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Lydia Mwikali

lydiamwikali5@gmail.com

Mbarara University of Science and Technology

Julia Räder

Navigo Proteins GmbH, Halle (Saale)

Grace Birungi

Mbarara University of Science and Technology

Urs Berger

Helmholtz-Centre for Environmental Research – UFZ

Kenneth Arinaitwe

Helmholtz-Centre for Environmental Research – UFZ

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Spatiotemporal Profiles, Pollution Sources and Ecological Risk Assessment of Poly-and Perfluoroalkyl Substances in Sediments of Lake Victoria, East Africa

Lydia Mwikali[a], Julia Räder[b,c], Grace Birungi[a], Urs Berger[c,d], Kenneth Arinaitwe[c,e]

[a] Department of Chemistry, Mbarara University of Science and Technology, P. O Box 1410

Mbarara, Uganda

[b] current address: Navigo Proteins GmbH, Halle (Saale), Germany

[c] Helmholtz-Centre for Environmental Research – UFZ, Leipzig, Germany

[d] current address: Faculty of Medicine, Department of Pediatrics, Children's Hospital, Adolescent Medicine and Neonatology, Freiburg University Hospital, Freiburg, Germany

[e] current address: Johann Heinrich von Thünen Institute for Fisheries Ecology, Bremerhaven, Germany.

*Corresponding author; E-mail address: lydiamwikali5@gmail.com

2024phd024@std.must.ac.ug

ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are persistent contaminants with potential bioaccumulative and ecotoxicological effects. Yet, little is known about their occurrence, sources, and ecological risks in Lake Victoria sediments. PFAS contamination may threaten the lake's ecological integrity and socio-economic importance. To address these gaps, we investigated PFAS occurrence and distribution in Lake Victoria sediments, elucidated their potential sources, and associated ecological risks. Sediment samples were collected in September/October 2017, May 2019 and September/October 2019 and extracted on an ultrasonic bath, cleaned up by solid phase extraction and analyzed for PFAS using liquid chromatography with tandem mass spectrometry. Total PFAS (∑PFAS) levels ranged from <6.78 to 1021 ng/kg dw in September/October 2017, 165 to 1004 ng/kg dw in May 2019, and <1.98 to 593 ng/kg dw in September/October 2019. Intra-region PFAS spatial distribution was site-dependent with the highest concentrations observed at sites near urban centers. PFAS in the core sediment implied a significant post depositional redistribution of PFAS in Lake Victoria. In general, there was no significant lake-wide spatial structure exhibited by the PFAS concentration. Despite the low PFAS concentration, Principal Component Analysis-Multiple Linear Regression (PCA-MLR) indicated that the primary source of PFAS into Lake Victoria was atmospheric deposition. PFAS in Lake Victoria posed moderate to negligible ecological risks except for PFBA. Occurrence of PFAS in Lake Victoria shows that contamination pressures are persistent and that continuous monitoring and mitigation are necessary to protect the ecological and socio-economic functions of the lake.

Keywords: Poly- and Perfluoroalkyl substances, Sediment, Spatiotemporal profiles, Source identification, Ecological risks

1. Introduction

Poly- and perfluoroalkyl substances (PFAS) are synthetic organic molecules with a minimum of one fully fluorinated methyl (-CF_3) or methylene (-CF_2) group (Wang et al., 2021). They have a hydrophobic fluoroalkyl moiety (C_nF_{2n+1}) and a hydrophilic functional group (Buck et al., 2011). The fluoroalkyl moiety imparts extreme chemical and thermal stability, making PFAS persistent in the environment. Since the 1960s, PFAS have been used extensively as water- and oil-repellents in textiles, food packaging, and upholstery (Richardson & Kimura, 2016; Hekster et al., 2003; Herzke et al., 2012) and in applications such as non-stick cookware and fire-fighting materials (Brendel et al., 2018). PFAS are classified as contaminants of emerging concern, because of their pervasiveness, persistence and possible toxicity even at low concentrations (Podder et al., 2021).

PFAS have been detected in all environmental compartments including: sediments, soil, food, surface waters, ground water, drinking water and living organisms (Ahrens & Bundschuh, 2014a; Fujii et al., 2007; Nakayama et al., 2019); although most of the data available is from the Global North. Measurable PFAS levels in air, snow and atmospheric particles were reported in remote regions such as the Arctic (Stock et al., 2007; Young et al., 2007); however the limited data from Sub-Saharan Africa creates a major geographical disparity in environmental toxicology literature. PFAS are persistence and bioaccumulative in nature hence have the potential to risk ecosystem and public health. Therefore, PFAS pollution remains a major focus of environmental research worldwide.

PFAS exposure is associated with negative health outcomes such as endocrine disruption, immunotoxicity, hepatotoxicity, cancer and neurodevelopmental impacts (Butenhoff & Rodricks, 2015; Grandjean & Clapp, 2015; Sunderland et al., 2019). Thus, several PFAS such as perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), perfluorohexanesulfonic acid (PFHxS) are now listed under

the Stockholm Convention with many others proposed for restriction (Stockholm Convention, 2017, 2022; UNEP, 2009;ECHA, 2020). Their environmental partitioning is strongly influenced by chain length: short-chain PFAS have a lower affinity for organic carbon and are thus more mobile over long distances whereas long-chain PFAS tend to accumulate near their sources as desorption potential decreases with increasing chain length (Prevedouros et al., 2006; Sepulvado et al., 2011; Zhao et al., 2016).

While PFAS contamination data from Africa compared to the Global North remains limited, studies from Uganda are exceptionally scarce. Occurrence of PFAS in water from Lake Victoria, the largest fresh water lake in Africa, (Kurwadkar et al., 2022; Ssebugere et al., 2020) has been reported. However, there is no data about sediment. Sediments are an important repository of PFAS, serving as both a long term sink and a potential secondary source to the water column, facilitating uptake into aquatic food webs and posing a threat to benthic ecosystems (Prevedouros et al., 2006; He et al., 2018). Therefore, it is essential to evaluate the ecological risks associated with PFAS in sediments, assess their possible negative effects on aquatic ecosystems, and promote efficient environmental management and regulatory decision-making.

Lake Victoria is a vital source of fish and domestic water, making it a critical PFAS-exposure pathway for humans (Arinaitwe et al., 2021; Orata et al., 2011; Bagumire et al., 2018; Knaap & Ligtvoet, 2010). Given the Lakes ecological significance and socio-economic importance, a comprehensive assessment of PFAS in the sediments is critical. Understanding their occurrence, sources and ecological risks is necessary for guiding pollution management in the lake. In this study, surface sediments from bays, nearshore regions, and the open lake, along with a sediment core from Lake Victoria, were analyzed to assess (i) the occurrence of PFAS, (ii) their spatial distribution, anthropogenic influences, and source pathways, (iii) their historical deposition profiles, and (iv) the ecological risks posed by PFAS contamination in the lake sediments.

2. Materials and Methods

2.1 Study Area

Lake Victoria (Figure 1) in East Africa, is the world's second-largest freshwater lake with an area coverage of about 68 800 km^2 and is shared across Uganda (45%), Kenya (6%), and Tanzania (49%). The lakes basin catchment area is 193 000 km^2 shared between Kenya (21.5%), Tanzania (44%), Uganda (15.9%), Burundi (7.2%) and Rwanda (11.4%) (Awange, 2021). Lake Victoria spans 412 km across the equator between latitudes 0˚ 300 N – 3˚120 S, and 355 km between longitudes 31˚ 370 W – 34˚ 530 E (Abuodha & Hecky, 2005; Okungu et al., 2005). The Lake has 3,450 km shoreline a maximum depth of 84 m, and a mean depth of 40 m, and a 140-year water retention period (Aloo et al., 2013). The lake is surrounded by a dense and rapidly expanding human population and sustains one of the largest inland fisheries in the world, aimed at both local and global markets (Kolding et al., 2014).

