "high consumer scenarios". Berger et al. (2009) and Squadrone et al. (2014), for

Table 1

PFOS exposure (μg/kg/yr) after consumption of Swedish freshwater fish for female and male consumers, when not making any distinction between the three datasets. Bold values indicate figures above the EFSA guidance value for the sum of PFOS, PFOA, PFNA and PFHxS (8 ng/kg/week, or 0.42 μg/kg/yr). Note that with the assumptions made in our calculations, with constant body weights and fish portion sizes, the figures are consistently 26 times higher for men than for women.

	Mean	Min	P5	P25	Median	P75	P95	Max
"NORMAL CONSUMERS"								
Men, mobile	0.14	0.0021	0.021	0.056	0.091	0.16	0.26	**3.6**
Women, mobile	0.11	0.0017	0.017	0.044	0.072	0.13	0.21	**2.9**
Men, immobile	0.11	0.0021	0.0046	0.014	0.036	0.082	0.26	**5.4**
Women, immobile	0.087	0.0017	0.0037	0.011	0.029	0.065	0.21	**4.3**
HIGH CONSUMERS"								
Men, mobile	**1.8**	0.10	0.19	0.33	**1.2**	**1.8**	**8.0**	**22**
Women, mobile	**1.4**	0.079	0.15	0.26	**0.95**	**1.4**	**6.3**	**17**
Men, immobile	**1.8**	0.036	0.11	0.35	**0.62**	**1.4**	**4.5**	**93**
Women, immobile	**1.4**	0.029	0.087	0.28	**0.49**	**1.1**	**3.6**	**74**

Values in bold > the porposed EFSA guidance value

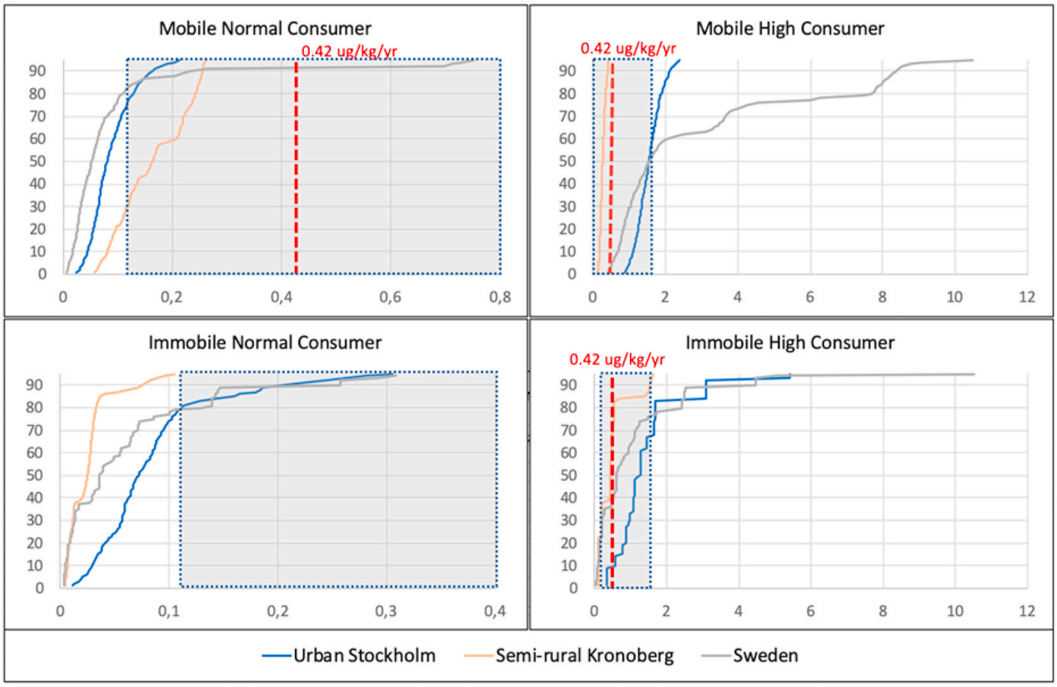

Fig. 3. Cumulative distribution functions, truncated at the 95th percentiles, of annual exposure rates (μg/kg/yr) for male individuals who consume freshwater fish on a "normal basis" (3 times per year) or as "high consumers" (once a week), and depending on whether their fishing pattern is mobile or immobile. Each subfigure contains three different lines, representing the three different datasets evaluated. The vertical, red, dashed lines represent the proposed EFSA guidance value for the sum of PFOS, PFOA, PFNA and PFHxS (8 ng/kg/week), expressed as an annual dose (0.42 μg/kg/yr). The grey areas mark the range in average total dietary PFOS intake reported by EFSA (2018), which is 0.11–1.5 μg/kg/yr. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

