Spatial profiles of perfluoroalkyl substances and mercury in fish from northern Lake Victoria, East Africa.

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Abstract

There is an acute deficit of data on per- and polyfluoroalkyl substances (PFASs) and mercury (Hg) in the open waters of Lake Victoria, East Africa, relative to nearshore areas. We analyzed stable isotopes ($\delta^{15}N$ and $\delta^{13}C$), PFASs and Hg in Nile Perch and Nile Tilapia muscle and liver samples from nearshore and open lake locations from the Ugandan part of the lake. The $\delta^{15}N$ values of Nile Perch muscle indicated a higher trophic level for samples from the open lake than from nearshore locations. Averages of ∑PFAS concentrations in Nile Perch muscle and liver (0.44 and 1.75 ng g^{-1} ww, respectively) were significantly higher than in Nile Tilapia (0.24 and 0.50 ng g^{-1} ww, respectively). ∑PFAS concentrations in muscle of open lake Nile Perch were significantly higher than for nearshore samples. A similar observation was made for total mercury concentrations in muscle ($THg_{\_Muscle}$) of Nile Perch. THg was dominated by methyl mercury ($MeHg^+$, 22-124 ng/g ww) and mercuric mercury (Hg^{2+}, <MDL-29 ng/g ww) in Nile Perch muscle. Strong correlation between $MeHg^+$ and some PFASs (e.g. PFOS: $r = 0.704$, $P = 0.016$) suggested similar exposure routes or factors. Estimated daily intake values of PFOS and Hg were below international limits.

Key finding:

PFAS and $MeHg^+$ profiles suggest a location-dependent and trophic-influenced spatial difference in bioaccumulation with open lake samples posing the higher human exposure risk than nearshore ones.

1. Introduction

Lake Victoria (1° 0′ S, 33° 0′ E, 68,000 km^2 surface area and about 195,000 km^2 drainage basin), a major regional economical resource in Eastern Africa, is experiencing rapid urbanization at a much faster rate than the urban development of supporting infrastructure, including waste management systems (GRID-Arendal and Lake_Victoria_Basin_Commission, 2017). Additionally, the lake's catchment has high agricultural and mining activities. Consequently, it is exposed to various chemical pollutants from these activities as well as urban domestic and industrial processes. The chemical pollutants which were of particular interest to this study are per- and polyfluoroalkyl substances (PFASs) and mercury (Hg).

PFASs are a huge class of anthropogenic organic substances, including perfluoroalkyl acids (PFAAs) such as perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkane sulfonic acids (PFSAs), whose surfactant properties have enabled their wide use in industry, such as in textiles, fire-fighting foams, hydraulic fluids and upholstery, among others (Zushi et al., 2012). Some PFASs may also be transformed via natural environmental processes to PFAAs (Ellis et al., 2004; Parsons et al., 2008). PFAAs are ubiquitous in the environment (Houde et al., 2011; Jian et al., 2017) although there exist limited environmental measurements from Africa. PFAAs are persistent in the environment, and long chain homologues bioaccumulate in animal tissues and biomagnify in food webs (Kannan et al., 2005; Houde et al., 2006; Müller et al., 2011; Pan et al., 2014). Fish and sea food have been reported to be a major source for human exposure to PFASs (Berger et al., 2009; Vestergren et al., 2012; Domingo and Nadal, 2017). The major possible input pathways for PFASs to fresh water lakes include atmospheric deposition, (Ahrens et al., 2010; Benskin et al., 2011) wastewater treatment plant (WWTP) effluents, (Boulanger et al., 2005; Xiao et al., 2012) storm water drainage (Kim and Kannan, 2007) and catchment release via riverine discharge

(McLachlan et al., 2007; Scott et al., 2009; Filipovic et al., 2015). These exposure routes are all very relevant for Lake Victoria.

Unlike the synthetic PFASs, Hg is a naturally occurring global element that exists in various species, including elemental mercury, organic mercury (largely methyl mercury – $MeHg^{+}$), mercurous mercury (Hg_2^{2+}) and mercuric mercury (Hg^{2+}). Elemental mercury and Hg^{2+} are predominantly dispersed via atmospheric transport, contaminating land and surface waters through precipitation and dry deposition(Driscoll et al., 2013). Upon deposition in sediment or hypoxic zones of the aquatic bodies, anaerobic bacteria transform Hg^{2+} to $MeHg^{+}$ (Ranchou-Peyruse et al., 2009). This $MeHg^{+}$, in addition to the atmospherically deposited and surface run-off fractions, is then able to bioaccumulate in fish food webs, which becomes the major exposure pathway to humans (Driscoll et al., 2013). Mercury in all forms is toxic to human health and can impair virtually any organ (Bernhoft, 2012). As of 2015 about 38% of the global Hg emissions could be attributed to artisanal and small scale gold mining activities, which constitute the major emission source for Sub-Saharan Africa and Latin America (UNEP, 2019). The Tanzanian section (and to a lesser extent the Kenyan and Ugandan ones) of the Lake Victoria catchment is home to artisanal and small scale gold mining. Indeed, the highest levels of total mercury (THg) measured to date in Lake Victoria, were found in samples collected close to the southern shoreline of the lake, near the gold mines (Campbell et al., 2003a). The highest concentrations of THg in fish samples from the lake have been observed in Nile Perch, possibly due to biomagnification of $MeHg^{+}$, since Nile Perch occupies the highest trophic level for fish in the lake(Campbell et al., 2003b).

More studies of Hg than of PFASs have been conducted on Lake Victoria. Very few studies on the Kenyan part of the lake (which is about 6% by surface area) have reported PFAAs in water (Orata et al., 2009), sediment (Orata et al., 2011), WWTP effluent (Chirikona et al., 2015) and fish

84 samples (Orata et al., 2008). One study has also reported PFASs in various matrices from wetlands
85 in the northern catchment of the lake (Dalahmeh et al., 2018). This limited coverage underscores
86 the need for studies which investigate the state of PFAS contamination in the wider lake and its
87 catchment.

88 The increasing urbanization in the Lake Victoria catchment, the acute deficit of PFAS data for the
89 lake and the knowledge gap on the occurrence and fate of Hg in the open lake motivated this study.
90 Aided by $\delta^{13}C$ and $\delta^{15}N$ stable isotope analysis to assess possible influence of dietary habits, we
91 investigated spatial profiles of PFAAs and Hg ($MeHg^{+}$, Hg^{2+} and THg) in the muscle and liver of
92 fish samples collected from nearshore and open lake areas from the Ugandan part of Lake Victoria.
93 Nile Tilapia is known to inhabit nearshore areas of the lake and its diet, reportedly, is composed
94 of algae, plant material, insects, juvenile fish, invertebrates, bivalves and detritus (Njiru et al.,
95 2004). Nile Perch is piscivorous (Agembe et al., 2019) and widely distributed all over the lake
96 wherever there is enough oxygen (Kitchell et al., 1997). Consequently, any differences between
97 the results of stable isotope analysis of open lake and nearshore samples could be a function of the
98 fish's residence period and differences in diets at either location. The lake's seasonal stratification
99 (Talling, 1957; MacIntyre et al., 2014) could, potentially, be a major factor in such spatial fish diet
100 differences. During the stratification period, the deep waters tend towards anoxic conditions,
101 thereby leading to upward migration of deep-feeding fish, such as haplochromine species and a
102 population increase in anoxia-tolerant benthic feeders, such as shrimps (Budeba and Cowx, 2007;
103 Njiru et al., 2012). The latter become a major component of the fish-diet in the mixed period, hence
104 a longer food chain in the deep waters (open lake). This could be reflected in the $\delta^{15}N$ profile which
105 is a better indicator of the food web structure, due to its stepwise enrichment into the higher trophic

106 level (Minagawa and Wada, 1984; Peterson and Fry, 1987; Cabana and Rasmussen, 1994) than
107 $\delta^{13}C$.

108 This study aimed at obtaining insight into the levels, spatial profiles of PFASs and Hg in fish
109 samples obtained from nearshore and open lake locations of the Ugandan waters of Lake Victoria
110 as well as human dietary exposure. We hypothesized that (i) the fish samples from nearshore areas
111 were more likely to have higher concentrations due to proximity to source areas, especially the
112 urban centers, than samples from open lake areas; (ii) there is no appreciable risk of dietary
113 exposure to PFASs and Hg residues from the consumption of fish from Lake Victoria in light of
114 internationally recognized tolerable intake limits.

115

116 **2. Materials and methods**

117 **2.1 Sampling**

118 Nile Perch (*Lates niloticus*, N=25) and Nile Tilapia (*Oreochromis niloticus*, N=11) samples (Table
119 S1 in the Supporting Information) were collected in 2017 using a trawling net during a lake-wide
120 hydro-acoustic survey of Lake Victoria under the auspices of the Lake Victoria Fisheries
121 Organization (LVFO). The samples were collected from nearshore (Sites 7, 8 and 12; total N = 15
122 Nile Perch and 11 Nile Tilapia samples) and open lake locations (Sites 1 and 5; total N =10 Nile
123 Perch samples) (Figure. 1). Site 8 is located in Napoleon Gulf which is the gateway of water
124 discharge from Lake Victoria to River Nile, the only outflowing water channel. Additional fresh
125 Nile Tilapia samples (N=6) caught in and around the Napoleon Gulf of the lake (using gill nets)
126 were purchased from fishermen at Masese Landing site in Jinja, Uganda. The fish weight, length
127 and sex were recorded (Table S1). Fish muscle, taken near the dorsal fin, and liver from all fish

were stored frozen at -20°C at the Department of Chemistry, Makerere University in Kampala, Uganda. These samples were later shipped via courier to the Helmholtz Centre for Environmental Research – UFZ in Leipzig, Germany for analysis.

Figure 1. Map of Lake Victoria showing the sampling sites and date of sampling. Samples were collected by trawling. Sites 1 and 5 are characterized as "open lake" locations. Sites 7, 8 and 12 are characterized as "nearshore" locations.

2.2 Chemicals and reagents

High purity organic solvents and ammonium acetate (purity >99.9% and UPLC/MS grade, respectively, BioSolve Chemicals, Valkenswaard, The Netherlands) and PFAS standards (native compound mix PFAC-MXB and single compound FOSA-I; isotope mass-labelled internal standard mix MPFAC-MXA and single compounds M3-PFPeA, M4-PFHpA, M8-FOSA; isotope mass-labelled recovery standards M8-PFOA, M8-PFOS; all from Wellington Laboratories, Ontario, Canada) were used for analysis. All gases used were of high purity >99.999%.