2.2 Chemicals and Reagents.

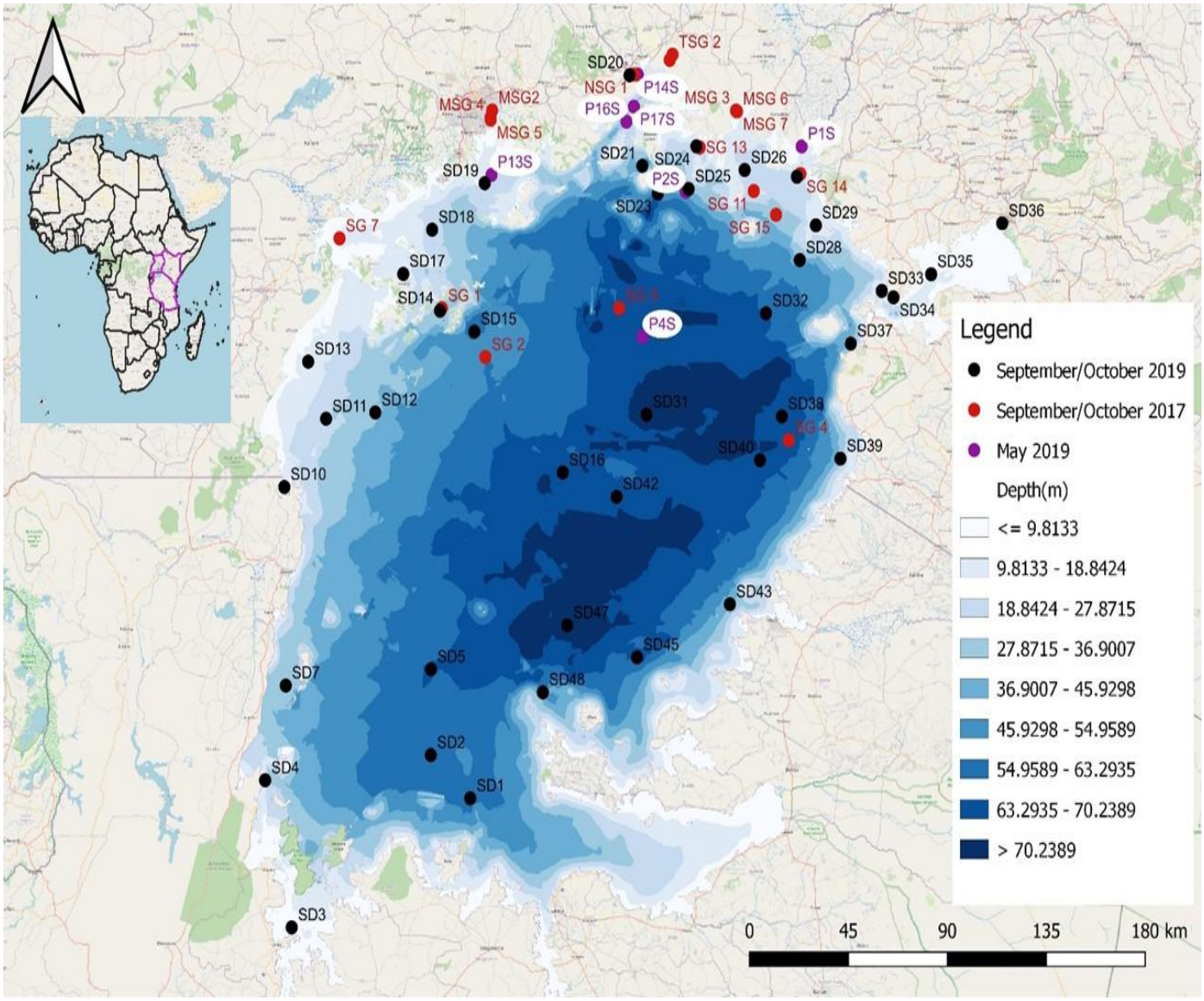
The internal and native standard mixtures of chemical purities > 98% and isotopic purities of ≥ 99% for this study were obtained from Wellington Laboratories, Canada (Table S1). Methanol (HPLC grade, purity > 99.9%), ammonium acetate (HPLC grade, purity >99.9%), ammonium hydroxide (NH_4OH, HPLC grade; v/v, 50%) and acetic acid (HPLC grade, purity>99.8%) were procured from Sigma Aldrich, United States. Sodium hydroxide and hydrochloric acid were obtained from Merck (Darmstadt, Germany). Ultra-pure water was obtained from the laboratory water purification system (MilliQ-Integral, Merckmillipore, Billerical, USA).

2.3 Sample Collection and Preparation

Sampling was carried out on three separate cruises in September/October 2017, May 2019 and September/October 2019 (Figure 1). The September/October 2017 and 2019 sediment samples were

collected during lake-wide hydro-acoustic surveys by the Lake Victoria Fisheries Organization and were taken using a grab sampler. The May 2019 samples were collected aboard the Uganda National Fisheries Resources Research Institute research vessel, RV Hammerkop, using a gravity sampler (UWITECH GmbH, Mondsee, Austria). September/October 2017 and May 2019 sediment samples were collected from the Ugandan part of Lake Victoria. The September/October 2019 samples were collected on a lake-wide scale.

The September / October 2017 batch composed of 21 surface sediment samples collected from Murchison Bay, Napoleon Gulf Bay, Thruston Bay and on the open lake and the May 2019 sample set consisted of 7 surface sediment cores and one sediment gravity core (P4S; Table S2). May 2019 surface sediment cores samples were cut into three sections of 5 cm each starting from the top: 0 - 5 cm, 5 - 10 cm, 10 - 15 cm. The gravity core sediment layer (0-16 cm) was sectioned into 8 sediment layers of 2 cm each. September/October 2019 batch composed of 41 surface sediment samples (Table S3). All the sediment samples were stored frozen at -20 °C till analysis.



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124 Figure 1. Map of Lake Victoria showing the sampling sites. The surface sediment samples are grab
125 samples. P4S is the core sediment sample collected using a gravity corer for long-term historical
126 deposition profiling.

127

2.4 Sample Extraction and Clean-up

Extraction was done following the method by Powley et al.(2005), with some slight modifications. Briefly, 1 g of sieved sediment sample was spiked with 20 µg/L (50 µL) of internal standards and 1 mL of 200 mM sodium hydroxide in MeOH was added. After standing for 15 minutes, MeOH /ACN 50:50 (3 mL) was added, the mixture vortexed for 30 seconds. Prior to centrifuging at 5000 rpm for ten minutes, it was placed in ultrasonic bath at 40 °C for 10 minutes. The supernatant was transferred to a PP Falcon tube (15 mL). To the residue, MeOH /ACN 50:50 (3 mL) was added, and again, vortexed, sonicated, centrifuged and the supernatant combined with previous portion. The combined solution was concentrated to 1 mL in a Horizon XcelVap at 40 °C.

The extracts were cleaned up using SPE as described by Filipovic & Berger (2015), with some modifications. The SPE cartridge (CUQAX256, United Chemical Technologies Inc., Bristol, PA) was preconditioned with 16 mL of 2% vol 1-methylpiperidine in MeOH /ACN 80:20 v/v, rinsed with Milli-Q-water (2 mL) and loaded with the extracts. PFAS were eluted with 2 mL of 2 % vol 1-methylpiperidine in MeOH /ACN 80:20 v/v into a 15 mL PP Falcon centrifuge tube (pre-cleaned with 2% vol 1-methylpiperidine in MeOH /ACN 80:20 v/v). Under nitrogen, the eluate was concentrated to dryness at 40 °C and resuspended with MeOH (500 µL). To the solution, 500 µL of 4 mM aqueous ammonium acetate was added, sonicated, transferred to Eppendorf centrifuge tube (1.5 mL), and centrifuged for 20 minutes at 0 °C at 7000 rpm. The supernatants were transferred to polypropylene vials and kept at 4 °C till analysis.

2.5 Analytical Method Validation and Quality Assurance

The performance of the method was assessed using linearity, sensitivity, recovery, and calibration tests. Method linearity was evaluated using multi-point calibration curves using standards in the range of 0.01µg/L to 20 µg/L. Sensitivity was determined through triplicate analyses of low concentration

standards of the PFAS analytes. The concentrations producing S/N ratios of 3.3 and 10 were defined as the LOD and LOQ respectively. Recoveries were determined by spiking the sediment samples with 50 µL of 20 µg/L of the internal standards. The percentage recoveries were calculated as;

$$\%\, Recovery = \frac{Cd}{Cs} * 100\% \ldots\ldots\ldots\ldots\ldots\ldots\ldots i$$

Where Cd and Cs are the detected and spiked concentration of each individual PFAS.

