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1 Perfluoroalkyl substances (PFASs) in the Ugandan
2 waters of Lake Victoria: Spatial distribution,
3 catchment release and public exposure risk via
4 municipal water consumption

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17

18 **Abstract**

19 Perfluoroalkyl substances (PFASs) have scarcely been studied in the Lake Victoria Basin and
20 Africa in general. We investigated spatial profiles of PFASs in the Ugandan part of Lake Victoria,
21 their influxes and human exposure via drinking water. We analyzed open lake water, riverine water
22 (Rivers Kagera and Sio), urban drainage water (Nakivubo Channel), over-lake bulk atmospheric
23 deposition and municipal tap water (Kampala, Jinja and Entebbe). The average concentrations
24 (ng/L) for individual target PFASs were in the ranges of 0.08-23.8 (Nakivubo Channel), 0.01-10.8
25 (Murchison Bay), <MDL-5.38 (Kampala tap water), 0.01-3.64 (R. Kagera), <MDL-3.56 (Jinja tap
26 water), <MDL-3.35 (R. Sio), <MDL-1.96 (Entebbe tap water), <MDL-1.46 (open lake) and
27 <MDL-1.00 (atmospheric deposition). Estimated contribution of input pathways to \sum PFAS fluxes
28 into Lake Victoria was in the order atmospheric deposition > R. Kagera, > R. Sio > Nakivubo
29 Channel. Perfluorohexanoic acid (PFHxA) and perfluorooctane sulfonic acid (PFOS) had the
30 highest influx and retention estimates, respectively. Perfluoroalkane sulfonates (PFASs) were
31 mostly associated with urban drainage samples. PFASs were likely recycled from the Nakivubo
32 Channel, through the Murchison Bay, into municipal drinking water. The estimated human
33 exposure to \sum_{11} PFASs via drinking water indicated low risk of adverse health effects.

34 **Keywords**

35 Perfluoroalkyl substances; Lake Victoria; Catchment drainage; Urban discharge; Human
36 exposure; Atmospheric deposition

37 **1. Introduction**

38 Perfluoroalkyl substances (PFASs) consist of a large class of anthropogenic compounds, including
39 some of their transformation products, whose strong C-F bonds, hydro- and lipophobicity

40 properties have paved way for numerous applications in industry and consumer products, such as
41 surfactants and polymers (Buck et al., 2011). Through these applications, PFASs are emitted into
42 the environment directly and indirectly. They are soluble in water, resistant to biodegradation and
43 bioaccumulate in both aquatic and terrestrial organisms (González-Gaya et al., 2014; Müller et al.,
44 2011; Ochoa-Herrera et al., 2016). Some of these PFASs can be toxic to living organisms,
45 especially following dietary exposure (Bull et al., 2014; EFSA, 2018). The environmental
46 occurrence and fate processes for most PFASs are not yet understood properly (Wang et al., 2017).
47 Increased research interest into the environmental behavior of PFASs in recent decades has
48 resulted in various legislation and stewardship programs (Buck et al., 2011; Dauchy, 2019),
49 especially with respect to perfluorooctanoic acid (PFOA), PFOS and their salts, which are listed
50 under Annexes A and B of the Stockholm Convention on Persistent Organic Pollutants,
51 respectively. These efforts have led to a rise in production of alternative PFASs, some of which
52 are thought to be “regrettable substitutions”, such as hexafluoropropylene oxide-dimer acid
53 (HFPO-DA) which is also known as Gen X and perfluorobutane sulfonic acid (PFBS) (Ahearn,
54 2019; Reade et al., 2019).

55 PFASs are ubiquitous in the environment and, more so, in the aquatic environment (Houde et al.,
56 2011; Jian et al., 2017). They have been reported in samples from the marine environment
57 (González-Gaya et al., 2014), lakes (Gewurtz et al., 2019; Orata et al., 2009), rivers (Orata et al.,
58 2009; Zhao et al., 2015), atmospheric deposition (Gewurtz et al., 2019) and drinking water
59 (Dauchy, 2019). Waste water treatment plants (WWTPs) are also known gateways of these
60 chemicals into the aquatic environment (Petrie et al., 2015) due to a concentrated use of PFAS-
61 containing consumer products in urban areas. PFASs have also been found in landfill leachate
62 (Gallen et al., 2017; Lang et al., 2017) and in soil and groundwater impacted by aqueous film-

63 forming foams (Barzen-Hanson et al., 2017; Houtz et al., 2013). The high solubility of some
64 PFASs in water calls for more robust water treatment procedures for municipal water grids
65 although this presents significant challenges for removal of highly polar PFASs, also known as
66 “mobile” PFASs (Reemtsma et al., 2016; Schulze et al., 2019). Thus, municipal drinking water is,
67 potentially, a major pathway for human-exposure to mobile PFASs, along with dietary exposure
68 to contaminated food (Chen et al., 2019; Dauchy, 2019).

