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Poly- and perfluoroalkylated substances (PFASs) in water, sediment and fish muscle tissue from Lake Tana, Ethiopia and implications for human exposure

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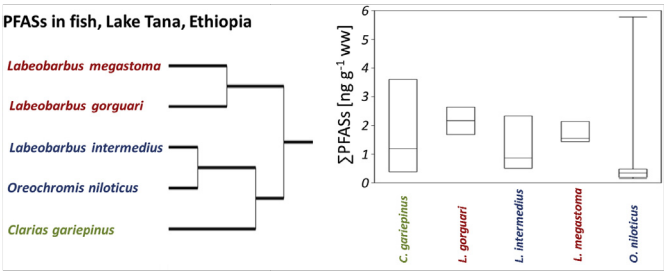
HIGHLIGHTS

- PFCAs were predominant in water (68%), sediment (91%), and fish (71%).
- PFAS levels are higher in piscivorous compared to non-piscivorous fish species.
- PFAS sorption depends on the CF_2 moiety and functional group for sediment or biota.
- PFAS levels were generally low and potential risks to humans are not expected.

GRAPHICAL ABSTRACT



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ABSTRACT

Lake Tana is Ethiopia's largest lake and there are plans to increase the harvest of fish from the lake. The objective of this study was to assess the levels of poly- and perfluoroalkyl substances (PFASs) in different compartments of the lake (water, sediment, and fish muscle tissue), and its implications for human exposure. The results showed higher PFAS concentrations in piscivorous fish species (*Labeobarbus megastoma* and *Labeobarbus gorguari*) than non-piscivorous species (*Labeobarbus intermedius, Oreochromis niloticus* and *Clarias gariepinus*) and also spatial distribution similarities. The ∑PFAS concentrations ranged from 0.073 to 5.6 ng L^{-1} (on average, 2.9 ng L^{-1}) in surface water, 0.22–0.55 ng g^{-1} dry weight (dw) (on average, 0.30 ng g^{-1} dw) in surface sediment, and non-detected to 5.8 ng g^{-1} wet weight (ww) (on average, 1.2 ng g^{-1} ww) in all fish species. The relative risk (RR) indicates that the consumption of fish contaminated with perfluorooctane sulfonate (PFOS) will likely not cause any harmful effects for the Ethiopian fish eating population. However, mixture toxicity of the sum of PFASs, individual fish consumption patterns and increasing fish consumption are important factors to consider in future risk assessments.

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1. Introduction

Poly- and perfluoroalkyl substances (PFASs) are persistent, bioaccumulative and toxic (PBT) substances of concern for environmental and human health (Ahrens and Bundschuh, 2014). PFASs have been widely used, for example, as stain repellents in commercial applications such as textile, paper, and household products over the past 50 years (Buck et al., 2011). Sources of PFASs to the environment include sewage treatment plant (STP) effluents, landfill effluents, and fire training facilities (Ahrens, 2011). STPs are also not effective in removing PFASs from wastewater since PFASs were detected at similar or higher concentrations in STP effluent when compared to the STP influent (Schultz et al., 2006).

An important exposure pathway for human intake of these substances is fish of both freshwater and marine origin (Dórea, 2008). PFASs have been widely investigated in fish of Europe (Berger et al., 2009; Labadie and Chevreuil, 2011), South America (Quinete et al., 2009) and Asia (Yang et al., 2012). There is however, a lack of knowledge of PFASs in African aquatic ecosystems and the potential exposure to humans (Hanssen et al., 2010; Mudumbi et al., 2014; Orata et al., 2008). Lake Tana, the largest lake in Ethiopia and the origin of the Blue Nile, is of interest for investigation of substances with PBT characteristics since production of fish from the lake is predicted to increase. Different hazardous health effects related to PFASs have been reported the aquatic ecosystem and humans, e.g. endocrine-disrupting effects, hepatotoxicity, immunotoxicity and reproductive toxicity (Ahrens and Bundschuh, 2014; Borg et al., 2013; Du et al., 2013). However, little is known about the levels of PFASs in the fish that will be produced.