2.3 Sample preparation and analysis

2.3.1 Stable isotope analysis of $\delta^{13}C$ and $\delta^{15}N$

Simultaneous $\delta^{13}C$- and $\delta^{15}N$-analysis of the samples was achieved with a combination system involving an auto-sampler (AS200, CE Instruments, Italy), a EuroEA3000 elemental analyzer system (HEKAtech, Germany), a gas chromatograph (HEKAtech, Germany) and an isotope ratio mass spectrometer (MAT253 IRMS, Thermo Fisher Scientific, Germany) (Renpenning et al., 2017). A subsample of the muscle or liver tissue was freeze dried, ground into a fine powder and approximately 1 mg of this sample was weighted into a tin foil capsule (3.5 mm × 5 mm, HEKAtech, Germany). The sample was combusted with a 10 mL oxygen pulse at 1050 °C in a combustion reactor filled with wolfram oxide and silver-cobalt oxide. The product gases, including nitrogen and carbon dioxide, were swept with helium carrier gas (gas flow 80 mL/min) through a copper-packed reduction furnace at 650 °C and then dried over phosphorus pentoxide. The gases were separated isothermally at 70 °C on a GC-column and transferred into the IRMS via a ConFlo IV open split system. Isotope ratios were expressed as delta notation ($\delta^{13}C$ or $\delta^{15}N$) in parts per thousand (‰) relative to the standards VPDB (Vienna Pee Dee Belemnite) for carbon or AIR for nitrogen according to equation:

$\delta^{13}C$ or $\delta^{15}N$ [‰] = ($R_{sample}/R_{standard}$ -1), where

R_{sample} and $R_{standard}$ are the isotopic ratio ($^{13}C/^{12}C$ or $^{15}N/^{14}N$) of the sample or the corresponding ratio of the standard, respectively.

2.3.2 PFAS analysis

The sample preparation procedure was an adaptation of a method published elsewhere (Berger et al., 2009). Briefly, approximately 1 g of the homogenized muscle or liver tissue was spiked with 1 ng of each internal standard (ISTD) in 50 µL methanolic solution. The sample was extracted with acetonitrile over an ultrasonic bath. The extract was concentrated to about 1 mL, treated with graphitized carbon (25 mg ENVI-Carb) and 50 µL of glacial acetic acid before centrifugation (10 min at 10 000 rpm). A volume of 0.5 mL of the supernatant was mixed with 0.5 mL of a 4 mM ammonium acetate solution in water and spiked with 50 µL of a recovery standard (RSTD) solution (20 pg/µL M8PFOA and 20 pg/µL M8PFOS in methanol). The mixture was filtered (0.2 µm) and transferred into a polypropylene vial for analysis.

The samples were analyzed on an Aquity Ultra Performance Liquid Chromatograph, equipped with a trapping column upstream of the injector and coupled to a Xevo TQ-S Tandem Mass Spectrometer (Waters, Eschborn, Germany). The target analytes and the associated multiple reaction monitoring (MRM) transitions are given in Tables S2.

2.3.3 Mercury (Hg) analysis

Separate total Hg (THg) and speciation analyses were done for selected fish and liver samples. THg was measured using a Direct Mercury Analyzer (DMA-80 evo, Milestone Srl, Sorisole, Italy) in accordance with the guidelines from the German Environmental Specimen Bank (Rüdel et al., 2011). The DMA method involves complete combustion of the sample followed by catalytic

conversion of gaseous products to release elemental Hg. This Hg is then amalgamated with gold and analyzed by atomic absorption spectroscopy. Approximately 10 - 50 mg of the fish samples, depending on Hg content, was analyzed in triplicates.

The procedure for Hg speciation had minor modifications to a published method (Arroyo-Abad et al., 2016) combining solid phase micro-extraction (SPME) and gas chromatography (GC) coupled to inductively coupled plasma mass spectrometry (ICPMS 7500, Agilent Technologies, Santa Clara, USA). About 200 mg of homogenized muscle or liver was heated in 5 mL of 3.6 M nitric acid solution in a water bath at 40 °C for 16 hours. The mixture was allowed to cool, centrifuged at 6 °C (15 min at 4950 rpm). An acetic acid/acetate buffer solution was used to adjust an aliquot of the supernatant to pH 4.5-5, before derivatization with tetrapropylborate (0.1 g mL^{-1}). Hg species were enriched by SPME onto a 100 µm polydimethylsiloxane (PDMS) fibre for 30 min at 30 °C with agitation. The Hg species were desorbed from the PDMS fibre directly in the injector port of the GC for 2 min at 220 °C.

2.4 Quantification and quality control

2.4.1 Stable isotope analysis

Normalization of measured isotope compositions to international isotope-delta scales was done by analyzing reference materials in the same way as the samples. For normalization of the $\delta^{13}C$ values, IAEA-CH6 (10.45 ‰), IAEA-CH7 (32.15 ‰) and IAEA-CH3 (-24.72 ‰) were used (Coplen et al., 2006). The normalization of $\delta^{15}N$ values was done with AS-3 (-0.35 ‰) and AS-11 (11.0‰) (Kornexl et al., 1999). All stable isotope analyses were done in triplicates.

2.4.2 PFAS analysis

For the analysis of PFASs, procedural blanks (total n=7) and a certified reference material IRMM-427 (European Commission, Institute for Reference Materials and Measurements, Geel, Belgium)

were included in each batch of samples. Multi-level calibration standards (13-point between 0.01 and 2.5 ng mL^{-1}) were used for quantification of PFASs. For all PFASs only the linear isomer was analyzed (branched isomers of PFOS and FOSA were generally below the quantification limit). For analytes detected in the procedural blanks, method detection and quantification limits were calculated as the average plus three or ten times the standard deviation of the blank concentrations, respectively. Where analytes were not detected in the blanks, a signal-to-noise (S/N) ratio in the chromatograms of sample extracts of three and ten was applied to estimate method detection and quantification limits, respectively (Table S2). Samples were spiked with stable isotope-labelled internal standards for quantification using the internal standard method and for the determination of apparent recoveries (combination of sample preparation recoveries and matrix effects). The measured concentrations of target analytes in IRMM-427 were within 20 % of the reference concentrations, except for PFTeDA (Bias + RSD = 31 %). In the analytical method, the internal standard M-PFDoDA was used for estimation of PFTeDA concentration. It is plausible that the difference in matrix effects on these two compounds may account, at least in part, to this discrepancy. Average apparent recoveries of the internal standards in the Nile Perch and Nile Tilapia muscle were generally better than for liver samples (Figure S1). The apparent recoveries were generally good with the exception of M8-FOSA (in all matrices) and PFOA (in Nile Tilapia Liver), likely due to strong matrix effects.

2.4.3 Hg analysis

For the DMA method, a mercury ICP standard (1000 mg/L; Merck KGaA, Darmstadt, Germany) was used for reference. An 18-point calibration in the concentration range of 0.1 – 100 ng was used for calibration (R^2 = 0.999). The THg MDL was 3 ng/g ww. Certified reference materials IAEA-407 and IAEA-436 were also analyzed and the trueness was 93% and 116%, respectively.

For the speciation, 5-point calibrations in the 2.9 – 34.5 ng range, were performed using an Hg ICP standard (1000 mg/L; Merck KGaA, Darmstadt, Germany) and Alfa Aesar MeHgCl (10 mg/L prepared from solid; Thermo Fisher GmbH, Kandel, Germany) standard for the measurement of Hg^{2+} (R^2=0.981) and $MeHg^{+}$ (R^2 = 0.999), respectively. The MDLs for $MeHg^{+}$ and Hg^{2+} were 5 and 3 ng/g ww, respectively. The trueness for THg and $MeHg^{+}$ for IAEA-407 (106% and 93%, respectively) as well as for IAEA-436 (91 % and 87 %, respectively) was good.

2.5 Statistical analysis

Descriptive statistics and outlier detection were done using Microsoft Excel. Outliers were identified as values outside the interquartile range (IQR) using an IQR multiplier of 1.5 ($Xi > Q3 + 1.5*IQR$ or $X_i < Q1 – 1.5*IQR$, where Xi is the outlier data point). During statistical analysis, "MDL/2" values were assigned to non-detects. Pearson correlations and probabilities and other statistical analyses were done using SigmaPlot 13.0. The t-test and One Way Analysis of Variance (ANOVA) were performed for site comparison for stable isotope, PFAS and Hg data. The Shapiro-Wilk test, the Brown-Forsythe test and Holm-Sidak test were used to test normality, equal variance and pairwise multiple site comparison at an overall significance level of 0.05. If a dataset did not pass the normality or equal variance test, Kruskal-Wallis One Way ANOVA on ranks was additionally performed. For sampling sites which had female and male fish samples, the t-test was used to assess influence of sex on the concentration data. During the t-test, if the dataset failed the normality or equal variance test, the Mann-Whitney rank sum test was performed.

2.6 Dietary intake estimation

Current fish consumption data for Uganda is not readily available. The fish consumption in Uganda in 2013 was reported to be 12.5 kg/person/year (Obiero et al., 2019). This value was used to estimate the total daily intake (TDI) of ∑PFASs and PFOS via consumption of Nile perch and Nile

Tilapia from the sampling locations mentioned above. Since fillet is the most often eaten part of fish, only concentrations of muscle tissue were considered for the TDI calculation. An average 70 kg body weight (BW) for an adult and a 360-day year are assumed.

3. Results and discussion

3.1 Stable isotope analysis

The results of stable isotope analysis ($\delta^{15}N$ and $\delta^{13}C$) of selected fish muscle and liver samples from the open lake and nearshore areas (Figures 2 and S2) provided an insight into the trophic structure of the samples.