example, presented average intake levels among high consumers of 2.7 ng/kg/d (= 0.99 μg/kg/yr) and 54.4 ng/kg/d (= 19.9 μg/kg/yr), respectively. In comparison to our assumptions they have, however, used higher consumption rates and slightly lower bodyweights (the former assumed 4 meals of freshwater fish per week (à 125 g) and 65 kg bodyweight, and the latter ca 150 g fish/day and 60 kg bodyweight). The "high freshwater fish consumers" targeted in the study by Yamada et al. (2014a) may resemble our "high consumer" group better, with a consumption frequency of ≥45 times per year. For this group, the authors assess the mean and P95 PFOS intake to be 2.7 and 11 μg/kg/yr, respectively.

When it comes to PFOA, PFNA and PFHxS only a worst-case scenario can be estimated since the concentrations were below detection limits (0.2, 0.1, and 0.02 μg/kg, respectively) in all samples. This worst-case scenario was calculated from the assumption that the true concentrations were exactly at the detection level in each sample. For a high consumer, eating 200 g fish per week and weighing 84 kg, the annual intake of PFOA, PFNA and PFHxS would then add up to 0.04 μg/kg/yr. This is about 10% of the tolerable intake level proposed in the recent draft opinion from EFSA (2020). In comparison to the exposure levels estimated for PFOS under the "high consumer scenarios" (Table 1 + Fig. 3), we see that this intake is negligible.

We can conclude, however, that freshwater fish must be viewed as a very important source of PFOS exposure, when related to normal dietary PFOS intake levels. According to EFSA (2018), the average total dietary PFOS intake from all types of food among European adults is 0.29–4.08 ng/kg/day, equaling 0.11–1.5 μg/kg/yr. At least 50% is said to relate to the consumption of (commercial) fish. Other estimates of the total dietary PFOS intake found in the scientific literature are within the same range, or slightly lower, than the EFSA figures. Food basket surveys from Holland (Noorlander et al., 2011), Sweden (Gebbink et al., 2015; Vestergren et al., 2012) and Norway (Haug et al., 2010) suggest an intake via the dominating food categories in these countries to lie between 0.2 and 1.0 ng/kg/day (=0.073–0.37 μg/kg/yr). A study by Fromme et al. (2007), which was based on analyses of 214 portions prepared as for consumption, estimated the median dietary intake to be 1.4 ng/kg/day (= 0.51 μg/kg/yr). The P10–P90 interval according to the same study was 0.7–3.8 ng/kg/day (=0.26–1.4 μg/kg/yr).

Comparing these figures with the distributions of Fig. 3, where the EFSA interval of 0.11–1.5 μg/kg/yr is marked in grey, we see that even the 3 fish meals per year that we ascribe to a "normal consumer" is enough to reach what is assumed to be a normal total annual dietary PFOS intake. For the high consumers, as much as 50% of the individuals (looking at the Sweden dataset) may even exceed it.

When comparing studies from different years, as well as when comparing different food categories, it is important to note that PFOS concentrations in a lot of commercial food has decreased over the last decades. The date of sampling of fishes that are the source of the PFOS concentration raw data is thus something to keep in mind when evaluating estimates of annual exposures. Based on Swedish market basket surveys, for example, a statistically significant decrease was seen in the overall median dietary per capita PFOS intake between 1999 and 2015, corresponding to a 9.6% decrease/year (Swedish National Food Agency, 2017). PFOS concentrations in commercial fish meat from Swedish grocery stores, however, did not display the same general decrease according to the report. And as mentioned in the introduction to this article, concentrations in environmental compartments and biota are not (yet?) decreasing to the same extent as in humans, where humans come in less direct contact with PFAS-containing consumer products today (Land et al., 2018). Fig. 4 shows time trends for the sampling sites of the "Urban Stockholm" dataset of our study. This is the only dataset for which we have data from more than one time point. It is a limited period of time but, as shown in the figure, we cannot say that average PFOS concentrations in the fishes caught have decreased since 2015. We therefore assume that our exposure estimates are representative for at least for the past 5 years or so.