The aim of this study was to assess *i)* the spatial distribution of PFASs in Lake Tana, Ethiopia, *ii)* the distribution of PFASs in five different fish species, *iii)* solid/liquid partition and bioconcentration behaviour of individual PFASs, and *iv)* the human health risk from PFASs if fish consumption increases. In this study, we collected surface water, sediment and fish muscle tissue samples from Lake Tana in October 2014. The risk of human exposure to perfluorooctane sulfonate (PFOS) was estimated based on a comparison of human fish consumption and pollutant levels in fish from Lake Tana relative to established international guidelines.

2. Material and methods

2.1. Sampling sites

The sampling was performed in Lake Tana, located in the Amhara region in northwestern Ethiopia at around 1800 m above sea level. Lake Tana has an area of 3000–3600 km^2 (84 km long, 66 km wide) with a volume of 28 000 km^3 and a mean depth of 8 m (max 14 m). In total, 61 rivers and streams feed the lake, of which six are perennial and contribute more than 95% of the inflow. The only natural outflow is the Abbay River (Blue Nile River) in the southeastern part of the Bahir Dar Gulf (Ligdi et al., 2010). The 7 sampling sites included *i)* two sites in the south close to wastewater outlets, one near Bahir Dar prison (P) and one near Bahir Dar hospital (H), *ii)* one sample near the Cherechera lake level regulatory weir (C), which is located at the lake's outflow to the Blue Nile River, *iii)* one sample at the Yegashu river inlet in the northeast part of the bay, with very shallow water (depth 2 m) and surrounding agricultural land (Y), iv) one sample in the south close to the forested Zegi Peninsula, where a wood industry was situated (Z), *v)* two samples in the northern part of the lake, one was located near the center of Gorgora Town (G) and the other north of the town, close to the Dirma River (D) (for details see Fig. 1 and Supplementary Table S1).

2.2. Sampling

Fish samples ($n = 30$) were collected between October 11th and 25th, 2014, from five species: *Labeobarbus megastoma* (*L. megastoma*), *Labeobarbus intermedius* (*L. intermedius*), *Labeobarbus gorguari* (*L. gorguari*), *Clarias gariepinus* aka African catfish (*C. gariepinus*) and *Oreochromis niloticus* aka Nile Tilapia (*O. niloticus*) (Supplementary Tables S2 and S3). The three *Labeobarbus* species were chosen due to their habitats and occurrence in the lake as well as their varying feeding habits, i.e. *L. megastoma* and *L. gorguari* are piscivorous and *L. intermedius* is omnivorous (Supplementary Table S4). The herbivorous *O. niloticus* is the most important fish in Ethiopia and stands for 60% of the commercial fishery in the country as well as 30% of the fishery in Lake Tana. The omnivorous *C. gariepinus* is also an important commercial fish as it is fast growing and thus a large protein source (Desta et al., 2007). All fish were weighed and measured (standard length) and directly dissected. The muscle sample was taken above the dorsal line, in between the dorsal and adipose fin. The samples were then carefully wrapped into aluminum foil and packed into a zip lock plastic bag together with an identification card. These samples were then frozen to −20 °C before transportation to Sweden.

Surface water and sediment samples were collected at five sites (i.e., P, H, C, Y, and Z). Surface water samples were collected as grab samples in plastic bottles and sediment samples were collected using a Van Veen Grab Sampler. After sampling, water and sediment samples were stored at −20 °C before transportation to Sweden.

2.3. Chemicals

In total, 26 PFASs were analyzed for four perfluoroalkane sulfonates (PFSAs) (PFBS, PFHxS, PFOS, PFDS), 13 perfluoroalkyl carboxylates (PFCAs) (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFHxDA, PFOcDA), three perfluorooctane sulfonamides (FOSAs) (FOSA, MeFOSA EtFOSA), two perfluorooctane sulfonamidoethanols (FOSEs) (MeFOSE, EtFOSE), three perfluorooctane sulfonamidoacetic acids (FOSAAs) (FOSAA, MeFOSAA, EtFOSAA) (purchased from Wellington Laboratories (ON, Canada)) and one fluorotelomer carboxylate (6:2 FTSA) (purchased from Chiron AS, Norway). In addition, 16 internal standards were used which were spiked before extraction (i.e. $^{13}C_8$-FOSA, d_3-MeFOSAA, d_5-EtFOSAA, d_3-MeFOSA, d_5-EtFOSA, d_7-MeFOSE, d_9-EtFOSE, $^{13}C_4$-PFBA, $^{13}C_2$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_5$-PFNA, $^{13}C_2$-PFDA, $^{13}C_2$-PFUnDA, $^{13}C_2$-PFDoDA, $^{18}O_2$-PFHxS, $^{13}C_4$-PFOS) and one injection standard (InjS) was used ($^{13}C_8$-PFOA) (purchased from Wellington Laboratories (ON, Canada)).