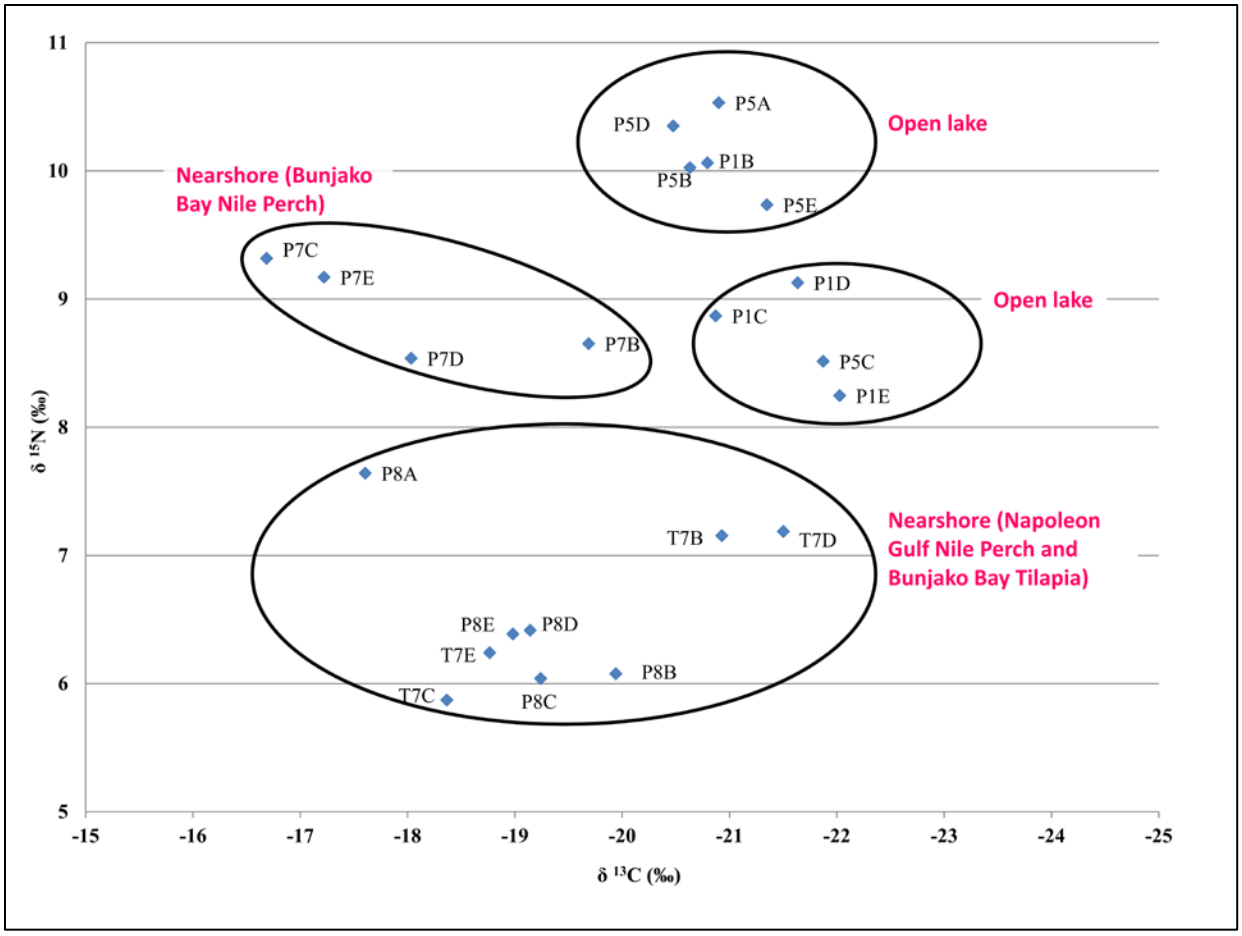

Figure 2. Trophic structure of fish muscle samples (Nile Perch and Nile Tilapia) from near shore and open lake areas of Lake Victoria (for sample codes see Table S1).

Fish muscle from Nile Perch samples obtained from the open lake had significantly higher $\delta^{15}N$ and $\delta^{13}C$ values than for muscle of Nile perch from Napoleon Gulf (Site 8), a nearshore location, with a $\delta^{15}N$ difference of about 3 – 4 ‰ between these locations (Two-tailed *P*-value <0.001). Trophic transfer enrichment factors of $\delta^{15}N$ in and around this range have been reported for aquatic organisms (Deniro and Epstein, 1981; Hansson et al., 1997; Post, 2002). These results, therefore, suggest that the open lake Nile Perch samples were at a higher trophic level than the ones from the Napoleon Gulf despite their young age (assuming that Nile Perch total length (Table S1) is proportional to age and given that Nile Perch can grow up to 2 m, and weigh up to 200 kg) (Ogutu-

Ohwayo, 2004). It is possible that the two locations have different food web structures, leading to location-dependent dietary shifts by the Nile Perch, associated with longer food chains in the open lake. Dietary shifts for Nile Perch in the Lake Victoria have been previously reported elsewhere (Kishe-Machumu et al., 2012; Cornelissen et al., 2018; Agembe et al., 2019). The $\delta^{15}N$ and $\delta^{13}C$ values for Nile Perch from Napoleon Gulf were similar to previous measurements for the same location (Campbell et al., 2003b). The $\delta^{15}N$ values for fish muscle samples from Bunjako Bay (Site 7, a nearshore area) were similar to those from the open lake, but the $\delta^{13}C$ values for the two locations significantly differed (Two-tailed *P*-value <0.001) by 2 - 5 ‰, with the samples from the former site having more ^{13}C enrichment. Assuming a $\delta^{13}C$ trophic-enrichment factor of 0.39 - 2 ‰ (DeNiro and Epstein, 1978; Post, 2002), these results suggest an ecologically significant difference in sources of dietary carbon for the two locations, suggesting a food chain richer in, for example, carbondioxide-deprived primary consumers or producers at Site 7 (Bootsma et al., 1996).

3.2 Per- and polyfluorinated alkyl substances (PFASs)

The concentrations of PFASs in fish muscle and liver are summarized in Figure 3 and Tables S3 - S5.

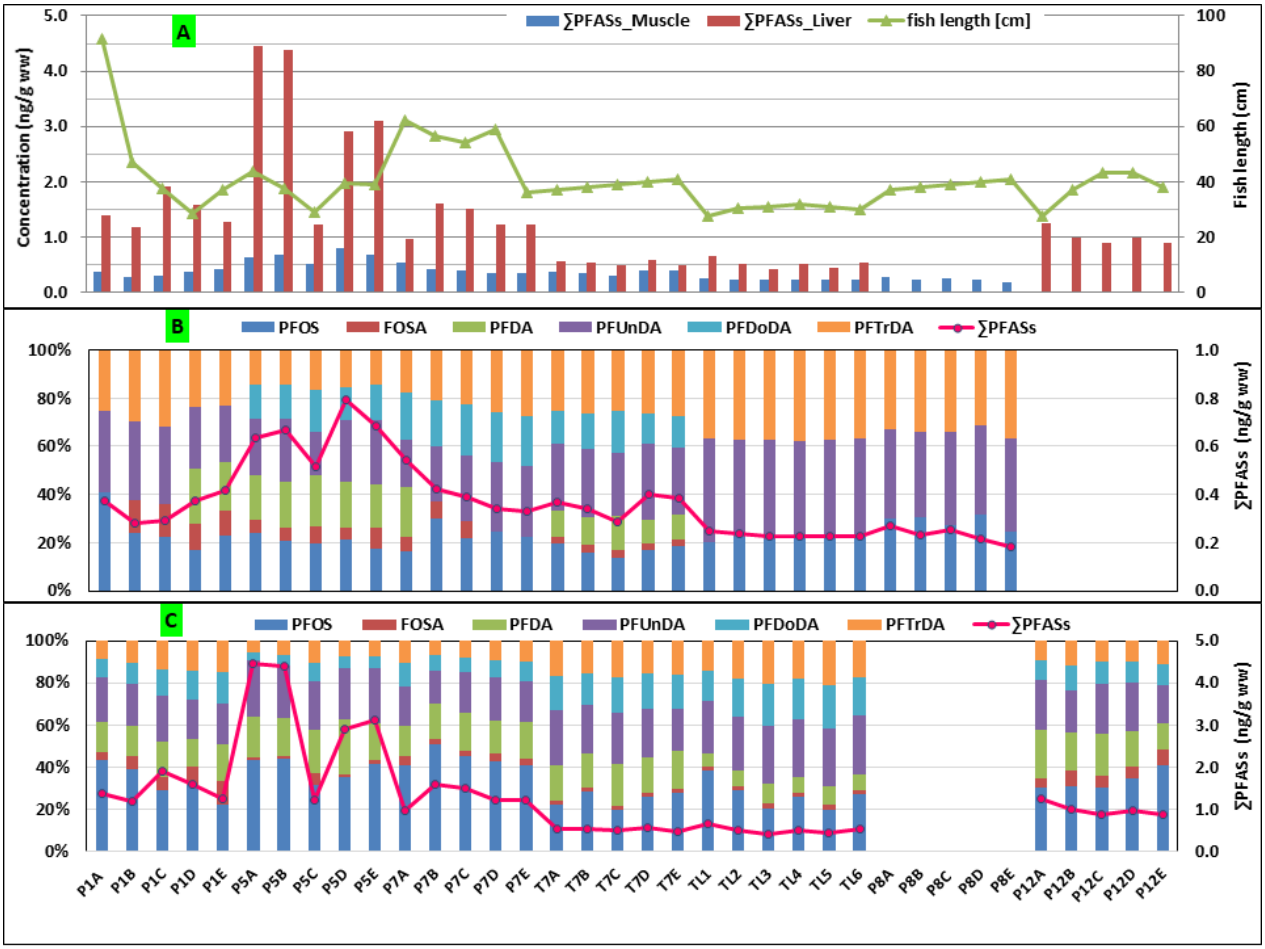

Fig 3. **A:** Concentrations of ∑PFASs in the fish muscle and liver samples from Lake Victoria (see Table S1 for details about the fish codes used). ∑PFASs is the sum of concentrations of PFOS, FOSA, PFDA, PFUnDA, PFDoDA and PFTrDA, which were detected above MQLs. **B:** Profiles of individual PFASs in muscle samples. **C:** Profiles of PFASs in liver samples. Note that for Locations 8 and 12, only muscle and liver samples, respectively, were analyzed for PFASs.