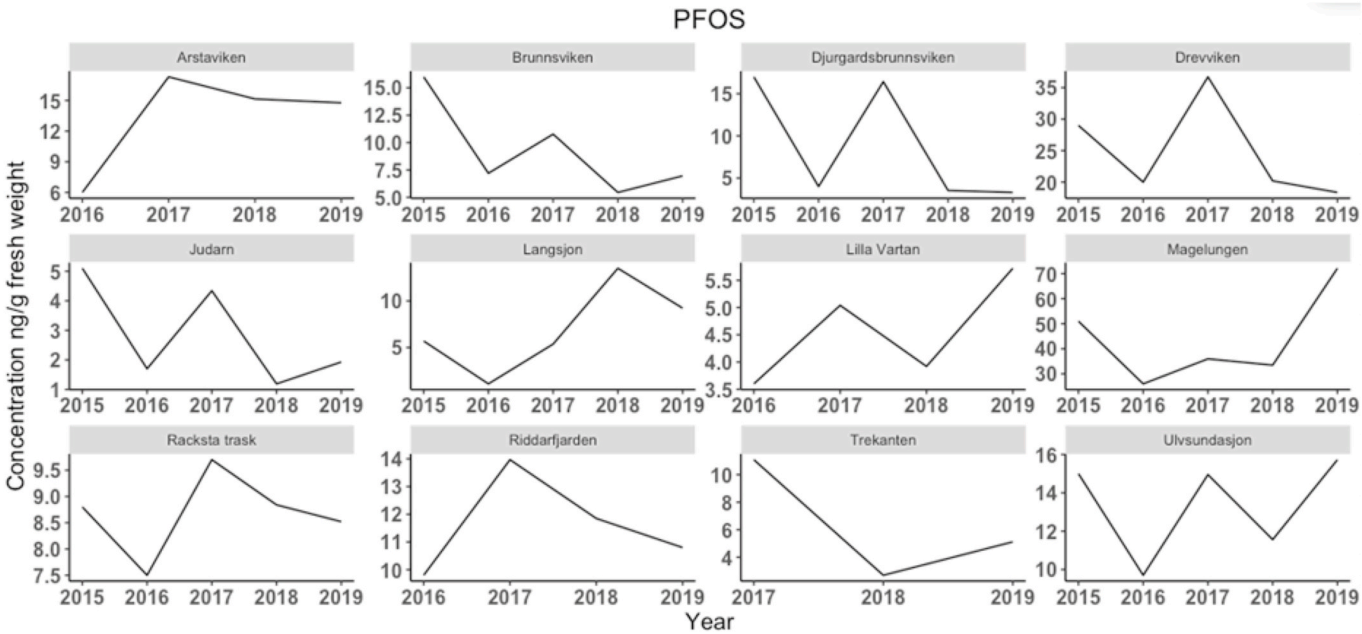

Fig. 4. Time series of mean PFOS concentrations in muscle tissue from edible-sized perches in 12 sites from "Urban Stockholm" (the ones with time series from 2015 to 2019). Each mean value is based on pooled material from 15 individual fishes. Note that the scales differ between lakes, due to differences in recorded concentrations.