2.4. PFAS analysis

Extraction and analysis of PFASs were performed using standardized and validated methods (Ahrens et al., 2010a, 2015). The muscle tissue samples were homogenized using Ultra-Turrax with a stainless steel probe in a 50 mL PP-tube. The extraction was performed using solid-liquid extraction (SLE). The water samples were filtrated (Whatman™ Glass Microfiber Filters GF/C™, 47 mm diameter, 1.2 μm) and extracted using solid phase extraction (SPE) using Oasis® WAX 6 cc cartridges, 6 cm^3, 500 mg, 60 μm (Waters). The extraction of the sediment samples was performed using SLE. The instrumental analysis was performed by high performance liquid chromatography (HPLC, Agilent Technologies 1200 Series, Palo Alto, CA, USA) with a triple quadrupole mass spectrometer interfaced with an electrospray ionization source in negative-ion mode ((−)ESI-MS/MS, Agilent 6460 Triple Quadrupole System, Palo Alto, CA, USA). Aliquots of 10 μL were injected on a Hypersil

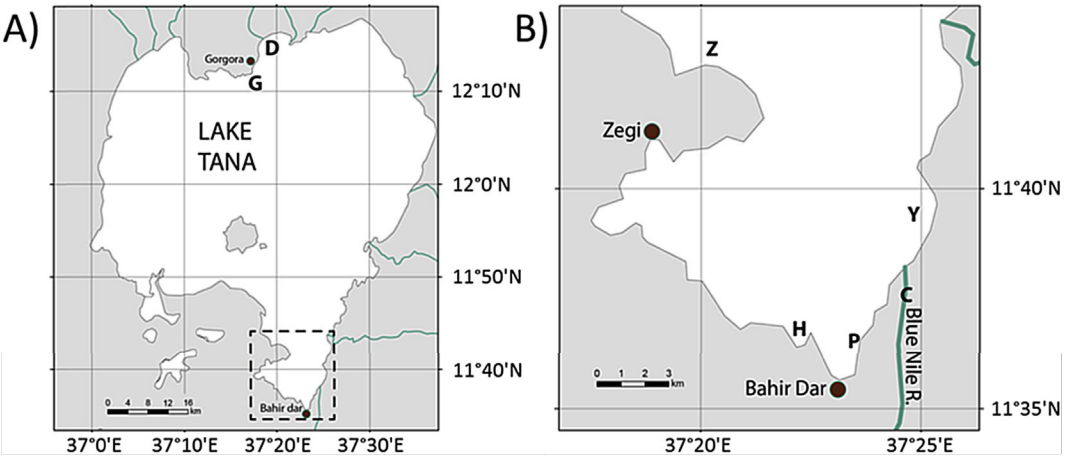

Fig. 1. Sampling sites at Lake Tana (Modified after Dejen et al., 2009 with permission (Dejen et al., 2009)) including map A) near the town of Gorgora (Gorgora (**G**)), and near Dirma River close to the town of Gorgora (Dirma (**D**)), and map B) near the wastewater outlet from Bahir Dar prison and from the entire town (Bahir Dar Prison (**P**)), near the wastewater outlet from the Bahir Dar Hospital (Bahir Dar Hospital (**H**)), near the Cherechera lake level regulatory weir at the outflow from Lake Tana to the Blue Nile River (Cherechera (**C**)), Yegashu river outlet near agricultural land (Yegashu (**Y**)), and forested peninsular with wood industry (Zegi Peninsular (**Z**)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Gold pre-column (10 × 2.1 mm, 5 μm particle size, Thermo Scientific, Waltham, MA, USA) coupled with a Betasil C18 column (50 × 2.1 mm, 5 μm particle size, Thermo Scientific, Waltham, MA, USA) using a gradient of 0.350 *m*L/min Millpore water and methanol (both with 10 mM aqueous ammonium acetate solution (NH_4OAc)). The initial gradient was set at 90/50 (v/v) Millipore water/methanol, then decreased for 3 min to 50/50 Millpore water/methanol and further decreased to 5/95 Millpore water/methanol (hold for 3 min) (total time 20 min). The MS/MS was operated in the multiple-reaction monitoring (MRM) mode at the most sensitive transition from precursor ion to product ion (Supplementary Table S5). The isotope dilution method was used for quantification using a five-point calibration curve (0.5, 2.5, 10, 40, 80 and 400 pg absolute amount injected onto the column). For details of the analytical method see Supplementary Data or elsewhere (Ahrens et al., 2010a, 2015).