The PFAS concentrations in fish muscle decreased in the order PFOS > PFDA = PFUnDA > PFDoDA = PFTrDA > FOSA while the order for the concentrations in the liver tissue was PFOS > PFUnDA > PFDA > PFTrDA > PFDoDA > FOSA. Apart from these, all the other target analytes indicated in Table S2 were either non detectable or below their MQLs. This distribution, in which PFOS and the long chain PFCAs (≥ C9) are quantifiable in fish tissue, but not the shorter chain PFAAs, has been reported elsewhere (Martin et al., 2004; Berger et al., 2009; Yoo et al., 2009; Fang et al., 2014; Babut et al., 2017). This is expected since the shorter chain PFAAs are highly

mobile in the aqueous phase and will be more readily excreted than the longer chain PFAAs. These studies, however, reported higher concentrations than the ones measured in the present study, probably due to relatively lower consumption of PFAS-containing consumer products and, therefore, less local emissions in the Lake Victoria region. The concentration levels of PFOS (in muscle) reported in the present study are also lower than the ones previously reported for Nile Perch and Nile Tilapia samples from Winam Gulf in the Kenyan Part of Lake Victoria (Orata et al., 2008). Winam Gulf is an embayment heavily impacted by urban emissions (mainly waste water and storm water drainage from Kisumu town) and discharge from the Nyando River. Lower levels in nearshore samples from the Ugandan part of the Lake may suggest relatively less fish exposure to local PFAA emissions. In the open lake, it is possible that dilution and partitioning into the sediment could lead to a general reduction in fish exposure to PFAAs. Elsewhere on the continent, there are very limited studies of PFASs in fish muscle. Ahrens et al. (2016) measured PFASs in fish muscle samples from Lake Tana, Ethiopia. All sampling site-specific PFOS average concentrations in our Nile Perch samples (0.067 – 0.136 ng/g ww) were higher than the ones for fish from Lake Tana (<0.025 – 0.046 ng/g ww). However, for our Nile Tilapia samples, average PFOS concentrations (0.06 and 0.05 ng/g ww for Site 7 and Masese landing site, respectively) were similar to the one for Nile Tilapia samples from Lake Tana (0.045 ng/g ww). All the other average site-specific concentrations of PFASs in our fish muscle samples were within the range of the levels detected in Lake Tana samples. Higher concentrations of PFASs were reported for fish muscle samples from the Vaal River in South Africa (e.g. <0.12 – 45.7 for PFOS, <0.71 – 1.9 for PFDA and <0.07- 0.3 for PFDoDA, all ng/g ww) (Groffen et al., 2018). This may be due to more local PFAS emissions to the river's catchment than in the Lake Victoria Basin. Mwakalapa et al. (2018) did not find any PFASs in farmed and wild fish (mullet and milkfish) in Tanzanian coastal

areas while Guillette et al. (2020) reported much higher PFAS concentrations in blood plasma of Striped Bass from the Cape Fear River, South Africa. Fish muscle or liver were not investigated in this study, but the high plasma PFAS concentrations speak to relatively higher local exposure of the Striped Bass to these contaminants than in our Nile Perch and Nile Tilapia samples.

In general, the concentrations of the predominant PFAAs in this study come close to measurements in fish samples from remote environments that are less impacted by urban/human emissions of these compounds, such as arctic char from some Canadian Arctic lakes (Lescord et al., 2015). It is a plausible assumption that the Lake Victoria Basin has, generally, a low importation and/or usage of PFAS-containing consumer products, but with increasing urbanization and inefficient waste management systems, (Okot-Okumu, 2012) PFAS emissions into the lake and subsequent accumulation in fish could rise significantly in future.

The concentrations of individual detected PFAAs in the fish muscle (range: <MQL – 0.20 and <MQL - 0.13 ng/g ww for Nile Perch and Nile Tilapia, respectively) was generally lower than in liver samples (range: 0.04 - 1.95 and <MQL – 0.25 ng/g ww for Nile Perch and Nile Tilapia, respectively). On average, PFUnDA slightly edged PFOS in the muscle (by about 10 %) but the latter was about twice as high as the former in the liver. Whereas the concentrations of ∑PFASs in Nile Tilapia liver were equal to approximately double those of muscle, ∑PFASs in Nile Perch liver were 2-7 times as high as in muscle. Exposure studies of fish elsewhere have reported preferential uptake and longer half-lives of PFASs in liver (Falk et al., 2015) while a PFAS–protein interaction, specifically albumin and fatty acid binding protein (FABP), has been suggested as a major factor in bioconcentration of PFASs in fish liver (Ng and Hungerbühler, 2013, 2014). This is a plausible explanation of the higher PFAS levels in liver than in muscle.

345 The highest ∑PFASs were measured in fish samples collected from the open lake as opposed to
346 the ones from nearshore areas. Statistical site-specific comparison of PFAS concentration data of
347 Nile Perch muscle samples indicated a significant difference between Sites 5 and 8 (P = 0.030),
348 but not for other sites and not for liver samples. Additionally, there was no significant difference
349 between the Nile Tilapia muscle and liver sample sets from nearshore sites (Site 7 and Masese
350 Landing site). The significant difference between Sites 5 and 8 mentioned above is likely due to
351 trophic magnification of PFASs in samples from Site 5. The ∑PFAS profiles were neither
352 significantly correlated to fish-length nor to fish-weight (r < 0.2). Only each of Sites 1, 7 and 8 had
353 both female and male samples (Table S1), but there was no significant sex-based differences in
354 concentrations of PFAS at any of the three sites (P = 0.818, 0.394 and 0.334, respectively). There
355 was a significant Pearson correlation between $\delta^{15}N$ and ∑PFASs (r = 0.733 and 0.693 in Nile Perch
356 muscle and liver, respectively; P<0.001) but not between $\delta^{13}C$ and ∑PFAS profiles. The
357 correlations between isotope profiles and the individual PFASs (only for cases where all isotope-
358 analyzed samples had >MQL PFAS values) were good for $\delta^{15}N$ (range of r = 0.640 - 0.818, all P
359 values < 0.006) for both Nile Perch muscle and liver samples but not for $\delta^{13}C$. These correlations
360 likely indicated influence of trophic magnification on the ∑PFAS concentration profile in fish
361 samples. However, it is worth noting that measured concentrations may include direct
362 bioconcentration of PFASs from the water, for example, across the gills.

363 **3.3 Mercury (Hg)**

364 The results of mercury measurements in fish muscle and a few of the liver samples are shown in
365 Table S6 and (only for muscle) in Figure 4. The concentration of total mercury measured using a
366 Direct Mercury Analyzer ($THg_{\_DMA}$) was in the range of 24.1 – 161 ng/g ww in Nile Perch muscle
367 (n=20) and 3.4 – 13.2 ng/g ww in Nile Tilapia muscle (n=10). As was observed with the PFAAs,

368 the concentration of THg in the open lake Nile Perch samples was generally over 2-3 times higher
369 than the one in nearshore Nile Perch. Statistical site-specific comparison of THg concentration
370 data for Nile Perch muscle samples indicated a significant difference between the open lake Site 5
371 and the nearshore Sites 7 and 8 (P = 0.008 and 0.007, respectively). There was no significant
372 difference between the nearshore sites (Site 7 and Masese Landing site) for THg in Nile Tilapia
373 samples. These observations also indicated more trophic magnification of Hg in the open lake
374 samples, likely due to $MeHg^{+}$ which is bioaccumulative and dominated the Hg profile (see Fig. 4
375 and Hg speciation results below). For the whole set of analyzed fish, $THg_{\_DMA}$ had weak Pearson
376 correlation with fish length and weight (r = 0.389 and 0.354, respectively), a sign that age is not
377 the major influencing factor. Also, there was no significant sex-based differences in the $THg_{\_DMA}$
378 concentrations for Sites 1, 7 and 8 which had both male and female sexes represented (P = 0.800,
379 0.445 and 0.079, respectively). Very few (n=4) $THg_{\_DMA}$ measurements on liver samples were
380 made and the results may not be conclusive, but the THg concentration ratio between liver and
381 muscle of 1.3 – 2.3 for the analysed samples seemed to indicated higher general abundance in the
382 liver than in muscle. Studies of freshwater fish liver tissues have reported the existence of several
383 Hg-binding proteins that could even serve as biomarkers of mercury contamination (Vieira et al.,
384 2017). If such proteins were to be present in the Nile Perch liver samples, that could explain, at
385 least in part, the elevated concentrations of Hg in the liver samples compared to muscle.

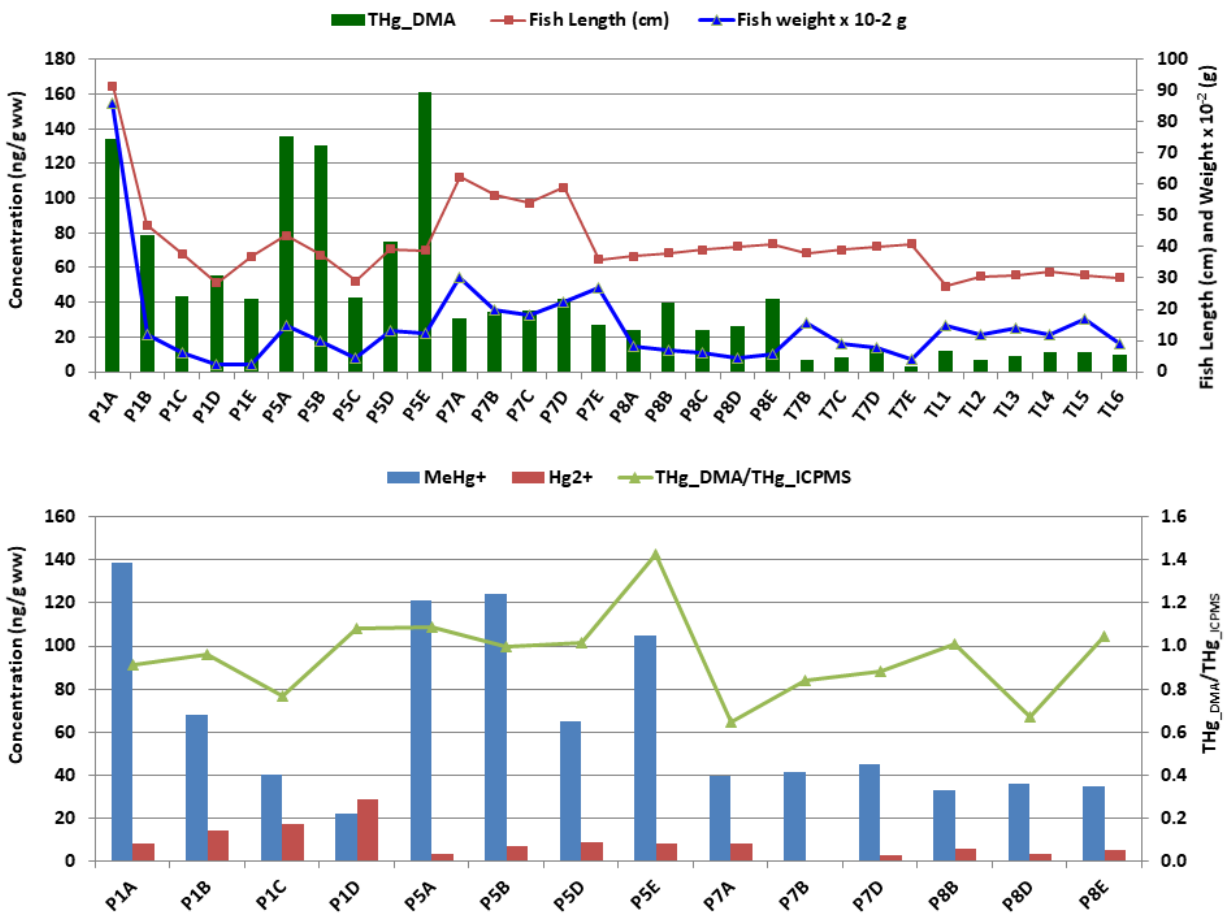

Figure 4. Profiles of total mercury concentrations in fish muscle samples in light of the parent fish length and fish weight profiles (top chart) and mercury speciation profiles in Nile Perch samples from open lake and nearshore areas (bottom chart). See Table S1 for details about the fish codes used.