2.6 PFAS Analysis - Instrument and Analytical Conditions

The purified extracts were analyzed using high-performance liquid chromatography (HPLC)-tandem mass spectrometry (MS) operated in a negative electrospray ionization (ESI) mode. The details of the LC-MS/MS instrument method settings are provided in Table S4. Multiple Reaction Monitoring (MRM) mode was used for the qualitative and quantitative analysis of the PFAS analytes (Table S5). Retention times and intensities of MRM transitions were used as diagnostic parameters for peak identification and selection.

2.7 Ecological Risk Assessment

Ecological risk was assessed using the risk quotient (RQ), calculated as the ratio of the measured environmental concentration (MEC) to the lowest predicted no-effect concentration (PNEC) for sediments as listed in the NORMAN Ecotoxicological Database (NORMAN, 2026) using equation *ii*

$$RQ = \left(\frac{MEC}{\text{Lowest PNEC}}\right) \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ ii$$

Risk quotients were calculated by taking each individual PFAS measured concentration and the lowest PNEC for sediments for the respective PFAS compound from the Norman ecotoxicological database (NORMAN, 2026). Risk levels were categorized as follows: RQ ≥ 1 indicates high ecological risk; 0.1 ≤ RQ < 1 reflects moderate risk; and RQ < 0.1 corresponds to low risk. Nevertheless, this rating does not offer a thorough evaluation of the chemical's overall ecological danger at the regional level (Liu et al., 2020). To overcome this constraint, we introduced a corrected risk quotient (CRQ) that considers the

detection frequency (DF) of the individual PFAS compounds. This reduces uncertainty and offers a more accurate evaluation of ecological risk.

$$CRQ = RQ_{mean} * DF \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots iii$$

Where RQ_{mean} is the calculated mean risk quotient and DF is the detection frequency for each individual PFAS.

2.8 Data management and Statistical Analysis

The data was managed in Microsoft Excel v21 and analyzed using SPSS v26 software (IBM corp.) Concentrations that were below the LOQ were substituted with a value equal to half the LOQ of the specific PFAS in statistical tests. Analysis of variance was performed to test for significant differences in mean concentrations of PFAS among the Lake Victoria region countries (Kenya, Uganda and Tanzania) and significance in mean concentrations among the sampling sites grouping. Independent t-test was used to test for significance differences in PFAS concentrations for same analytes in the September/October 2017 and May 2019 samples. All the statistical tests were carried out at a 5% level of significance. QGIS 3.14.1 (https://qgis.org/) and an in-built OpenStreetMap tile as a base map was used. The bathymetry data was obtained from http://faculty.salisbury.edu/~sehamilton/LV_bathy/, Accessed 12 March 2024. The spatial autocorrelation was calculated using the global and local Moran's I statistics. For source identification of PFAS, diagnostic homologue ratios and principal component analysis multiple linear regression (PCA-MLR) model was employed. To calculate source contributions, PCA factor scores were converted to absolute principal component scores (APCS). The overall PFAS concentrations were modelled as the dependent variable and the components found in PCA as independent. The regression coefficients from the model were employed in determining the source contributions using equation *iv*

$$i\% = \left(\frac{Bi}{\sum_i^n Bi}\right) * 100 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots iv$$

Where *Bi* represents beta coefficient for a specific factor and *n* is the number of factors (Thurston & Spengler, 1985).

3. Results and Discussion

3.1 Quality Control and Assurance

Overall, the matrix spike recoveries ranged from 43.9 % to 98 % with precisions in the range of 0.2 to 13 %, except for the neutral precursor compound FOSA (Table S5). During sample preparation, procedural blanks were added to each batch of samples. The calibration curves had linearity r^2 >0.99. The instrumental limits of detection and quantification were set as the PFAS concentration required to produce a signal to noise ratio of 3.3 and 10 respectively. Where multiple MRM transitions were targeted, the one with the highest intensity was used for quantification.

3.1 PFAS Concentration in Lake Victoria

Overall, over the course of the three sampling campaigns, PFAS concentrations demonstrated significant temporal fluctuation (Table 1). The values of total PFAS (∑PFAS) varied from <6.78 to 1021 ng/kg in September/October 2017, 165 to 1004 ng/kg in May 2019, and <1.98 to 593 ng/kg in September/October 2019. May 2019 had the highest mean ΣPFAS values (424 ± 320 ng/kg), which were roughly twice as high as those found in September/October 2017 (219 ± 310 ng/kg) and September/October 2019 (167 ± 140 ng/kg).

In September/October 2017 samples, PFBA had the highest mean concentration of 82.1 ± 108 ng/kg dw with a detection frequency of 43% despite its high solubility in water, possibly due to its presence in sediment pore water (Feng et al., 2020). PFTrDA had the lowest mean concentration of 0.78 ± 0.94 ng/kg

dw. The low abundance of PFTrDA; a long-chain PFAS is likely governed by source availability with high hydrophobicity and fluorophilic (Prevedouros et al., 2006) limiting its mobility and widespread dispersion across the sampling sites. PFOA was the most prevalent with a detection frequency of 67 % due to its historical large scale production (Kissa, 2005) in addition to its high solubility in pore water (Feng et al., 2020).

In May 2019 sampling campaign, PFOA had detection frequency of 100% with mean of 131 ± 110 ng/kg dw. PFNA, PFUnDA and FOSA had the lowest detection frequencies of 5%. PFAS concentration in May 2019 increased compared to those in September/October 2017 although the increase was not statistically significant except for PFOA ($p = 0.046$), PFUnDA ($p = 0.41$) and PFTrDA ($p < 0.001$) suggesting continued contamination of the lake sediments with PFAS. For PFBA and FOSA, PFAS mean concentrations showed a clear decrease between 2017 and 2019. PFBA is highly mobile in water while FOSA degrades to PFOS (Prevedouros et al., 2006; Yan et al., 2025). The observed declines in PFBA and FOSA could therefore be attributed to their environmental behaviors assuming no new, ongoing inputs.

PFNA was the most abundant during the September/October 2019 sampling campaign. Samples from the Ugandan, Tanzanian and Kenyan parts of the lake had total PFAS ($\sum_7$PFAS) concentration ranges of 12.0 - 583 ng/kg dw, < 4.78 - 456 ng/kg dw and <17.1-367 ng/kg dw with means of 169 ± 147 ng/kg dw, 163 ± 135 ng/kg dw and 168 ± 132 ng/kg dw, respectively (Table S6). However, the total mean concentrations were not significantly different among the three countries ($p = 0.981$, $\alpha = 0.05$) pointing to a significant overarching source. Atmospheric deposition was recognized as a major source of PFAS in Lake Victoria (Arinaitwe et al., 2021) hence the similarity by means. In the Ugandan part, all PFAS had detection frequencies of 100% implying widespread PFAS contamination. PFNA had the highest mean concentrations in each of the three countries and was the most predominant PFAS suggesting distinct source profiles.