2.5. Statistical analysis

Distribution of concentration data did not follow a normal distribution according to Shapiro Wilk W-test (Shapiro and Wilk, 1965), hence normalization to logarithmic values was performed before statistical analysis. To be able to conduct statistical analysis of the analyzed fish samples half of the detection limit was used for PFASs not detected in the fish species (zero-values). Analysis of variance (ANOVA) in EXCEL 2013 was used to determine if mean values between groups (sampling sites or fish species) differed significantly. The level of significance was taken as $p < 0.05$.

2.6. Partitioning

The interaction of sorption and desorption between sediment and water can be described by the partition coefficient (K_d) for individual PFASs.

$$K_d = \frac{c_s}{c_w} \quad (1)$$

where c_s is the adsorbed PFAS on sediment in ng g^{-1} dw and c_w is the mass concentration of PFAS in the aqueous phase in ng mL^{-1}.

Bioconcentration factors (BCF) were estimated in fish muscle tissue for individual PFASs and individual species.

$$BCF = \frac{c_b}{c_w} \quad (2)$$

where c_b is the PFAS concentration in fish in ng g^{-1} ww.

2.7. Human health assessment

To evaluate the uptake of PFASs by humans with varied fish species consumption from Lake Tana, an average daily intake (ADI) [μg g^{-1} d^{-1}] was estimated according to the equation

$$ADI = substance\ concentration \cdot fish\ consumption \quad (3)$$

where the substance concentration can be either for PFAS in ng g^{-1} ww, and the fish consumption in g kg^{-1} body weight $(bw)^{-1}$ d^{-1} (Assuming bw of 60 kg). Then the relative risk (RR) were calculated with the equation

$$Relative\ risk\ (RR) = ADI / Reference\ Dose\ (RfD) \quad (4)$$

where RfD is in μg g^{-1} d^{-1} and a RR value greater than 1 suggests that the average exposure level exceeds the benchmark concentration.

These calculations were done for two scenarios, with consumptions of 0.67 g d^{-1} (national average) and 27 g d^{-1} (Lake Tana average) in 1995 (Breuil, 1995) and in 2016 after an estimated 44% increase in fish consumption in the country (Gordon et al., 2007). Also different country averages were included. The RR for PFASs was only calculated for PFOS (mean = 0.137 ng g^{-1} ww, $n = 30$) since there were no other RfDs available for the other PFASs detected in fish samples. Four RfD were used for PFOS including TDI = 150 ng kg bw^{-1} d^{-1} from the European Food Safety Authority (EFSA) (EFSA, 2008), Rfd = 100 ng kg bw^{-1} d^{-1} from the German Federal Institute for Risk Assessment (BfR) (BFR, 2006), RfD = 30 ng kg bw^{-1} d^{-1} from the Agency for Toxic Substances and Disease Registry (ATSDR) (ATSDR, 2009), and 25 ng kg bw^{-1} d^{-1} from the Environmental Working Group (EWG) (Thayer and Houlihan, 2002).