Mercury speciation was done on 14 Nile Perch muscle samples from the open lake and nearshore areas (Fig. 4). The average (and standard deviation) ratio of $THg_{\_DMA}$ to the total mercury measured using ICPMS ($THg_{\_ICPMS}$ = sum of concentrations of detected Hg species) was 0.95 (0.2), indicating excellent agreement between the two analytical procedures. The THg in all fish except one were dominated by $MeHg^{+}$ which was about 2-35 times and 5-13 times higher than Hg^{2+} in the open lake and nearshore samples, respectively. The $MeHg^{+}$ profile had moderate Pearson correlation with the $\delta^{15}N$ profile ($r = 0.660$, $P < 0.03$) (Table S7), supporting a trophic magnification pathway in the open lake fish. There was strong positive correlation between $MeHg^{+}$

399 and PFOS as well as between $MeHg^{+}$ and PFUnDA in the Nile Perch (r = 0.704 and 0.701,
400 respectively, P-values <0.02), further underscoring the role of biomagnification in their profiles.
401 Deep-water anoxia (Verschuren et al., 2002) in the open lake could favor more formation of
402 $MeHg^{+}$ and its incorporation into the base of the Nile Perch food chain than in oxic nearshore
403 areas. The relatively higher concentrations of Hg^{2+} at Site 1 of the open lake may indicate more
404 oxic conditions here than at Site 5 (Fig. 4). Such habitat-influenced mercury variation in Nile Perch
405 has been suggested elsewhere (Hanna et al., 2016).

406 The THg concentrations measured in Nile Tilapia fish samples in our study (3.4 – 13.2 ng/g ww;
407 Figure S3 and Table S5) are within the same range as previously measured in similar samples from
408 Ugandan nearshore locations (1.7 – 59.7 ng/g ww) (Campbell et al., 2003a). The Nile Perch
409 samples from nearshore areas in our study (24.1 – 42.1 ng/g ww) had generally lower $THg_{\_DMA}$
410 values than previously measured from Napoleon and Thruston Bays on the Ugandan shoreline
411 (36.4 – 252 ng/g ww) as well as in samples collected near Bugaia Island in Northern Lake Victoria
412 (19.6 – 156 ng/g ww) (Campbell et al., 2003a).

413 Hg measurements have not yet been reported for fish samples from the open lake. However, the
414 average concentration from Site 5 of the open lake (109 ng/g ww) is comparable to the levels
415 previously reported for fish samples from nearshore areas, including the ones proximate to the
416 gold mines of Tanzania, some of which had higher average fish weights (Campbell et al., 2003a).
417 Similar values (average THg concentration: 90.5 – 137 ng/g ww) have also been reported for larger
418 fish groups from Napoleon Gulf (Campbell et al., 2003a). This is contrary to the hypothesis that
419 open lake fish samples would have lower concentrations than nearshore areas.

The THg values in this study were compared with some of the most recently reported measurements elsewhere globally (Table S8). On average, the THg levels in our Nile Perch samples were in the range of average THg levels observed in fresh fish from Laurentian Great Lakes (Visha et al., 2018), but below the estimated North American continental averages (Eagles-Smith et al., 2016; Willacker et al., 2020). The THg levels observed in our Uganda samples are also below the levels observed in mining-impacted areas elsewhere, such as in South Africa (Verhaert et al., 2019), Colombia (Marrugo-Negrete et al., 2018), and French Guiana (Gentès et al., 2019). THg levels in our samples are comparable, and in some cases higher, than levels observed in Europe (Kalisinska et al., 2017; Łuczyńska et al., 2017; Fliedner et al., 2018; Kucukosmanoglu and Filazi, 2020; Rüdel et al., 2020). The average THg concentration in our Nile Tilapia samples was lower than the one reported for the same fish species from Mangala Lake in Egypt (Sallam et al., 2019) but similar to THg concentrations in other fish species in the same lake and in both farmed and wild fish from Tanzanian coastal areas (Mwakalapa et al., 2019).

3.4 Dietary exposure risk to humans

Figure S4 shows the estimated sampling site-specific average daily dietary intake of PFASs. Open lake Nile Perch would present the highest total PFAA and PFOS dietary exposures at 0.32 ng kg^{-1} bw day^{-1} and 0.07 ng kg^{-1} bw d^{-1} (about 20 % of total PFAA exposure), respectively. This is about twice the highest estimated exposure via Nile Tilapia consumption (0.13 ng kg^{-1} bw day^{-1} and 0.03 ng kg-1 bw day^{-1} for total PFAA and PFOS, respectively, at Site 7). The estimated TDI for PFOS is lower than the recommended tolerable daily intake (TDI) values, such as Europe's recent recommendation of 13 ng kg^{-1} bw $week^{-1}$ (about 1.9 ng kg^{-1} bw day^{-1}) (European Food Safety Authority, 2018).

442 The United States Environmental Protection Agency (US EPA) derived an epidemiological study-
443 informed reference dose RfD (0.1 μg/kg/day) for $MeHg^+$, which they defined as "a daily intake
444 that is likely to be without appreciable risk of deleterious effects during a lifetime". The estimated
445 daily intake of $MeHg^+$ based on concentrations measured in Nile Perch muscle in this study (0.8 –
446 4.8 μg/kg/day) exceeded the US EPA RfD. It is noteworthy that the THg concentrations measured
447 in our study were, nevertheless, below the WHO and European Commission Hg trade limit of 500
448 µg/kg in fish (EC, 2006).

449

450 **4. Conclusions**

451 In this study, we have seen indications that fish in the open lake locations, though moved away
452 from shoreline point sources, are more likely to biomagnify PFASs and Hg than similar age fish
453 from nearshore areas. This is contrary to our hypothesis (i) in the *Introduction* section above. We
454 have also observed that based on Ugandan average fish consumption rate and on the concentrations
455 measured in this study, the dietary intake to PFASs was lower than international tolerable daily
456 intake limits. This is consistent with hypothesis (ii) in the *Introduction* section above. Although
457 the potential dietary human exposure to Hg is higher for the open lake fish than the nearshore fish,
458 the measured concentrations are still below the international trade limits. It is noteworthy that
459 previous mercury measurements have shown that Nile Perch larger than 10 kg was more likely to
460 bioaccumulate THg to levels higher than the trade limit of 500 µg/kg (Campbell et al., 2003a).
461 This is a reasonable expectation given the long life spans and large sizes to which Nile Perch can
462 grow. The biomagnification of mercury, in Nile Perch, to high concentrations in the open lake may
463 pose a health risk to the lake's island fishing communities due to chronic mercury exposure from

above-average fish consumption. In light of the nutritional benefits of fish consumption, especially as a source of protein and omega-3 fatty acids, there is need for increased vigilance in reducing anthropogenic release of these and other contaminants into the lake to ensure long term chemical safety of the fish.

The findings in the current study underscore the need for further investigation of trophic magnification of chemical contaminants in Lake Victoria fish, especially Nile Perch, in the open lake waters. This is because there could be significant spatial variation in benthic ecologies (and in fish trophic structures) within the open lake due to factors such as (i) varying bathymetry of the lake, which influences focusing of sediment-bound chemicals; (ii) spatial differences in benthic oxygen distribution, which affects chemical fate processes and prevalence of benthic anaerobic/aerobic life forms and fish distribution and (iii) differences in proximity or exposure to inflowing riverine and urban effluent discharges.

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Supporting information

Sample and sampling site descriptions; the target analytes, their quantifier MRM transitions and detection limits; recoveries of PFAS internal standards, trophic structure of fish liver samples from Lake Victoria, detailed concentration data for the analyzed samples; Pearson correlation coefficients for mercury vs $\delta^{15}N$, $\delta^{13}C$ and $\sum$PFASs in Nile Perch muscle samples; graphical comparison of THg data in this study with previously reported data for Lake Victoria and estimated daily intake of PFASs.

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726

Supporting information

Spatial profiles of perfluoroalkyl substances and mercury in fish from northern Lake Victoria, East Africa.

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Number of Tables **6**

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Table S1. Sampling locations, sample codes, sex, species, parent fish total length and weight.

Sampling location	Fish species	Fish code (Muscle)	Fish code (Liver)	Fish length [cm]	Fish weight [g]	Fish sex
Site 1 – Open lake (Off Bukasa Island)	Nile Perch	P1A_M	P1A_L	91.5	8600	f
	Nile Perch	P1B_M	P1B_L	47.0	1200	m
	Nile Perch	P1C_M	P1C_L	37.7	590	f
	Nile Perch	P1D_M	P1D_L	28.5	240	m
	Nile Perch	P1E_M	P1E_L	36.9	235	m
Site 5 - Open lake	Nile Perch	P5A_M	P5A_L	43.7	1500	f
	Nile Perch	P5B_M	P5B_L	37.3	1000	f
	Nile Perch	P5C_M	P5C_L	29.0	450	f
	Nile Perch	P5D_M	P5D_L	39.2	1300	f
	Nile Perch	P5E_M	P5E_L	38.9	1240	f
Site 7 - Nearshore (Bunjako Bay)	Nile Perch	P7A_M	P7A_L	62.4	3000	m
	Nile Perch	P7B_M	P7B_L	56.5	2000	m
	Nile Perch	P7C_M	P7C_L	54.0	1800	m
	Nile Perch	P7D_M	P7D_L	59.0	2230	f
	Nile Perch	P7E_M	P7E_L	36.0	2700	f
	Tilapia	T7A_M	T7A_L	37.0	1500	m
	Tilapia	T7B_M	T7B_L	38.0	1550	m
	Tilapia	T7C_M	T7C_L	39.0	900	m
	Tilapia	T7D_M	T7D_L	40.0	780	m
	Tilapia	T7E_M	T7E_L	41.0	400	m
Site 8 - Nearshore (Napoleon Gulf)	Nile Perch	P8A_M	P8A_L	37.0	820	m
	Nile Perch	P8B_M	P8B_L	38.0	700	f
	Nile Perch	P8C_M	P8C_L	39.0	600	m
	Nile Perch	P8D_M	P8D_L	40.0	450	f
	Nile Perch	P8E_M	P8E_L	41.0	580	f
Site 12 - Nearshore (Bugaia)	Nile Perch	P12A_M	P12A_L	27.5	700	f
	Nile Perch	P12B_M	P12B_L	36.8	1200	f
	Nile Perch	P12C_M	P12C_L	43.0	1000	f
	Nile Perch	P12D_M	P12D_L	43.4	1000	f
	Nile Perch	P12E_M	P12E_L	38.0	700	f
Purchased from fishermen at Masese Landing Site, Jinja - Caught in Napoleaon Gulf*	Tilapia	TL1_M	TL1_L	27.5	1500	m
	Tilapia	TL2_M	TL2_L	30.5	1200	m
	Tilapia	TL3_M	TL3_L	31.0	1400	m
	Tilapia	TL4_M	TL4_L	32.0	1200	m
	Tilapia	TL5_M	TL5_L	31.0	1700	m
	Tilapia	TL6_M	TL6_L	30.0	900	m

*The location of fishing activity (Napoleon Gulf) was self-reported by the fishermen.