69 Although PFASs are globally prevalent in the environment, there is a conspicuous lack of data
70 from PFAS measurements from the African aquatic environment. The occurrence of PFASs in
71 Lake Victoria and its basin has not been adequately studied. A few studies have reported PFASs
72 in samples from Winam Gulf of the lake (Chirikona et al., 2015; Orata et al., 2008; Orata et al.,
73 2009), in Nakivubo Channel (which drains Kampala city center in Uganda) and in the lake’s
74 northern wetlands near Kampala (Dalahmeh et al., 2018). Rising urbanization in the lake’s
75 watershed, coupled to poor waste management systems, increasingly expose the lake to chemical
76 pollution. There is a need for a better understanding of the sources and input pathways of PFASs
77 to the lake, along with other chemical contaminants, and their fate processes in the lake so as to
78 facilitate better management of the lake’s ecosystem. To this end, we studied spatial distribution,
79 atmospheric deposition, as well as riverine and urban drainage influxes of selected PFASs into the
80 Ugandan part of the lake. We sought to understand the relative contribution of the influx-pathways
81 to PFAS loading into the lake. We also analyzed municipal piped water, relative to the lake water,
82 so as to assess the efficiency of PFAS removal during the water treatment stage and to get an
83 insight into potential PFAS exposure associated with drinking municipal grid water in the selected
84 urban areas of Uganda’s Lake Victoria watershed.

85 **2. Materials and Methods**

86 **2.1. Study site and sample collection**

87 Maps of Lake Victoria, Kampala and the sampling sites are shown in Figure 1.



88 **Figure 1.** Sampling sites: (A) Google Earth map of the Ugandan part of Lake Victoria showing
89 sampling points. Red markers are for riverine water sampling points; (B) Google Earth map of part
90 of Kampala showing the sampling points in Murchison Bay (green markers) and along the
91 Nakivubo Channel (NCB and NCA are sampling points before and after the waste water treatment
92 plant, respectively (red markers)).
93

94 Approximately 500 mL water samples were collected weekly from Nakivubo Channel (which
95 drains Kampala city center into Murchison Bay of Lake Victoria) and from Rivers Kagera and Sio.
96 River Kagera was accessed at Kasensero landing site, Kyotera District in Uganda while River Sio
97 was accessed via Majanji landing site (near Berkeley Bay, Lake Victoria), Busia District, Uganda.
98 The samples from Nakivubo Channel were collected at two points located before and after the inlet
99 from the National Water and Sewerage Corporation’s WWTP. Tap water samples were collected
100 monthly from different sites of the municipal piped-water grids of Kampala (Nakasero (N=4),
101 Bugolobi (N=4), Makindye (N=4) and Kansanga (N=4)), Entebbe (N=3) and in Jinja at Bugembe
102 (N=4) and at the Uganda National Fisheries Resources Research Institute (NaFIRRI, N=4). The

103 municipal raw water pumping station for tap water production for Kampala City is located in the
104 Murchison Bay south of the inlet of the Nakivubo Channel (Figure S1). The pumping station for
105 Jinja Municipality is situated at Masese on the Napoleon Gulf shoreline while the one for Entebbe
106 Municipality is located on the Entebbe Bay shoreline (Figure S1). Atmospheric bulk deposition
107 samples were collected at Bukasa Island, a site chosen to represent the general over-lake
108 atmospheric deposition. All these samples were collected between 25th February 2019 and 22nd
109 June 2019, using polyethylene bottles. Bulk deposition samples were collected using a simple set
110 up of a large polypropylene bottle and a funnel (Figure S2). Open lake water samples were
111 collected aboard *RV Hammerkop* in May 2019, the research boat of NaFIRRI. The samples were
112 transported in cooler boxes to the Department of Chemistry of Makerere University, Kampala for
113 processing. More details about the samples are given in Table S1.

114 **2.2. Sample preparation**

115 **2.2.1. Chemicals and reagents**

116 We used native standards of 14 PFASs targeted for analysis (Table S2) and isotopically labelled
117 compounds (standard mix MPFAC-MXA) and single labelled compounds; perfluoropentanoic
118 acid (M3-PFPeA), perfluoroheptanoic acid (M4-PFHpA), perfluorooctane sulfonamide (M8-
119 FOSA), all from Wellington Laboratories, Ontario, Canada. High purity nitrogen and argon gases
120 (>99.999%) were used. The organic solvents (UPLC/MS grade) and ammonium acetate (>99.9%)
121 (BioSolve Chemicals, Valkenswaard, The Netherlands) used were of highest commercial purity
122 and were used as received. Purified “Milli-Q” water (Merck Millipore, Darmstadt, Germany) was
123 used for sample preparation and in the UPLC eluent system.