3. Results and discussion

3.1. Species specific PFAS concentrations

The mean ∑PFAS concentrations of the five evaluated species at site Z were quite similar, but with slightly elevated concentrations for the piscivores *L. gorguari* (2.1 ng g^{-1} ww), followed by the piscivorous *L. megastoma* (1.7 ± 0.38 ng g^{-1} ww) and omnivorous *C. gariepinus* (1.7 ± 1.7 ng g^{-1} ww) and *L. intermedius* (1.2 ± 0.96 ng g^{-1} ww). The lowest ∑PFAS concentrations were in the herbivorous *O. niloticus* (0.85 ± 1.4 ng g^{-1} ww) (Fig. 2). Between the five analyzed fish species, PFCA and PFSA concentrations differed significantly (ANOVA, F = 6.4, p = 0.018). PFDA was the PFCA with the highest concentrations for all species. These were highest in *L. gorguari* (0.93 ± 0.33 ng g^{-1}) and lowest in *L. megastoma* (0.32 ± 0.076 ng g^{-1}) (Fig. 2, Supplementary Table S6). Three species (*L. intermedius, C. garipenius, O. niloticus*) showed higher values of PFBS than of PFOS whilst *L. megastoma and L. gorguari* had higher PFOS concentrations. These results can be compared with another African study for PFOS in *O. niloticus*, where mean values ranged from 1.23 ± 0.19 to 4.89 ± 2.11 ng g^{-1} ww between five sites in Lake Victoria (Orata et al., 2008). To the best of the authors' knowledge, other studies on PFAS concentrations in freshwater fish from Africa are lacking.

The composition profile showed that the dominant PFASs in all species were PFCAs ranging from 54 to 84% (on average, 71%), while PFSAs were just ranging from 16 to 46% (29%) (Fig. 3). PFDA was present in all species independent of trophic level with the highest composition in the species *L. intermedius*, *L. gorguari* and *O. niloticus* with 45%, 43% and 40% of the ∑PFASs, respectively. PFBS was found in low percentages (~0–5%) in piscivores but higher (~20–30%) in nonpiscivores. The opposite was discovered for PFOS and PFUnDA, which were found in higher percentages in piscivores (~25–45% and ~15–25%, respectively) compared to non-piscivores (~0–5% and ~0–10%, respectively). Interestingly, *C. gariepinus* had high percentages of PFBS (~30%), PFDoDA (~30%) and PFDA (~30%), while *L. megastoma* had high percentages of PFOS (~45%) and PFUnDA (~25%). Overall a larger proportion of short-chain PFASs were found in herbivores and omnivores, while longer-chain PFASs were more prevalent in piscivores. This might be explained by the fact that short-chain PFASs dominate in plants (Felizeter et al., 2012), which are consumpted by herbivores and omnivores, whereas longer-chain PFASs have a higher bioaccumulation potential and therefore have higher concentrations in piscivores.

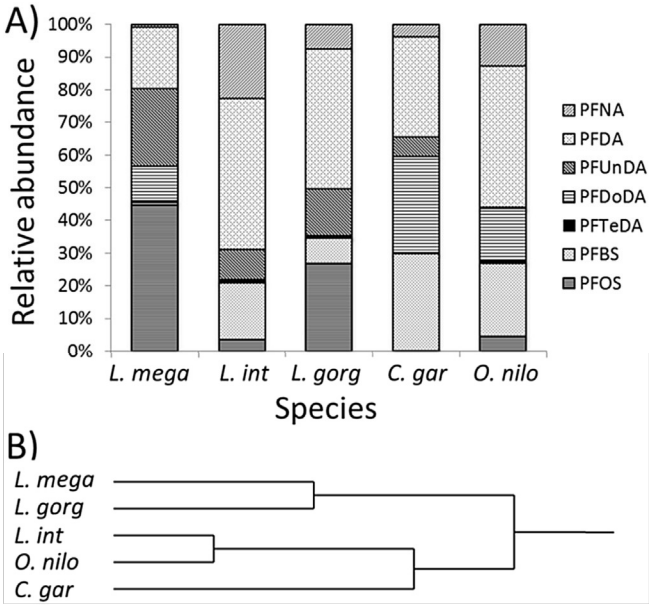

Fig. 3. A) Relative abundance and B) cluster diagram of PFASs in five sampled fish species (*L. megastoma*, *L. intermedius*, *L. gorguari*, *C. gariepinus*, *O. niloticus*) at Lake Tana.

The cluster analysis of the PFAS distribution in fish species showed that *L. intermedius* and *O. niloticus* had the closest relationship between their PFAS composition (Fig. 3). The two piscivores *L. megastoma* and *L. gorguari* also had a close relation but a bit further apart, and *C. gariepinus* was more similar to the non-piscivores than the piscivores. This may imply that the PFAS composition depends on the trophic position of the species.