241 Table 1: PFAS Concentrations in ng/kg dw for September/October 2017 and May 2019 Surface Sediment Samples (0-5 cm) and
242 September/October 2019 in Lake Victoria (Min = minimum, Max = maximum, SD = standard deviation, DF =detection frequency, n.a
243 =not analyzed)
244

PFAS	**Sept/Oct 2017 (n=21)**			**May 2019 (n=7)**			**Sept/Oct 2019 (n=41)**		
	Min- Max	**Mean±SD**	**DF (%)**	**Min- Max**	**Mean±SD**	**DF (%)**	**Min- Max**	**Mean±SD**	**DF (%)**
PFBA	<0.75 -368	82.1±108	43	<0.75 -180	43.4±75.4	29	n.a	n.a	n.a
PFHxA	<0.38 -208	59.6±79.6	38	90.0 -330	218±90.2	100	n.a	n.a	n.a
PFHpA	<0.38 - 21.0	1.18±4.54	5	n.a	n.a	n.a	n.a	n.a	n.a
PFOA	<1.50 -146	41.1±40.1	67	71.0 -378	131±110	100	n.a	n.a	n.a
PFNA	n.a	n.a	n.a	<1.50 -20.0	5.68±8.49	29	0.80 -139	46.7±34.8	100
PFDA	<0.38 -25.0	7.73±9.54	43	<0.38 -29.0	10.1±10.8	57	<0.05 -97.9	33.8±28.2	98
PFUnDA	<0.38 -23.00	7.39±8.89	43	<0.38 -52.0	10.1±19.6	29	<0.03 -82.4	24.1±21.7	98
PFDoDA	<0.75 -32.0	1.88±6.90	5	n.a	n.a	n.a	<0.07 -41.5	13.3±10.5	95
PFTrDA	<0.75 -4.00	0.78±0.94	19	<0.75 -12.0	5.39±4.72	71	1.00 -53.0	10.1±10.2	98
PFTeDA	n.a	n.a	n.a	n.a	n.a	n.a	<0.01 -156	32.6±29.3	98
PFHxDA	n.a	n.a	n.a	n.a	n.a	n.a	<0.02 -23.1	6.35±4.91	98
PFOS	<1.13 -186	15.9±49.1	10	n.a	n.a	n.a	n.a	n.a	n.a
FOSA	<0.38 -8.00	1.23±2.17	29	<0.38 -3.00	0.71±1.06	29	n.a	n.a	n.a
∑PFAS	<6.78 -1021	219±310		165 -1004	424±320		<1.98 -593	167±140	

245

3.2 Spatial Distribution Profiles for PFAS in Lake Victoria Surface Sediments

PFAS were detected in all the sediment samples. The spatial profiles were generated from the surface sediments and the uppermost layer (0-5 cm) sedimentss. The upper most layer of the core sediment (0-2 cm) was also used. Spatial distribution profiles for September/October 2017 and May 2019 samples were grouped based on the location of the sampling site in the lake into near-shore rural region, close to near-shore region, near-shore urban region and open lake (Figure 2). September/October 2019 samples provided a lake-wide trend (Figure 3). Spatial distribution for September/October 2017 and May 2019 samples showed a site-dependent pattern linked to anthropogenic activity.

3.2.1 September/October 2017 and May 2019 sediment samples

In the near-shore rural region, PFOA was detected in all samples except SG15. The highest concentration of PFOA was 127 ng/kg dw. PFHxA had concentration of 330 ng/kg dw in site P16S. In the close to the nearshore region, FOSA was only found in sample TSG2 and P13S. In the near urban region PFBA and PFOA were the most dominant with high concentrations of 368 ng/kg dw and 101 ng/kg dw respectively. PFHxA was the most dominant PFAS in the open lake. PFOA had the highest concentration of 378 ng/kg dw at site P2S. PFBA was only found on site SG2.

Higher total PFAS concentrations were detected nearshore, near urban centers like Kampala (Murchison Bay) and Jinja (Napoleon Gulf), supporting an anthropogenic origin from industrial effluent, wastewater, and domestic discharge (Mafuta et al., 2017; Davis et al., 2007). Previous studies in Lake Victoria linked the higher PFAS to pronounced enrichment in urban drainage (Arinaitwe et al., 2021). Similar urban hotspots have also been reported in other African freshwater systems (Kasambala et al., 2024; Ssebugere et al., 2020). However, these PFAS concentrations remain lower than those reported in heavily industrialized regions in Asia, Europe and America (Mukonza & Chaukura, 2025). The prevalence of

PFAS and the spatial gradient from landfill and wastewater sources to nearshore lake sediments reflect worldwide contamination pathways while emphasizing the unique susceptibility of urban shorelines in areas that are not adequately monitored.

Nearshore rural regions had fewer and lower-concentration PFAS detections. PFHxA showed more homogeneous concentrations (150-200 ng/kg dw) across all regions, possibly indicating a significant atmospheric input source (Yao et al., 2025).

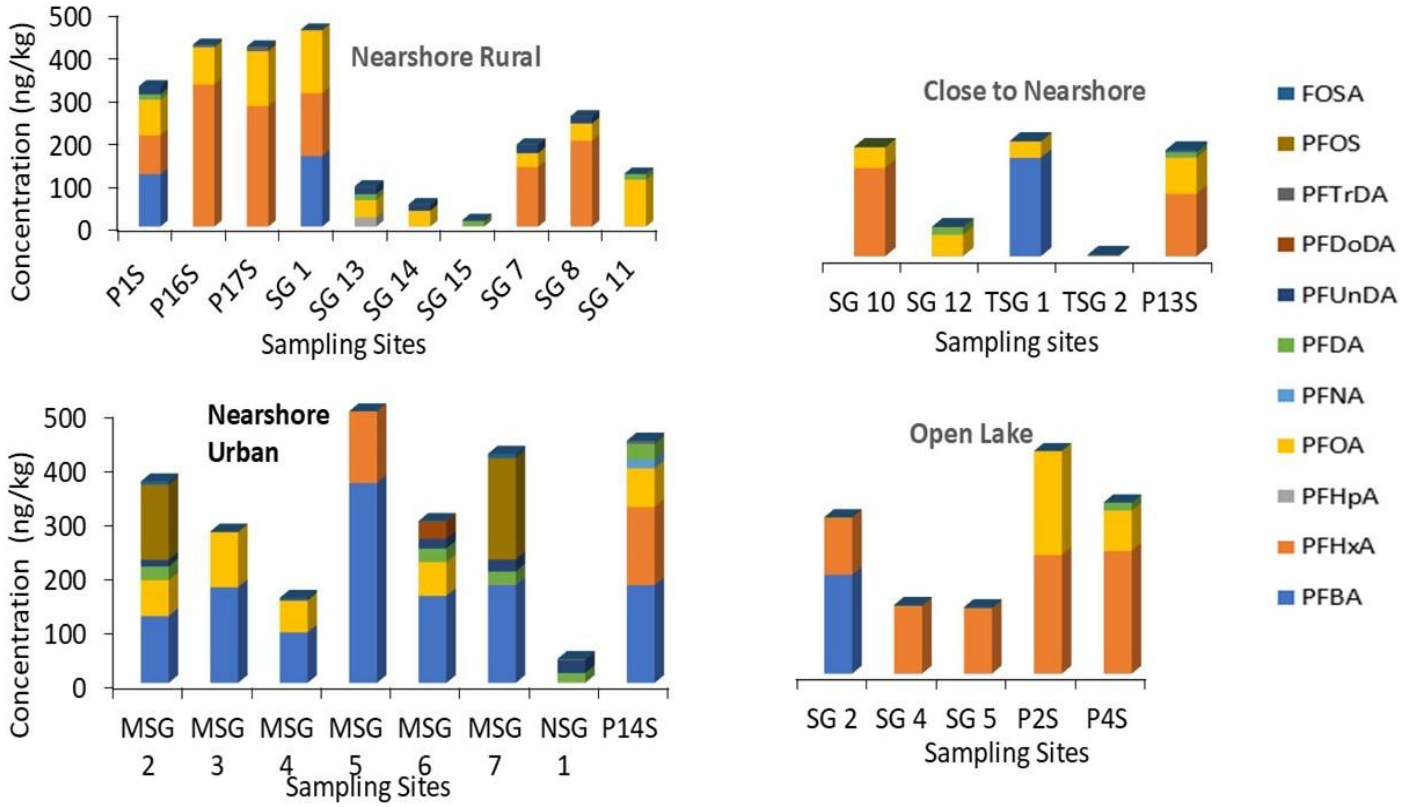

Figure 2: PFAS concentrations (ng/kg dw) in the surface sediments collected in September/October 2017 and May 2019 based on the sampling locations

The differences between mean concentrations of PFAS in the sampling site groupings were all statistically insignificant ($p \geq 0.05$) except for PFBA ($p = 0.026$). PFBA dominated the sampling sites in the near-

shore urban region. The high PFBA in the near-shore urban region could be due to continuous flooding by urban runoffs and wastewater discharges. This is consistent with (Xu et al., 2021), PFBA was dominant in sediments suggesting that the growing use of short-chain PFAS alternatives, potentially augmented by urban-derived inputs, contributes significantly to sediment contamination.