Table S2. Target analyte PFASs, their quantifier MRM transitions and method detection (MDL) and quantification limits (MQL).

Analyte	Abbreviation	Quantifier MRM transition (m/z>m/z)	MDL (ng/g ww)	MQL (ng/g ww)
Perfluorobutanesulfonic acid	PFBS	299>80	0.02	0.03
Perfluorohexanesulfonic acid	PFHxS	399>80	0.02	0.05
Perfluorooctanesulfonic acid	PFOS	499>80	0.02	0.02
Perfluorooctanesulfonamide	FOSA	498>78	0.02	0.02
Perfluorodecanesulfonic acid	PFDS	599>99	0.02	0.02
Perfluorobutanoic acid	PFBA	213>169	0.78	2.03
Perfluoropentanoicacid	PFPeA	263>219	0.11	0.21
Perfluorohexanoic acid	PFHxA	313>269	0.13	0.24
Perfluoroheptanoic acid	PFHpA	363>319	0.12	0.18
Perfluorooctanoic acid	PFOA	413>369	0.14	0.27
Perfluorononanoic acid	PFNA	463>419	0.04	0.12
Perfluorodecanoic acid	PFDA	513>469	0.05	0.08
Perfluoroundecanoic acid	PFUnDA	563>519	0.04	0.08
Perfluorododecanoic acid	PFDoDA	613>569	0.05	0.1
Perfluorotridecanoic acid	PFTrDA	663>619	0.06	0.08
Perfluorotetradecanoic acid	PFTeDA	713>669	0.06	0.17
Perfluorohexadecanoic acid	PFHxDA	813>769	0.03	0.08
Perfluorooctadecanoic acid	PFODA	913>869	0.02	0.09

Figure S1. Average apparent recoveries (including matrix effects) of PFAS internal standards for both Nile Perch (NP) and Nile Tilapia (NT). *Error bars represent standard deviation.*

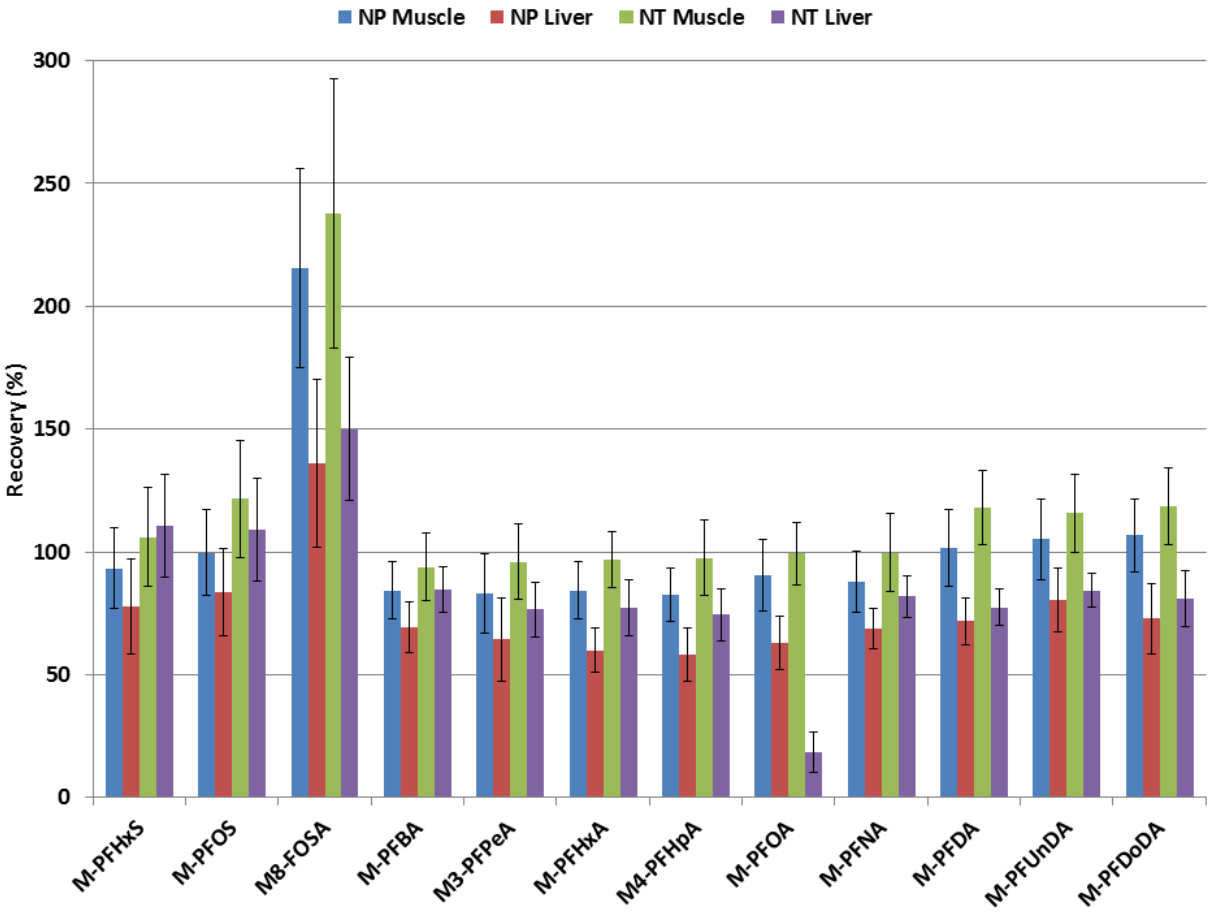


Figure S2. Trophic structure of fish liver samples from near shore and open lake areas of Lake Victoria.

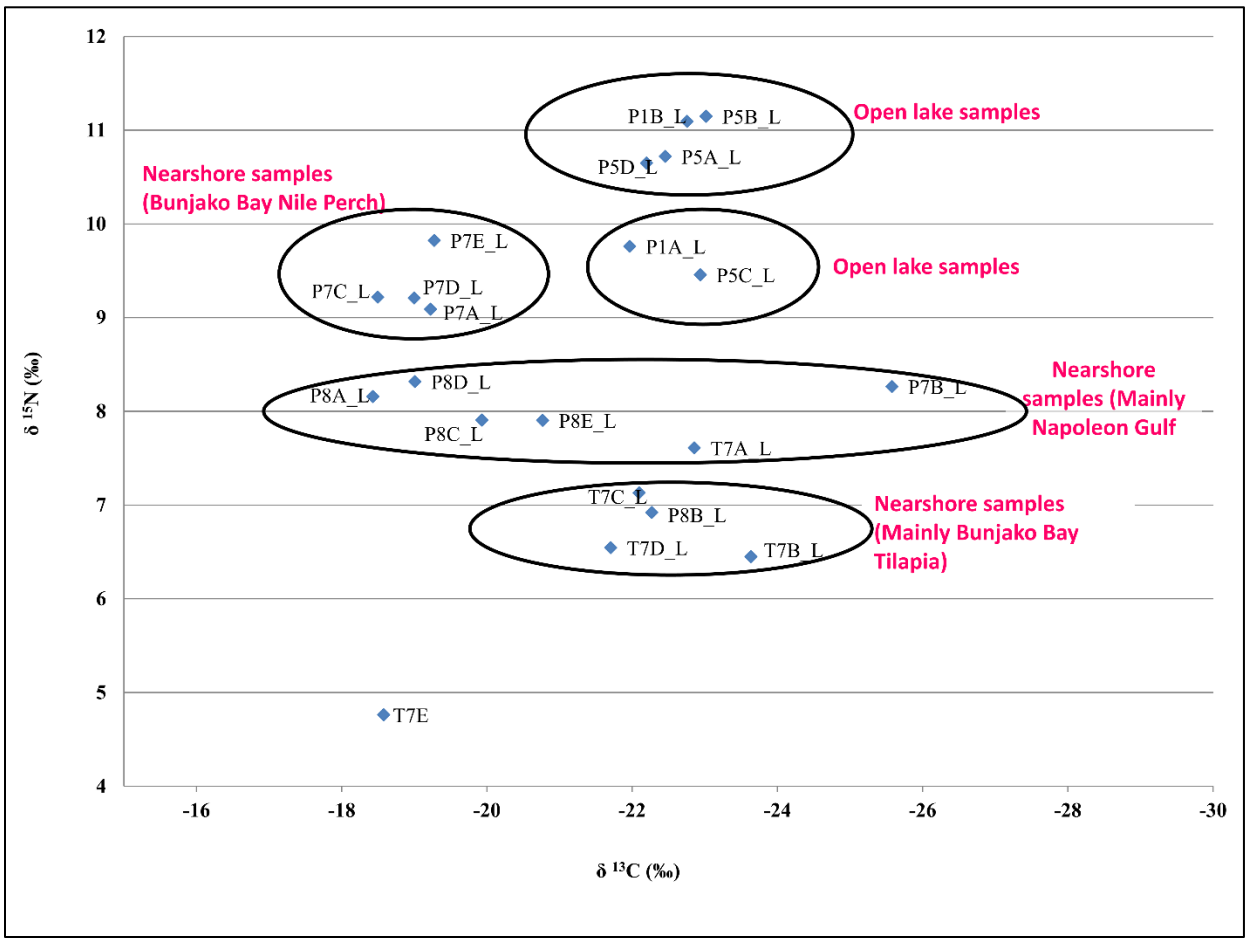


Table S3. Concentrations (ng g^{-1} ww) of PFASs in Nile Perch muscle and liver tissues.