3.2. Spatial distribution of PFASs in fish, water and sediment

The highest ΣPFAS concentrations in fish were found at the Cherechera weir (site C = 2.1 ± 1.6 ng g^{-1} ww), Yeagashu River (site Y = 1.9 ± 0.99 ng g^{-1} ww) and Zegi peninsular (site Z = 0.97 ± 0.40 ng g^{-1} ww) (Fig. 2, Supplementary Table S7). The sites located close to Bahir Dar (H and P) showed lower levels (ΣPFAS = 0.20 and 0.27 ng g^{-1} ww, respectively). There were significantly higher PFCA concentrations compared to PFSA for *O. niloticus* at the seven sampling sites (ANOVA, F = 21, $p < 0.0001$).

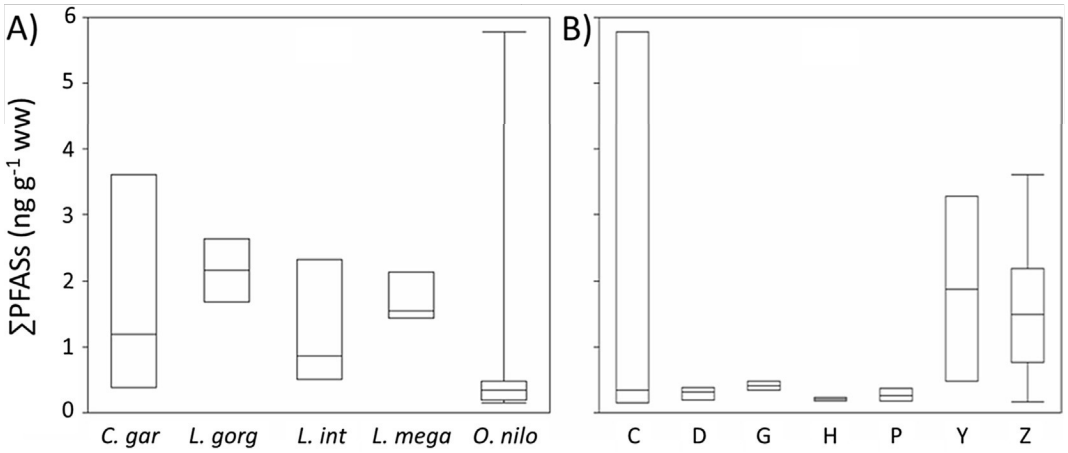

Fig. 2. Box and whisker plots of PFAS concentration for A) individual fish species and B) between sampling sites at Lake Tana. The box-whiskers indicate the maximum (upper error bar), 3rd-quartile (upper box), median (horizontal line), 1st-quartile and minimum values.

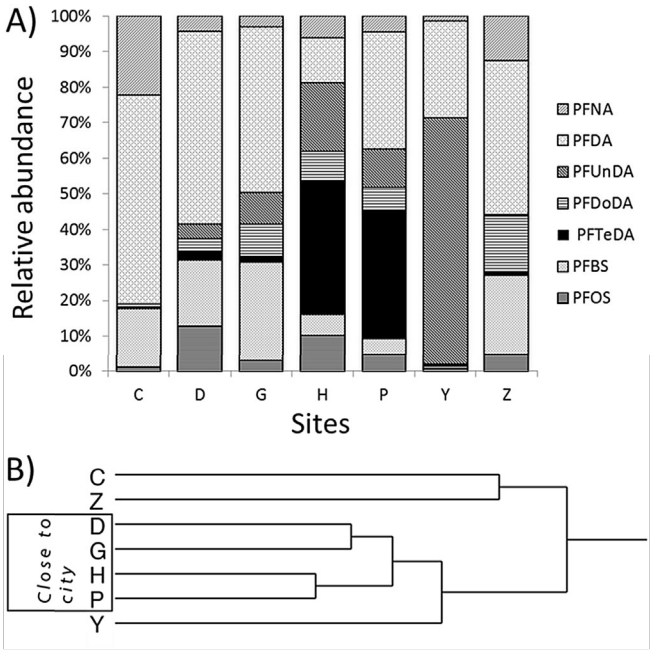

Fig. 4. A) Relative abundance and B) cluster diagram of PFASs in *O. niloticus* within sites (C, D, G, H, P, Y, Z) at Lake Tana. For details of the sampling sites see Fig. 1.