3.2.2 September/October 2019 Sediment Samples

The lake-wide spatial distribution of PFAS concentrations is presented in Figure 3. The lowest Σ_7PFAS concentration (27.2 ng/kg dw) was found in sample SD37 taken near Takawiri island which is a pristine freshwater island characterized by sandy, low–organic-carbon sediments with inherently limited PFAS sorption capacity (Odede et al., 2014). This is in agreement with a study that investigated the relationship between PFAS and overall organic carbon and grain size of sediments (Li et al., 2024). Stronger PFAS sorption capabilities are typically found in sediments with smaller particles and greater organic carbon levels as the sediment's hydrophobic organic carbon components aid in binding the PFAS molecular tails, resulting in stable sorption sites (Feng et al., 2024). Long chain PFAS are more hydrophobic and therefore are likely to adsorb to fine-grained sediments unlike sand which has coarse grains (Li et al., 2024). The highest Σ_7PFAS concentration (354 ng/kg dw) was in sample SD32, near Winam Gulf, influenced by urban and riverine discharge (Abuodha & Hecky, 2005) indicating a significant contamination hotspot.

In sediment sample SD10 from Rubafu Bay, Kagera region (Tanzania), only PFNA (42.5 ng/kg dw) and PFTrDA (16.8 ng/kg dw) were quantifiable. Their presence in this agriculturally dominant region is notable, as fluorine-containing agrochemicals are commonly used (Jeschke, 2017). This suggests that PFNA, PFTrDA, and/or their precursor compounds may originate from the application and environmental degradation of such agricultural products. Most of the nearshore sites had higher ΣPFAS, as long chain

PFAS (C ≥ 6 for PFSAs and C ≥ 7 for PFCAs) tend to accumulate near sources due to their high organic carbon partitioning and low mobility (Cousins et al., 2011; Ahmed et al., 2020; Sepulvado et al., 2011). Lower concentrations offshore may be due to dilution, reduced sedimentation or different sediment dynamics.

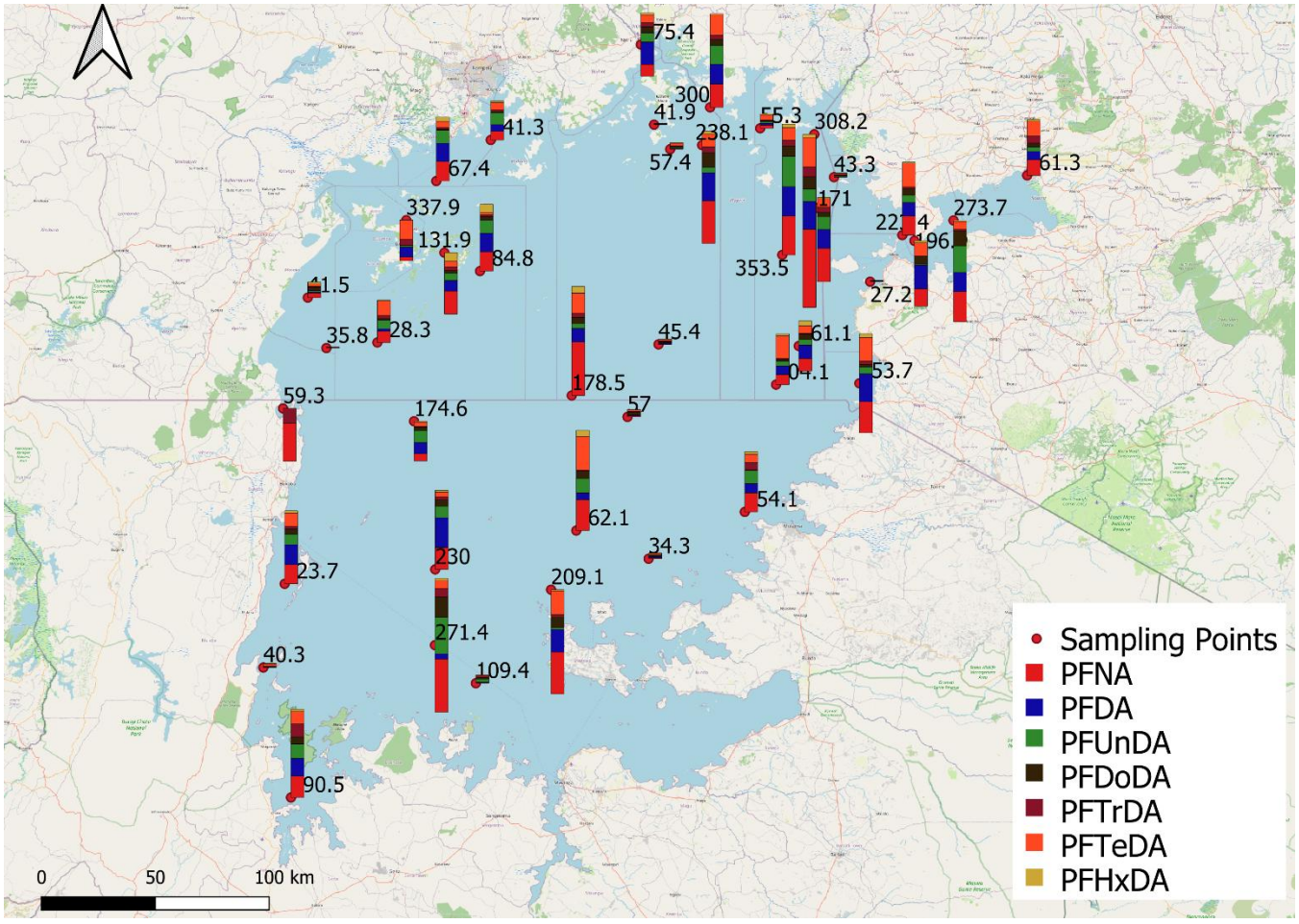

Figure 3: Spatial distribution profiles of total PFAS concentrations (ng/kg dw) in the September/October 2019 samples of Lake Victoria, East Africa

The global and local Moran's I statistics collectively indicate that the spatial distribution of ΣPFAS in Lake Victoria surface sediments is predominantly random and devoid of strong underlying spatial

structure. The global Moran's I was negative and non-significant ($I = -0.102$, expected $I = -0.025$, $p = 0.782$, one-sided test for clustering), confirming that ΣPFAS concentrations across the 68,000 km² lake do not exhibit a systematic tendency toward spatial aggregation, nor do they display a significant checkerboard dispersion beyond what would be expected by chance. At the local scale, only a single statistically significant feature was detected: a Low–Low cluster at site SD12 (West Lujabwa; local $Ii = 0.039$, $p = 0.028$; $\log10\ \Sigma PFAS \approx 2.11$).

This isolated cold spot, rather than indicating a source, delineates an area of consistently low concentrations that is hydrologically or sedimentologically decoupled from the broader lake system. Crucially, the complete absence of any High–High or High–Low clusters implies that no dominant point-source (e.g., an industrial outfall or major river plume) exists, and that the contaminant distribution is governed instead by numerous small, diffuse inputs along the shoreline such as runoff from informal settlements and multiple small streams. These are subsequently homogenized by vigorous wind-driven wave action and sediment resuspension in addition to atmospheric deposition.

These results suggest that PFAS entering Lake Victoria undergoes substantial post-depositional mixing, resulting in a spatially fragmented pattern in which sediment concentrations at adjacent sites exhibit little to no statistical dependence. The fact that there is one major cluster (Figure 4), reinforces the observations from Moran's that there is no significant spatial structure. This random background, punctuated only by isolated deviations, underscores the need for source attribution studies to prioritize high-resolution nearshore land-use analysis rather than deterministic spatial interpolation.