	Fish Muscle							Fish Liver					
	PFOS	**FOSA**	**PFDA**	**PFUnDA**	**PFDoDA**	**PFTrDA**		**PFOS**	**FOSA**	**PFDA**	**PFUnDA**	**PFDoDA**	**PFTrDA**
P1A_M	0.15	<MQL	<MQL	0.12	<MQL	0.09	**P1A_L**	0.60	0.05	0.20	0.30	0.12	0.12
P1B_M	0.07	0.04	<MQL	0.09	<MQL	0.08	**P1B_L**	0.46	0.08	0.17	0.23	0.11	0.13
P1C_M	0.07	0.04	<MQL	0.09	<MQL	0.09	**P1C_L**	0.56	0.11	0.32	0.41	0.23	0.26
P1D_M	0.06	0.04	0.08	0.10	<MQL	0.09	**P1D_L**	0.50	0.14	0.20	0.30	0.22	0.22
P1E_M	0.10	0.04	0.08	0.10	<MQL	0.10	**P1E_L**	0.28	0.13	0.22	0.25	0.19	0.19
P5A_M	0.15	0.03	0.12	0.15	0.09	0.09	**P5A_L**	1.94	0.05	0.87	1.18	0.17	0.24
P5B_M	0.14	0.04	0.13	0.18	0.09	0.10	**P5B_L**	1.95	0.05	0.79	1.10	0.21	0.30
P5C_M	0.10	0.04	0.11	0.09	0.09	0.08	**P5C_L**	0.38	0.08	0.25	0.29	0.11	0.13
P5D_M	0.17	0.04	0.15	0.20	0.11	0.12	**P5D_L**	1.03	0.04	0.75	0.71	0.17	0.21
P5E_M	0.12	0.06	0.12	0.19	0.10	0.10	**P5E_L**	1.30	0.05	0.54	0.81	0.17	0.24
P7A_M	0.09	0.03	0.11	0.11	0.11	0.10	**P7A_L**	0.40	0.04	0.14	0.18	0.11	0.11
P7B_M	0.13	0.03	<MQL	0.10	0.08	0.09	**P7B_L**	0.82	0.04	0.27	0.26	0.11	0.11
P7C_M	0.09	0.03	<MQL	0.10	0.08	0.09	**P7C_L**	0.68	0.04	0.27	0.29	0.11	0.12
P7D_M	0.08	<MQL	<MQL	0.10	0.07	0.09	**P7D_L**	0.52	0.05	0.20	0.25	0.09	0.12
P7E_M	0.07	<MQL	<MQL	0.10	0.07	0.09	**P7E_L**	0.51	0.04	0.22	0.23	0.11	0.12
P8A_M	0.08	<MQL	<MQL	0.10	<MQL	0.09	**P12A_L**	0.38	0.05	0.29	0.29	0.12	0.11
P8B_M	0.07	<MQL	<MQL	0.08	<MQL	0.08	**P12B_L**	0.31	0.08	0.17	0.20	0.12	0.12
P8C_M	0.07	<MQL	<MQL	0.10	<MQL	0.09	**P12C_L**	0.26	0.05	0.18	0.20	0.09	0.09
P8D_M	0.07	<MQL	<MQL	0.08	<MQL	0.07	**P12D_L**	0.34	0.06	0.16	0.23	0.10	0.10
P8E_M	0.05	<MQL	<MQL	0.07	<MQL	0.07	**P12E_L**	0.37	0.07	0.11	0.16	0.09	0.10

MQL is Method Quantification Limit. Note that from sites 8 and 12 only muscle or liver samples, respectively, were analyzed for PFASs. All the target analytes that are not shown here (but listed in Table S2) were either not detected or below MQL in all samples.

Table S4. Concentrations (ng g^{-1} ww) of PFASs in Nile Tilapia muscle and liver tissues.

	Muscle			Liver				
	PFOS	**PFUnDA**	**PFTrDA**	**PFOS**	**PFDA**	**PFUnDA**	**PFDoDA**	**PFTrDA**
T7A	0.07	0.10	0.09	0.12	0.09	0.14	0.09	0.09
T7B	0.05	0.10	0.09	0.15	0.09	0.13	0.08	0.09
T7C	0.04	0.08	0.07	0.10	0.10	0.12	0.08	0.09
T7D	0.07	0.13	0.11	0.15	0.10	0.13	0.09	0.09
T7E	0.07	0.11	0.10	0.13	0.09	0.10	0.08	0.08
TL1	0.05	0.11	0.09	0.25	<MQL	0.16	0.09	0.10
TL2	0.05	0.09	0.09	0.15	<MQL	0.13	0.10	0.09
TL3	0.05	0.09	0.08	0.08	<MQL	0.11	0.08	0.08
TL4	0.05	0.09	0.09	0.13	<MQL	0.14	0.10	0.09
TL5	0.05	0.09	0.08	0.09	<MQL	0.12	0.09	0.09
TL6	0.05	0.09	0.08	0.15	<MQL	0.15	0.10	0.09

MQL is Method Quantification Limit. All the target analytes that are not shown here (but listed in Table S2) were either not detected or below MQL in all samples.

Table S5. Descriptive statistics of ∑PFAS and THg concentrations (ng/g ww) by sampling site.

		Average	Standard Deviation	Min	Max	Count
∑PFASs in Nile Perch Muscle	**Site 1**	0.39	0.05	0.33	0.44	5
	Site 5	0.66	0.10	0.51	0.79	5
	Site 7	0.43	0.07	0.37	0.54	5
	Site 8	0.29	0.03	0.24	0.33	5
∑PFASs in Nile Perch Liver	**Site 1**	1.46	0.29	1.18	1.90	5
	Site 5	3.22	1.32	1.23	4.46	5
	Site 7	1.31	0.25	0.97	1.61	5
	Site 12	1.00	0.15	0.88	1.24	5
∑PFASs in Tilapia Muscle	**Site 7**	0.26	0.05	0.19	0.31	5
	Masese	0.23	0.01	0.22	0.25	6
∑PFASs in Tilapia Liver	**Site 7**	0.52	0.03	0.48	0.56	5
	Masese	0.49	0.09	0.38	0.63	6
THg_{DMA} in Nile Perch Muscle	**Site 1**	70.9	38.4	42.2	134	5
	Site 5	109	48.7	42.4	161	5
	Site 7	34.0	5.63	27.0	42.1	5
	Site 8	31.2	8.65	24.1	41.7	5
THg_DMA in Tilapia Muscle	**Site 7**	7.82	4.07	3.40	13.2	4
	Masese	10.1	1.76	7.12	11.9	6

Table S6. Concentrations of $MeHg^{+}$, Hg^{2+} and THg in Nile Perch and Nile Tilapia muscle, a few liver samples and Certified Reference Materials **IAEA-407** and **IAEA-436**.

Muscle						
	$MeHg^{+}$ (ng/g ww)	**Hg^{2+} (ng/g ww)**	**THg_{ICPMS} (ng/g ww)**	**THg_{DMA} (ng/g ww)**	**$MeHg^{+}$/ THg_{ICPMS} (%)**	**THg_{DMA}/THg_{ICPMS}**
P1A	139	7.90	147	134	94.6	0.91
P1B	68.0	14.0	82.0	78.7	82.9	0.96
P1C	40.0	17.0	57.0	43.8	70.2	0.77
P1D	22.0	29.0	51.0	55.3	43.1	1.08
P5A	121	3.50	125	135	97.2	1.09
P5B	124	7.00	131	131	94.7	1.00
P5D	65.0	9.00	74.0	75.2	87.8	1.02
P5E	105	8.00	113	161	92.9	1.43
P7A	39.7	8.30	48.0	30.9	82.7	**0.64**
P7B	41.3	<MDL	41.3	34.7	100	**0.84**
P7D	44.9	<MDL	47.7	42.1	94.1	0.88
P8B	33.0	6.00	39.0	39.5	84.6	**1.01**
P8D	36.0	3.10	39.1	26.3	92.1	**0.67**
P8E	35.0	5.00	40.0	41.7	87.5	**1.04**
Selected Liver Samples						
P5A	77.1	198	287	316	26.9	1.10
P5E	135	78.0	213	209	63.4	**0.98**
P8D	24.7	22.8	47.5	45.7	51.9	**0.96**
TL5	15.0	7.00	22.0	15.7	68.2	**0.71**
Certified Refererence Materials						
	IAEA-407		**IAEA-436**			
	$MeHg^{+}$ (mg/kg)	**THg (mg/kg)**	**$MeHg^{+}$ (mg/kg)**	**THg (mg/kg)**		
Concentration (ICPMS)	0.20	0.24	3.41	3.79		
Concentration (DMA)		0.22		4.41		
Reference Concentration	0.22	0.22	3.95	4.19		
Trueness (ICPMS)	**92.6 %**	**106 %**	**86.5 %**	**90.5 %**		
Trueness (DMA)		**99.1 %**		**105 %**		

MDL is Method Detection Limit.

Table S7. Pearson correlation coefficients for mercury vs $\delta^{15}N$, $\delta^{13}C$ and ∑PFASs in Nile Perch muscle samples.

	$MeHg^{+}$	Hg^{2+}	THg_{ICPMs}	THg_{DMA}	$\delta^{13}C$ ‰	$\delta^{15}N$ ‰
Hg^{2+}	-0.262					
THg_{ICPMs}	**0.971**	-0.023				
THg_{DMA}	**0.952**	-0.001	**0.986**			
$\delta^{13}C$ ‰	-0.301	**-0.701**	-0.486	-0.504		
$\delta^{15}N$ ‰	**0.660**	0.259	**0.749**	**0.734**	-0.577	
∑PFASs_Muscle	**0.708**	-0.069	**0.716**	**0.727**	-0.385	**0.771**

Bold values have *P* values less than 0.05.

Figure S3. Comparison of THg concentrations measured in Nile Perch and Nile Tilapia muscle in this study with previously reported measurements[1] from Lake Victoria. Sites 1, 5, 7 and 8 are from our study.