3.3. Comparison with a more "traditional" fish contaminant - mercury

Another contaminant, for which the human dietary exposure is also mainly related to the intake of fish and other kinds of seafood, is mercury. While this element has nothing to do with PFOS, the restrictions proposed for it still puts the PFOS situation in perspective. Mercury is a toxic metal that has long been known to accumulate in aquatic food chains, mainly as methylmercury, MeHg (Clarkson, 1995; Lavoie et al., 2013), where negative effects in humans include damage to the kidneys, liver, and to the nervous, immune, reproductive and developmental systems (Bernhoft, 2012). To aid in protecting the public from harmful mercury exposure, EFSA has assesses the maximum dietary intake of MeHg and inorganic mercury to 1.3 and 4.0 μg/kg/*week*, respectively (EFSA, 2012). In addition, maximum mercury concentrations in different food categories have been determined. For the EU member states (Commission Regulation (EC) No 1881/2006), the maximum concentration in fish meat is 0.50 mg/kg fresh weight. Using the most conservative of the two tolerable intake levels (1.3 μg/kg/*week* for MeHg, or 68 μg/kg/*year*), a man of average weight (84 kg) could eat as much as 11 kg of fish with 0.50 mg mercury per kilo before reaching the annual limit. This equals 57 average-sized fish portions (200 g). Still, environmental- and food safety agencies in many countries recommend a restricted intake of freshwater fish as a measure to reduce the intake of mercury in the population. In Sweden, for example, pregnant women are advised to avoid freshwater fish altogether. Since mercury concentrations in Swedish soils and natural waters are relatively high in a global perspective (Salminen, 2005), the mercury contamination of Swedish fish has been much debated. In a report from the Swedish university of agricultural sciences, as much as 50% of the freshwater fishes analysed (20 802 pikes and 5122 perches) were shown to exceed the 0.50 mg/kg maximum concentration (Åkerblom and Johansson, 2008). However, the same report also shows that the spatial variability in fish mercury is low; concentrations very rarely exceed 1 mg/kg. It means that about 30 portions per year can still be consumed before the provisional intake level is reached, even if every single fish contains mercury at the national maximum level. Obviously, one should never aim to reach the maximum intake of detrimental substances, and certainly not via one single type of food. But this comparison points to the safety margins adopted for another, more "traditional" food contaminant and thus also the need for future safety measures (eg advising) to restrict the PFOS exposure among recreational fishermen and their relatives.

4. Conclusion

The majority of the assessments found in the literature on total dietary PFOS exposure address central tendency exposures in the general population, by applying average consumption rates of various foodstuff and average PFOS concentrations in these foods. The rationale for using only mean contaminant concentrations in dietary exposure assessments is that the focus is usually on chronic exposure, where means are more representative than extreme values. For food items that are consumed on a regular basis this makes sense. However, this study gives an example of how food types that are seldom consumed can also play an important role in the dietary intake of certain contaminants. If a specific category of food (such as freshwater fish) contains highly variable concentrations of contaminants (such as PFOS), the use of mean values can be misleading. In addition, the study demonstrates the significance of identifying non-commercial food categories that can play a key role in the dietary intake of certain contaminants among certain subpopulations.

We would like to sum up by emphasising that highly variable PFOS concentrations in fish from different water systems make it difficult to assess a representative level of intake and risk for freshwater fish consumers. The mean PFOS concentrations in edible fish meat from our 78 different sites, for example, varied from 0.3 to 750 μg/kg. Another aspect worth highlighting is that most data on PFOS concentrations in fish, at least in Sweden, comes from analyses of pooled samples. Additional research is needed to better characterise variability within and between different fish species as well as between sites. It appears, however, as if the fishing location is more critical than the kind of fish (pike, perch or pikeperch) that is caught. Based on the three datasets evaluated for the present study, it is implied that the median intake among "normal consumers" (eating freshwater fish 3 times per year) is within 0.03–0.09 μg/kg/yr, and that the intake at the 95th percentile is in the range 0.21–0.26 μg/kg/yr. While not as high as the proposed tolerable intake according to EFSAs draft opinion from 2020, which is

0.42 μg/kg expressed as an annual dose, it is still considerable in relation to the average total dietary PFOS intake, which is about 0.11–1.5 μg/kg/yr (EFSA, 2018). For "high consumers" (eating freshwater fish once a week), however, the intake via freshwater fish consumption will outcompete the annual dietary intake from other sources in the majority of cases. The median "high consumer" intake was estimated to be 0.49–1.2 μg/kg/yr and the P95 intake 3.6–8.0 μg/kg/yr. Although all the analyses of PFOA, PFNA and PFHxS were below the limit of detection, we can conclude that the risk associated with freshwater fish consumption is related (almost) exclusively to the intake of PFOS.

In comparison to more traditional aquatic pollutants, like mercury for example, there is for PFOS no general awareness of the potential risks associated with consumption of food from aquatic environments, and in particular from freshwater. Our results point to an urgency in the need to expand the knowledge of freshwater fish as a source of PFOS exposure, and a need for discussions on intake recommendations.

Author contributions

A Augustsson designed the study, set up the exposure model, analysed the data (with the help of T Lennqvist), and did most of the manuscript writing. The fishing was made by P. Tibblin and CMG Osbeck, and the chemical analyses at IVL were coordinated and run by MA Nguyen, E Westberg and R Vestergren. A Glynn was responsible for retrieving data on fish consumption. All the authors have participated in discussing the results, the story outline/main messages, and have commented on several manuscript versions.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2020.110284.

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