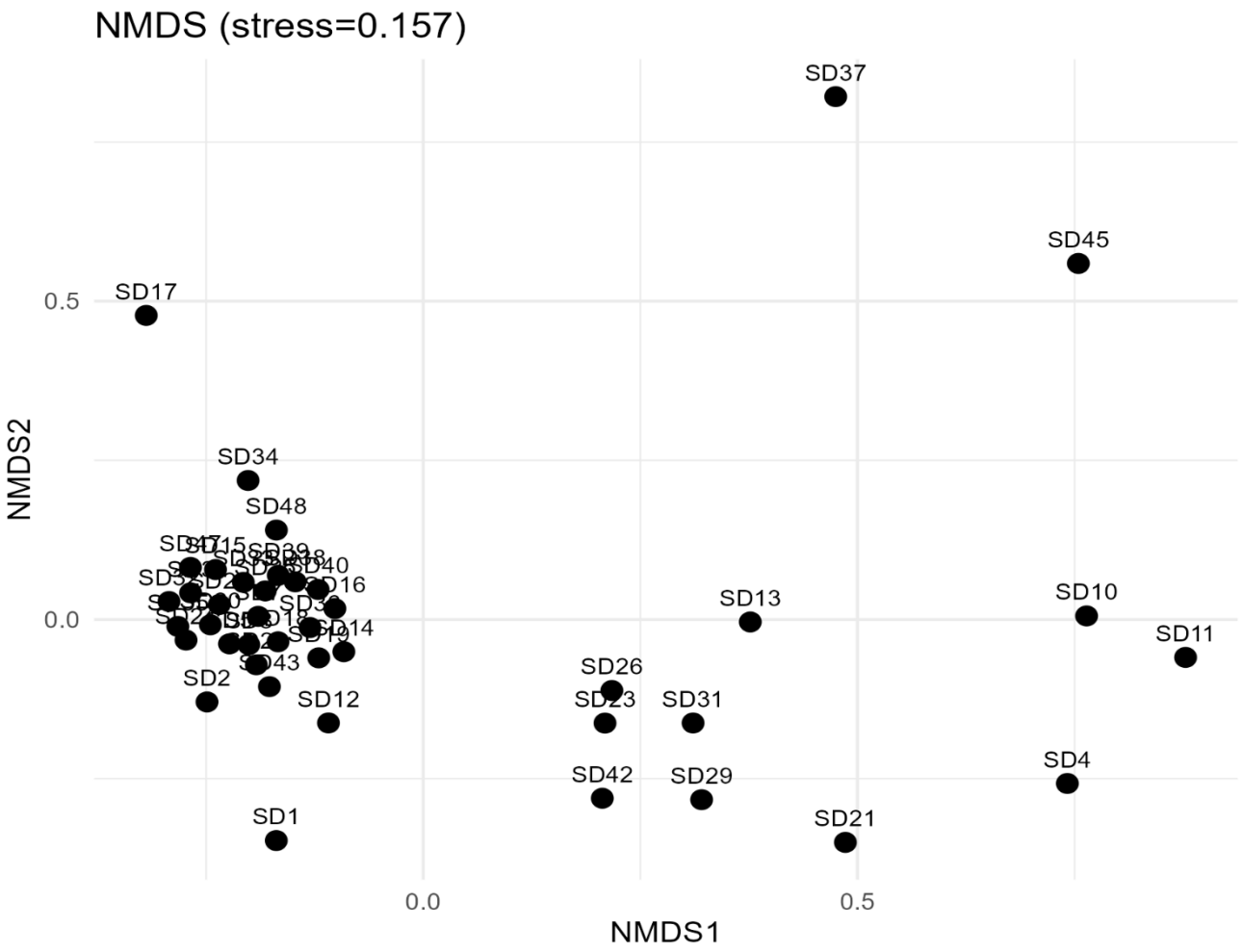

Figure 4: Nonmetric multidimensional scaling (NMDS) plot showing the similarities and dissimilarities in the September/October 2019 samples

3.2.3 PFAS Profiles in the Sediment Core

Using a previously dated core sediment collected from the open lake (0° 33' 13" S, 33° 15' 30" E) for reference (Arinaitwe et al., 2016), the analyzed core sediment (2019) collected from coordinates (0° 30' 26.28" S, 33° 15' 35.94" E) was estimated to represent deposition from 1964 – 2019 (Table S7) through linear interpolation of the reference core. Using the average sediment accumulation rate from the reference core, the 2019 surface shift was estimated to be 4.2 cm.

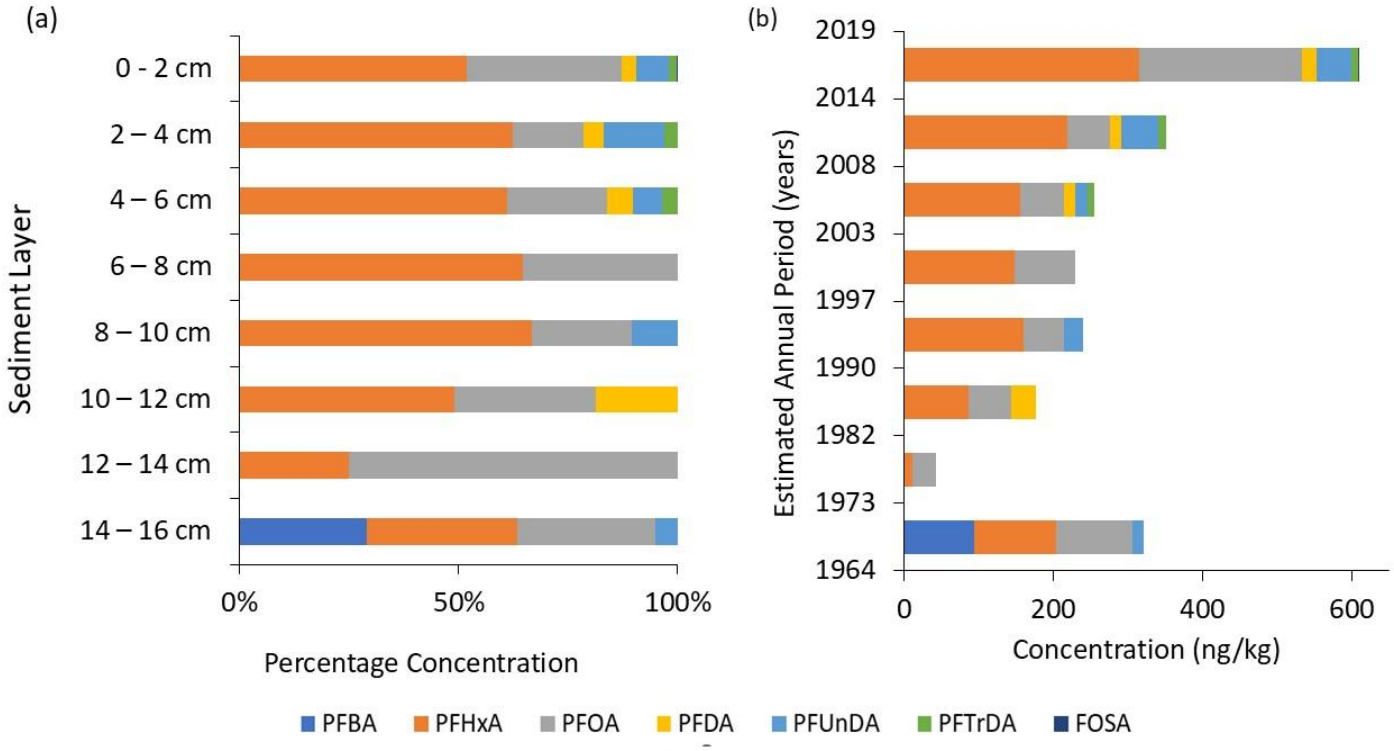

Figure 5: Relative percentage of PFAS vertical composition profiles (a) and temporal pattern of PFAS concentrations (ng/kg) dw (b) in Lake Victoria over the period of 1964-2019

Occurrence of PFAS in the core sediment showed that the PFAS concentrations varied over the period of 1964 -2019 (Figure 5). The uppermost layer of the sediment core had the highest ΣPFAS concentration

suggesting an ongoing PFAS contamination in Lake Victoria and a rise in the overall loading from the growing PFAS sources around Lake Victoria.

PFBA was detected in the 12-16 cm layer (1964 -1982). PFBA; a short-chain PFAS, has low volatility and high water solubility and thus higher vertical mobility and therefore it could have diffused through pore water movement into the deeper layers of the core sediment (Ahrens & Bundschuh, 2014a). PFBA was also found to have higher vertical mobility, partitioning into deeper sediments of Hudson River, USA (Liu & Li, 2024). Downward migration via pore water or bioturbation by benthic organisms may obscure true trends (Ahrens et al., 2009). PFHxA and PFOA were detected throughout the core sediment. PFHxA showed a clear increase over time, possibly linked to its use as a substitute for PFOA (Wang et al., 2013).