For the PFCAs, PFDA had higher values (on average, 0.44 ± 0.66 ng g^{-1}) in most sites compared to other PFCAs. For the two PFSAs, PFBS was generally found at higher concentrations (on average, 0.16 ± 0.30 ng g^{-1}) compared to PFOS (0.14 ± 0.25 ng g^{-1}) which can be explained by the replacement of PFOS by shorter chained PFASs such as PFBS (Möller et al., 2010). Furthermore PFBS was correlated with three PFCAs (R = 0.71 for PFNA, R = 0.71 for PFDA, R = 0.60 for PFDoDA) (Supplementary Table S8), indicating a different source origin compared to PFOS. Interestingly, high ΣPFAS concentrations were observed in *O. niloticus* at sites C and Y (2.1 and 1.7 ng g^{-1} ww, respectively) and in the piscivores *L. gorguari* and *L. megastoma* at site Z (2.1 and 1.7 ng g^{-1} ww, respectively, Fig. 2). This might mean that the ΣPFAS concentration does not depend on trophic levels, since piscivores and non-piscivores showed the same concentration values, or that the pollutant loading at site C and Y was higher than at site Z.

The relative abundance between all PFASs in *O. niloticus* at the different sites showed that PFDA was the dominant compound with 58%, 55%, 48% and 41% of the ∑PFASs at sites C, D, G and Z, respectively (Fig. 4). Site Y contained 70% of PFUnDA and 25% of PFDA. At sites H and P, PFTeDA was the prominent compound at ~30% but barely seen at any other site indicating that the wastewater outlets from Bahir Dar near site H and P are a point source for this compound. At site Y, PFUnDA was predominant with 70%, which is close to the agricultural lands. Cluster analysis for the distribution of PFASs between sampling sites showed that there were close similarities between sites D, G close to Gorgora and H, P close to Bahir Dar (Fig. 4). Site C and Z have a weak relationship, and are far from similar to the other sites. Site Y is closer to the near-city sites than sites C and Z.

In surface water, the highest ΣPFAS concentrations were found at site P with 5.6 ng L^{-1}, indicating that the wastewater outlets from Bahir Dar near site P is a point source for PFASs (Supplementary Table S9). The site C near the Cherechera weir showed the lowest ΣPFAS concentration (0.073 ng L^{-1}). PFCAs were dominant in surface water (68% of the ∑PFASs) with mainly the shorter chain PFBA (46%) and PFHxA (30%). In surface sediment, the ΣPFAS concentrations showed similar contamination levels at all sites ranging between 0.22 ng g^{-1} dry weight (dw) (site H) and 0.50 ng g^{-1} dw (site Z) (Supplementary Table S10). PFCAs were the predominant PFASs in sediment with, on average, 91% of the ∑PFASs, whereas of the PFSAs only PFOS was detected (9%).

3.3. Partitioning

K_d and BCF values were calculated for individual PFASs (Table 1). In general, the perfluoroalkyl chain length and functional group had an influence on the K_d values. For the PFCAs, the log K_d increased from 0.93 for PFBA (with a perfluorocarbon chain length of C_3) to 2.6 ± 0.32 for PFUnDA (C_{10}). Thus, for each additional CF_2 moiety the log K_d values increase by ~0.25 log units. These findings are in agreement with previous reported log K_d values (Ahrens et al., 2010b, 2015; Higgins and Luthy, 2006).

For the log BCF, no clear trend was observed for the influence perfluoroalkyl chain length and functional group on the bioaccumulation potential of PFASs. For the PFCAs, the log BCF increased by 0.1 log units for each additional CF_2 moiety from, on average, 3.3 for PFNA (C_8) to 3.6 for PFUnDA (C_{10}), whereas PFOS (C_8) had an average log BCF of 3.2. In addition, no species specific log BCF was observed for the five investigated fish species. Overall, the log BCF values calculated in this study were in the same range as previous reported (Martin et al., 2003).