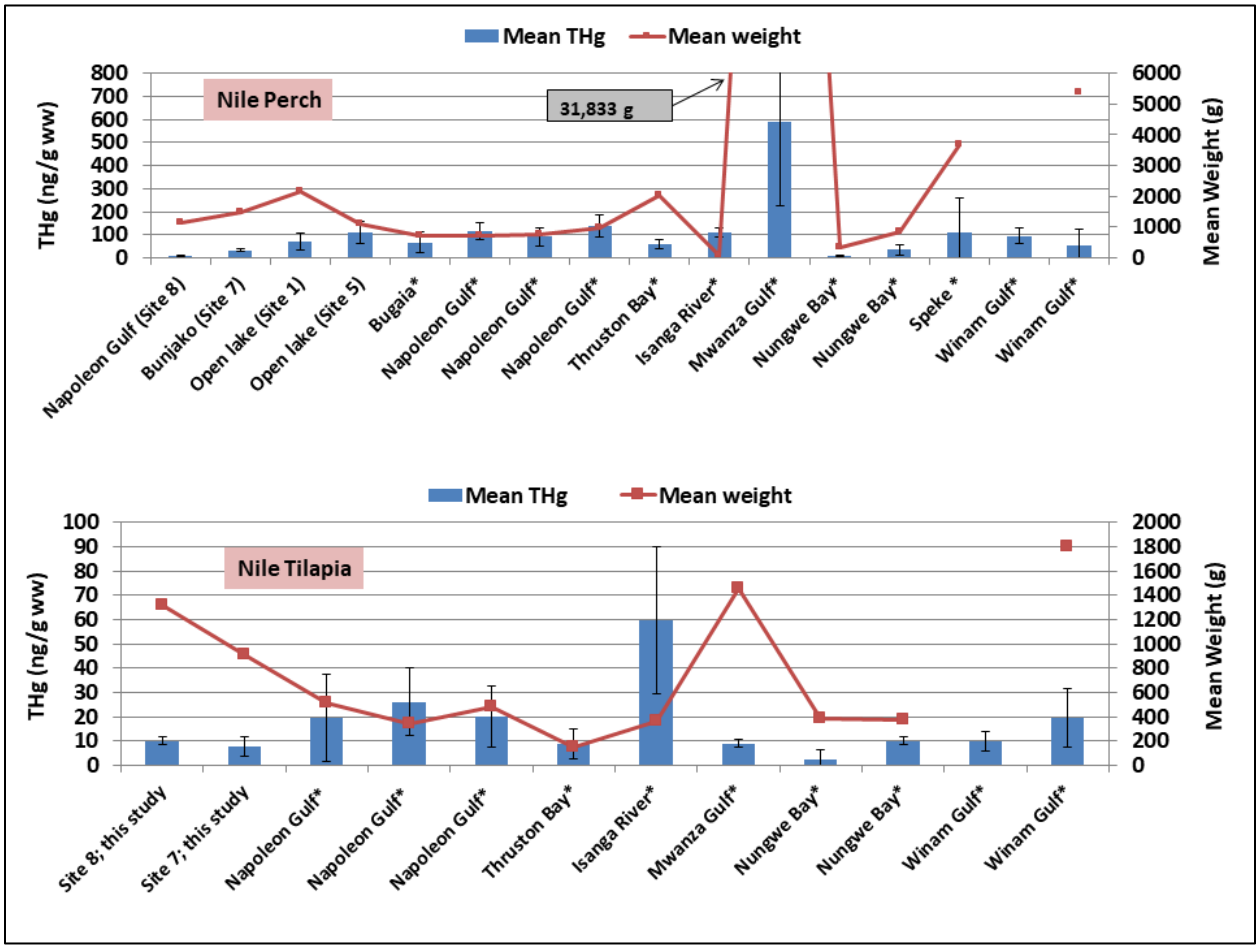
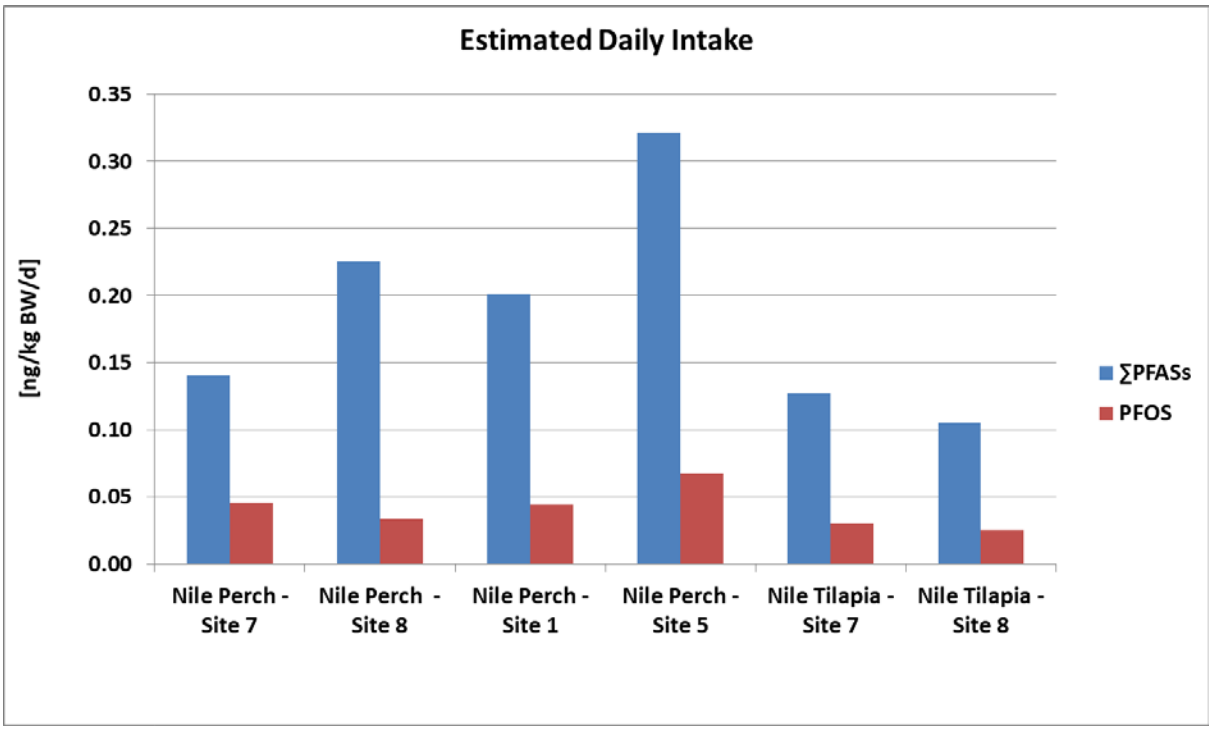


Table S8. Comparison of THg measurements in this study with other recently reported measurements from studies elsewhere globally.

Country	Water body	Fish species	Sample type	Year of Sampling	THg Concentration (µg/kg ww)	Statistic	Reference
Uganda (Lake Victoria)	Site 1	Nile Perch	Muscle	2017	70.9 (42.2 - 134)	Mean (Range)	This study
	Site 5				109 (42.4 - 161)		
	Site 7				34.0 (27.0 - 42.1)		
	Site 8				31.2 (24.1 -41.7)		
	Site 7	Tilapia			7.82 (3.40 -13.2)		
	Masese Landing site (caught from Napoleon Gulf)				10.1 (7.12 - 11.9)		
Germany	Rivers Weser, Elbe, Moselle & Havel; Lake Starnberg and Coastal lagoon of Baltic Sea (Kleines Haff)	Chub, Roach, Bream, Whitefish, Perch	Muscle	2016/2017	5 - 87	Mean	Rödel et al., 2020
USA	Chesapeake Bay area	Multiple freshwater and marine fish (61 species)	Muscle and muscle-equivalents estimated from liver	1990-2017	220 (1.00 - 4850)	Geometric mean (range)	Willacker et al., 2020
Turkey	Lake Mogan	Carp, Pike,Tench	Muscle	2017/2018	0.7 - 1.3	Annual mean	Kucukosmanoglu and Filazi, 2020
			Liver		0.8 - 1.0	Annual mean	
South Africa	Olifants River Basin	Multiple freshwater species (8 species)	Muscle	2012	21 - 1100	Range	Verhaert et al., 2019
			Liver		33 - 1200	Range	
Egypt	Manzala Lake	Nile Tilapa	Muscle	2016	19 (72)	Mean; Summer (Winter)	Sallam et al., 2019
		Flathead grey mullet			5 (24)		
		African cat fish			12 (24)		
South Africa	Vaal Dam	*Labeobarbus aeneus,*	Muscle and Liver	2016	96.2 (55.6)	Mean; Muscle (Liver)	Plessl et al., 2019
		Labeobarbus kimberleyensis			133 (72.6)		
		Labeo umbratus			49.8 (34)		
Tanzania	Coastal Region of Tanzania, Zanzibar and Pemba	Farmed Milkfish	Muscle	2016	10 (8)	Mean (Median)	Mwakalapa et al., 2019
		Mullet (wild)			10 (13)		
French Guiana	Multiple Creeks and rivers	Piscivores	Muscle and whole fish for fish with weight ≤ 1g ww	2004-2015	234 (509)	Mean; Creeks (Rivers)	Gentès et al., 2019
		Carnivores			179 (184)		
		Omnivores			156 (146)		
		Periphytophagous			171 (46)		
		Benthivores			203 (144)		
		Herbivores			65 (11)		
Canada (Lawrentian Great Lakes)	Lake Superior	Multiple species (11 species)	Muscle	1970s - 2000s	80 - 360	Mean	Visha et al., 2018
	North Channel				40 - 300	Mean	
	Georgian Bay				50 - 600	Mean	
	Lake Huron				60 - 390	Mean	
	Lake Erie				30 - 220	Mean	
	Lake Ontario				70 - 400	Mean	
	St.Lawrence River				140 - 600	Mean	
Colombia	La Raya marsh	Carnivores	Muscle	2010	1020 (640)	Mean; Dry Season (Rainy Season)	Marrugo-Negrete et al., 2018
		Non-carnivores			260 (190)		
	Ayapel marsh	Carnivores			560 (420)		

		Non-carnivores			190 (130)		
South Africa	Ga-Selati River	Multiple (8 fish species)	Muscle	2014	10 - 300	Mean	Govaerts et al., 2018
Germany	Danube River	Chub	Muscle	2015	150 (4.8 - 349)	Mean (Range)	Fliedner et al., 2018
		Bream			174 (28.1 - 372)		
		Perch			275 (131 - 509)		
Namibia	Northern Benguela (off the coast)	Cape monkfish	Muscle	2016/2018	126 (12 - 647	Mean (Range)	Erasmus et al., 2018
			Liver		106 (20 - 381)		
Poland	Bought on Polish market	Multiple (6 Species)	Muscle	not mentioned	6 - 138	Mean	Łuczy ´nska et al., 2017
Poland	Warta Mouth National Park	Multiple (8 species)	Muscle	2009-2014	67 - 308	Mean	Kalisinska et al., 2017
USA and Canada	Multiple sites (4262) west of the continental divide	Multiple (206 species)	Muscle and muscle-equivalents estimated from average whole body-to-muscle ratios reported elsewhere	1969-2014	170 (1 - 28540)	Geometric mean (range)	Eagles-Smith et al., 2016

Figure S4. Estimated average daily intake of PFASs through fish consumption from Lake Victoria (for Uganda), assuming 2013 average fish consumption of 12.5 kg/person/year. [2]



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