PFDA and PFUnDA had no clear trend over the years. PFDA had its highest concentration at 10-12 cm, corresponding to 1982 -1990. From 2003, PFDA, PFUnDA and PFTrDA were all quantifiable in the core sediment, a time when chemical industry had shifted to fluorotelomerization, after the global phase out of PFOS (USEPA, 2000). Fluorotelomerization produces polyfluorinated compounds which are volatile, possess the capacity for long-distance travel and break down into PFCAs through atmospheric oxidation (Ellis et al., 2004; ITRC, 2018). Moreover, long chain PFAS have a stronger affinity for organic carbon than short chain PFAS, and therefore are likely to get adsorbed on the upper layers of the core sediment limiting their mobility to deeper sediments (Ahrens & Bundschuh, 2014b). FOSA, was only found in the topmost layer (0-2 cm) in minimal concentrations (1 ng/kg dw). FOSA is biodegradable and is transformed to stable PFOS through aerobic microbial transformation (Ma & Olivares, 2025; Yan et al., 2025). Its occurrence on the topmost layer could indicate its local inputs into the lake. No clear vertical trend was observed for the total PFAS concentrations in the sediment core implying significant post depositional redistribution of PFAS through processes such as bioturbation and the diversity of PFAS shifts. However,

the increasing trend for most individual PFAS suggests their rising use and environmental release coinciding with regional urbanization and industrialization.

4. Source Identification

Diagnostic ratios and multivariate analysis were used to investigate the sources of PFAS in sediments of Lake Victoria. Diagnostic ratios of PFAS concentrations have been used to identify process based sources of PFAS in aquatic environments including sediments (Barhoumi et al., 2026; Gao et al., 2014; Guo et al., 2015). In this study, the ratios for PFOA/PFNA, PFOS/PFOA and PFHpA/PFOA without total organic carbon normalization were used for macro-level qualitative screening and relative spatial source tracking. The distinct clustering of the diagnostic ratios is shown in Figure 6.

PFOS/PFOA ratio greater than one indicates a direct PFOS source. The ratios were all below 1 except for sampling sites MSG2 and MSG 7 which had ratios of 2.09 and 124 respectively suggesting a combination of both point and diverse sources. The concentration of PFOA in these sites was less than that of PFOS, and thus, these sites had a direct source of PFOS. These sites are in Murchison Bay in the nearshore urban region. The bay is subjected to a mixture of pollutants from industrial effluents and urban storm drains from Kampala city (Arinaitwe et al., 2021). Gao et al. (2014) reported PFOS/PFOA ratios in surface marine sediments of less than 1 for most sites with few spots showing ratios >1, indicating proximity to PFOS-dominated sources. PFOS/PFOA ratios in sediments ranged from 1.7 to 6.9 at every sampling location in the Cau River, Vietnam. The location with the highest PFOS to PFOA ratio was directly influenced by wastewater effluent and the potential sources for the other sites were suggested to be domestic and industrial wastewater (Tran et al., 2022).

PFOA/PFNA ratios exceeding 1 indicate atmospheric emissions (Armitage et al., 2009). The PFOA/PFNA ratios were all above 1 in the ranges of 4.44 to 84.67 indicating that the concentrations for PFOA were all higher than for PFNA in all the sampling sites, proposing atmospheric deposition as a major source. This is in agreement with previous studies carried out in Tahiu lake where the PFOA/PFNA ratios were reported to be in the range of 1.7 to 56.8 attributing presence of secondary sources such as degradation of PFAS precursors (Guo et al., 2015) and also in soil samples from North America (Rankin et al., 2016). PFHpA/PFOA ratios have been used to establish a source tracer of atmospheric precipitation, wastewater treatment effluents and sediment accumulation (Gałęzowska et al., 2020; Simcik & Dorweiler, 2005). In this study, the PFHpA to PFOA ratios in all the sample sites were less than 1. Higher ratios above 1 would suggest atmospheric deposition as a PFHpA source (Simcik & Dorweiler, 2005)

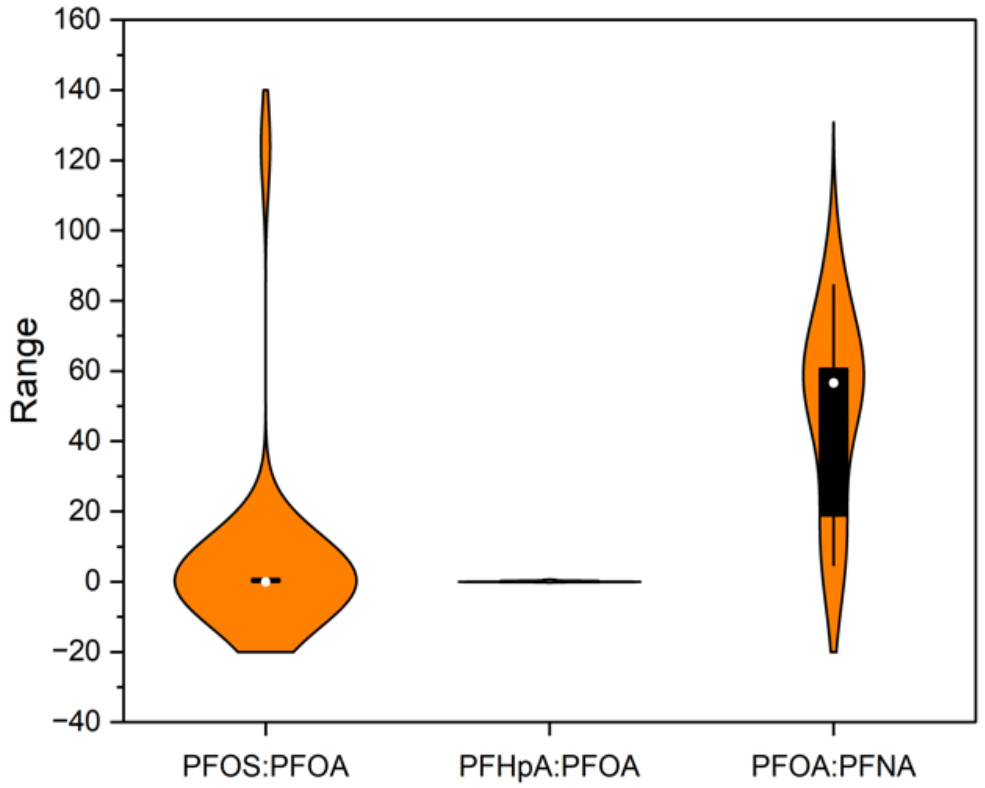

Figure 6: Violins plot showing the distinct clustering of the diagnostic ratios used for source identification in the sediment samples. The violins show the spread of the ratios, with the white dot indicating the median and the black box representing the interquartile range

Using PCA-MLR, Linear correlation and Kaiser Meyer-Olkin (KMO) values ($p < 0.000$, KMO = 0.708) revealed suitability of the model in source apportionment. Two principal components identified with the eigenvalues >1 were extracted through varimax rotation with Kaiser normalization. Multilinear regression R^2 value was 0.98. The modelled total concentrations correlated well with the actual concentrations for the total PFAS concentrations (Figure S1).