3.4. Human health assessment

The RR ratios for PFOS were orders of magnitude below 1 (Table 2), indicating that the current PFOS concentrations in fish are not harmful for human health. Neither people with fish consumption according to the national or Lake Tana average seem to be at any contamination risk in 1995 (Breuil, 1995) and 2016 (after an estimated 44% increase fish consumption) (Gordon et al., 2007). The largest ratio ($2.2 * 10^{-4}$) was found for the EWG reference dose in the production areas of Lake Tana for 2016. Overall it seems that fish consumption is not a risk for PFOS.

Table 1
Solid/liquid partition coefficients (K_d) and bioconcentration factors (BCF) for individual PFASs.[a]

	PFBA	PFHxA	PFNA	PFDA	PFUnDA	PFHxS	PFOS
log K_d[b]	0.93	2.5 ± 0.59	NC	2.1 ± 0.20	2.6 ± 0.32	NC	2.3 ± 0.47
log BCF[c]							
O. niloticus	NC	NC	3.4 ± 0.77	3.4 ± 0.46	3.0 ± 0.68	NC	2.6 ± 0.20
L. megastoma	NC	NC	NC	3.4 ± 0.19	3.7 ± 0.11	NC	3.7 ± 0.04
L. gorguari	NC	NC	3.4	3.8 ± 0.35	3.9	NC	3.6 ± 0.025
L. intermedius	NC	NC	3.4 ± 0.56	3.8 ± 0.19	3.6	NC	2.9
C. gariepinus	NC	NC	3.2	3.8 ± 0.07	3.6	NC	NC

[a] NC = not calculable, because the compound was not detected in sediment, water or/and biota.
[b] $K_d = c_s/c_w$, where c_s is the adsorbed PFAS on sediment in ng g^{-1} dw and c_w is the mass concentration of PFAS in the aqueous phase in ng mL^{-1}.
[c] BCF = c_b/c_w, where c_b is the PFAS concentration in fish in ng g^{-1} ww.

Table 2
PFOS relative risk (RR) for the national and Lake Tana fish consumption using scenarios for 1995 and 2016 (estimated).

		National		Lake Tana	
		1995	2016 (estimated)	1995	2016 (estimated)
PFOS	EFSA[a]	$6.1 * 10^{-7}$	$8.8 * 10^{-7}$	$2.5 * 10^{-5}$	$3.6 * 10^{-5}$
	BfR[b]	$9.2 * 10^{-7}$	$1.3 * 10^{-6}$	$3.8 * 10^{-5}$	$5.4 * 10^{-5}$
	ATSDR[c]	$3.1 * 10^{-6}$	$4.4 * 10^{-6}$	$1.3 * 10^{-6}$	$1.8 * 10^{-5}$
	EWG[d]	$3.7 * 10^{-6}$	$5.3 * 10^{-6}$	$1.5 * 10^{-4}$	$2.2 * 10^{-4}$

[a] European Food Safety Authority (EFSA). PFOS TDI = 150 ng kg bw^{-1} d^{-1} (EFSA, 2008).
[b] The German Federal Institute for Risk Assessment (BfR). PFOS Rfd = 100 ng kg bw^{-1} d^{-1} (BFR, 2006).
[c] Agency for Toxic Substances and Disease Registry (ATSDR). PFOS RfD = 30 ng kg bw^{-1} d^{-1}(ATSDR, 2009).
[d] Environmental Working Group (EWG) in the US. PFOS RfD = 25 ng kg bw^{-1} d^{-1} (Thayer and Houlihan, 2002).

4. Conclusions

The Ethiopian population as a whole have had very low fish consumption for centuries, so a large increase might affect the pollutant loading in the population and consequences regarding the uptake of PFASs needs to be considered. The maximum concentration in fish was 5.8 ng g^{-1} ww for ∑PFAS (on average for all species = 1.2 ng g^{-1} ww). However, the human health assessment using RR indicate that concentrations of PFOS in Lake Tana fish were of no harm to the Ethiopian population, even with an increased fish consumption in 2016. However, mixture toxicity of the sum of PFASs, the possibility that certain species diets, high fish consumption by certain groups, (e.g. fishing communities), and collection of fish near pollution sources might create situations with intake levels harmful to humans. It is important to gain a better understanding of the spatial distribution of PFASs in Africa like Lake Tana since only very few studies were performed in this region. Ultimately, further research is required regarding tropical aquatic systems and bioaccumulation of toxic pollutants to provide adequate information about the risks associated with fish consumption in Africa.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2016.09.007.

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