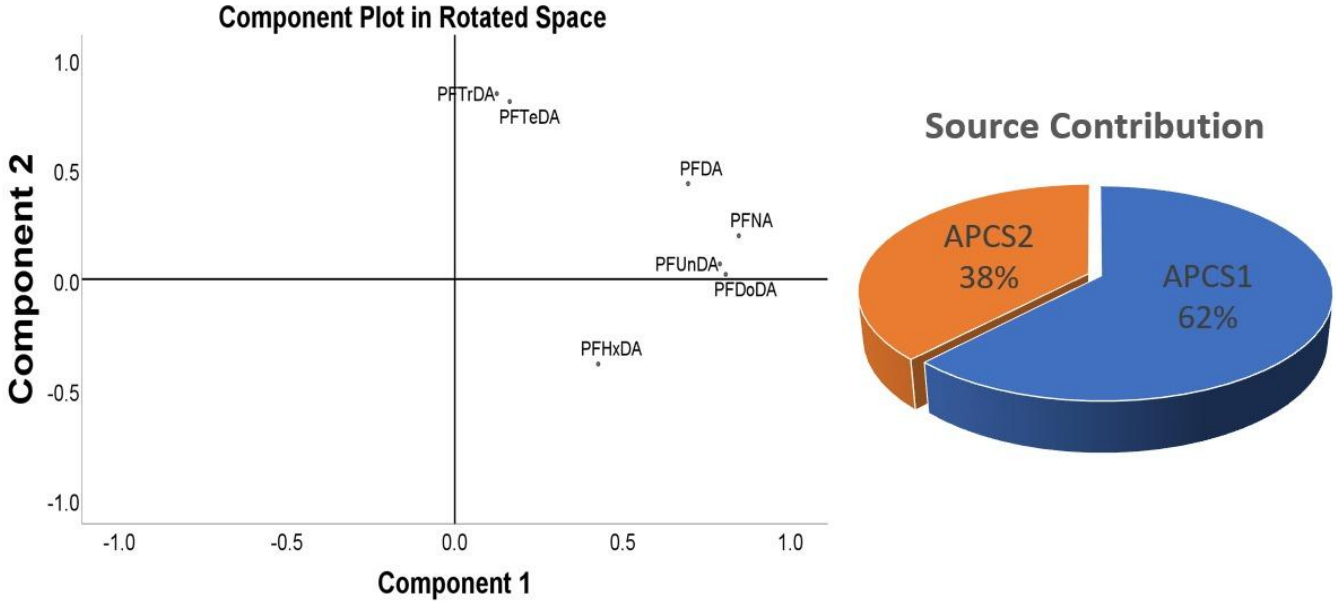

Figure 7: Component plot in Rotated space and Source Contributions for September/ October 2019 samples in PCA-MLR model

As shown in Figure 7, component 1(APCS1) had a source contribution of 62%, depicting its significant influence on occurrence of PFAS in Lake Victoria. It is characterized by high loadings of PFNA (0.846), PFUnDA (0.790), PFDoDA (0.807) and PFDA (0.695) and moderate loadings of PFHxDA (0.427). fluorotelomer olefins are oxidized to produce PFNA-based compounds, which are also utilized as processing aids in the production of fluorotelomer (Prevedouros et al., 2006). Additionally, PFNA may also be generated alongside other PFAS such as PFDA during the electrochemical fluorination (ECF) of perfluoroctane carbonyl fluoride and perfluorooctyl sulfonic fluoride as impurities. The electrochemical fluorination processes have reported higher levels of PFNA than PFDA as impurities (Reagen et al., 2007). However, it has been reported that emissions of PFNA from PFNA-based products during their lifecycle has reduced worldwide since 2002 (Group, 2003). PFNA and other homologue PFAS are not commercially produced at the vicinity of Lake Victoria. Additionally, PFNA, PFDA, PFUnDA and PFDoDA have been reported to be related to 10:2 and 8:2 fluorotelomer alcohols (Ellis et al., 2004; Kotthoff et al., 2015). Fluorotelomers are volatile and have long range transport potential and therefore can travel far from their sources through the atmosphere (Thackray et al., 2020). PFUnDA was identified in precipitation more than in and around cities (Kim & Kannan, 2007), an indication of oxidation of volatile precursors and atmospheric transport as its origin. Globally, long chain PFAS (about 2-50 tons per year) have been reported to be produced through degradation of fluorotelomers in the atmosphere (Thackray & Selin, 2017). Moderate loadings of PFHxDA (0.427) could be attributed to its low pKa, which makes it more anionic hence slow to sorb into sediments (Nguyen et al., 2020; Rayne & Forest, 2009). Therefore, Component 1 is designated as a non-point source (atmospheric deposition). Also, earlier findings by Arinaitwe et al (2021) indicate that atmospheric deposition had a source contribution of 70% and thus it's a predominant source of PFAS in Lake Victoria.

Component 2 (APCS2) had high correlations of 0.842 and 0.806 with PFTrDA and PFTeDA respectively with a source contribution of 38 %. The strong positive correlations indicate a distinct source. PFTrDA, and PFTeDA have been detected in consumer products such as skiwax products, food contact materials, personal care products and textiles (Herzke et al., 2012; Kotthoff et al., 2015). They have also been reported to occur in firefighting foams (Munoz et al., 2017). The presence of PFTeDA and PFTrDA in Lake Victoria sediments could therefore be linked to surface runoffs, landfill leachates or wastewater discharges due to poor waste disposal. PFTeDA and PFTrDA also have positive loadings on component 1, indicating that atmospheric deposition also contributes to their presence in Lake Victoria. Moreover, all the other PFAS show positive loadings for component 2 except PFHxDA which could imply a partitioning profile in the sediment-lake system. Unlike the other PFAS, PFHxDA displays a distinct behavior attributed to its low pKa making it highly sensitive to sediment chemistry (Nguyen et al., 2020).

PFAS Ecological Risk Assessment in the Sediments

Risk quotient (RQ) analysis showed significant variations in ecological risk between sampling periods and PFAS compounds (Table S8). Overall, PFBA showed remarkably high RQ values>1, making it the primary cause of ecological risk. The extremely low PNEC of PFBA (0.026 ng/kg) is the main cause of these increased findings, suggesting that even moderate environmental concentrations may pose significant ecological concerns.

PFOA had mean RQ of 0.27±0.26 (September /October 2019) and 0.88±0.73 (May 2019) with CRQ of 0.18 and 0.88 respectively posing moderate risks to benthic organisms. However, in May 2019 the RQ maximum value exceeded 2.52 indicating potential ecological effects at site P14S. PFDA in September/October 2019 had high risk with RQ>1. During September and October of 2017, PFOS showed a mean RQ of 2.33 ± 7.35, with highest values reaching 27.76, indicating localized areas of considerable

ecological concern. PFNA, PFDA and PFUnDA had moderate risks. PFTrDA, PFTeDA, PFHxDA and FOSA posed negligible risks despite their frequent detection in sediments.

6. Limitations

Spatial distribution profiles were determined from a finite number of samples, which may have provided a coarse resolution of the spatial PFAS variability in Lake Victoria. Also, since only one sediment core was used in this study, it does not indicate that the temporal deposition profiles are consistent for the whole lake as the kinetics of sedimentation and sediment resuspension mechanisms may differ across the lake. The temporal trend relied on an estimated chronology from a different core from the same part of the lake with similar bathymetry. The uncertainty around the PNEC values utilized for PFAS risk characterization is a major study constraint. The low toxicity threshold for PFBA resulted in disproportionately high-risk quotients, underscoring the assessment's sensitivity to PNEC selection and the possibility of conservative risk assessments. Therefore, comprehensive target and non-target screening of PFAS in the lake sediments as well as refined ecological risk assessment to lower the uncertainty surrounding conservative toxicity limits and increase the precision of PFBA risk characterization.

5. Conclusion

This study provided a comprehensive assessment of the occurrence, potential sources and ecological risks of PFAS in Lake Victoria sediments. PFAS occurrence in Lake Victoria indicates substantial inputs. Spatial analysis showed that PFAS occurrence in Lake Victoria is associated with the intensity of anthropogenic activities, pollution sources as well as the environmental conditions. Waterflow pathways and atmospheric deposition were identified as sources of PFAS in Lake Victoria. Despite relatively low concentrations of PFAS in the lake, the persistence, bioaccumulative potential, and toxicity of PFAS as well as their increasing levels in the core warrant concern. Ecological risk assessment showed that most

PFAS compounds posed low to moderate risks except for PFBA which exhibited elevated risk quotients. However, the high-risk estimates for PFBA should be interpreted cautiously because they could strongly be influenced by conservative toxicity thresholds used in screening-level risk assessments. Therefore, regular monitoring of PFAS and refined ecological risk assessment in Lake Victoria is recommended. Improved waste management practices are needed to mitigate contaminant inputs from urbanization. Future research should include novel PFAS (such as GenX), investigate sources and fate and correlate PFAS levels with sediment characteristics (e.g. organic carbon, clay content) to better understand bioavailability.

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