124 **2.2.2. Solid Phase Extraction (SPE) of the water samples**

125 The SPE extraction of the water samples was done at the Department of Chemistry of Makerere
126 University, Kampala, in Uganda. The water samples were filtered (Glass microfibre filters, 0.7 μ m
127 particle retention, 47 mm diameter, LABSOLUTE). This was necessary to prevent clogging of the
128 SPE cartridges during enrichment of water samples, especially given that some samples had a lot
129 of suspended particulate matter. For each filtered sample, a sub sample of 250 mL (250 g) was
130 weighed into a polypropylene bottle and spiked with surrogate standard (1.0 ng of each compound)
131 and agitated to mix. Solid Phase Extraction (SPE) cartridges (CUQAX256, C8+quaternary amine,
132 500 mg – 6 mL, United Chemical Technologies, UCT, Bristol, PA, USA) were washed with 24
133 mL of a solution of 2% 1-methylpiperidine in a methanol/acetonitrile 80:20 v/v mixture. The
134 spiked water samples were then extracted (after approx. one hour's time from spiking) onto the
135 washed SPE cartridges, which were thereafter dried with nitrogen gas. The dry enriched SPE
136 cartridges were wrapped in aluminum foil and shipped to the Helmholtz Centre for Environmental
137 Research-UFZ, Leipzig in Germany for further processing and analysis.

138 **2.2.3. Elution of SPE cartridges and analysis**

139 The procedure for the elution of the dry enriched SPE cartridges was an adaptation of a previously
140 published method (Filipovic et al., 2015). The cartridges were washed with 1 mL of methanol and
141 thereafter, eluted with 8 mL of a solution of 2% 1-methylpiperidine in a methanol/acetonitrile
142 80:20 v/v mixture. The eluent was evaporated to dryness with nitrogen gas on a 40 °C water bath.
143 The residues were re-dissolved in 150 μ L of methanol and 100 μ L of 4 mM aqueous ammonium
144 acetate. The extract was filtered (Minisart RC4 0.2 μ m RC-membrane Filter, Sartorius Stedim Lab
145 Ltd., Stonehouse, United Kingdom) and kept at -20 °C in a plastic vial until analysis. Prior to
146 analysis, the samples were allowed to thaw and vortex-agitated. They were analyzed on an Acquity
147 Ultra Performance Liquid Chromatograph, equipped with a trapping column upstream of the

148 injector and coupled to a Xevo TQ-S Tandem Mass Spectrometer (Waters, Eschborn, Germany).
149 Details of the instrumental method are given in Table S3. The target analytes and their quantifier
150 MRM transitions are given in Table S2, while those of the mass labelled standards have been given
151 elsewhere (Arinaitwe et al., 2020).

152 **2.3. Quantification, quality control and data analysis**

153 All samples, from collection to analysis, were handled with polyethylene or polypropylene
154 materials. Procedure blanks were analyzed along with the samples to evaluate background
155 contamination from the entire method and absolute recoveries were determined for all internal
156 standards (see “Method Performance” section of the supporting information for a discussion of the
157 results). Multilevel calibration (7-point calibration solutions in the concentration range of 0.01 –
158 5.0 ng/mL) was used for analysis. Two batches of SPE cartridges were used across the sampling
159 campaign. Therefore, SPE batch-based method detection limit (MDL) calculation was done (Table
160 S2). Generally, the MDL profiles for the two SPE batches were not significantly different
161 ($P=0.435$). All sample data was blank corrected with the average procedure blank level. Thus, the
162 MDL was determined from the procedure blanks as 3 standard deviations. In the procedure blanks,
163 where there were no detections, the average concentration equivalent to signal-to-noise ratio of 3
164 was used as the MDL. To include as many “real” values as possible (i. e. from integrated signals)
165 in statistical calculations, we quantified all data above MDL. All data values below MDL were
166 designated as “non-detects” and for statistical analyses, they were assigned values equivalent to
167 MDL/2. Back air trajectories were computed using NOAA’s HYSPLIT Model (Draxler and Rolph,
168 2011; Rolph, 2011). Estimation of annual influxes of chemicals into the lake through River Kagera,
169 River Sio and over-lake atmospheric deposition were based on long term (1950-2004) average
170 water inflows (261, 11.3, and 3610 m³/sec, respectively) (EAC, 2006) and a 1997-1999 average

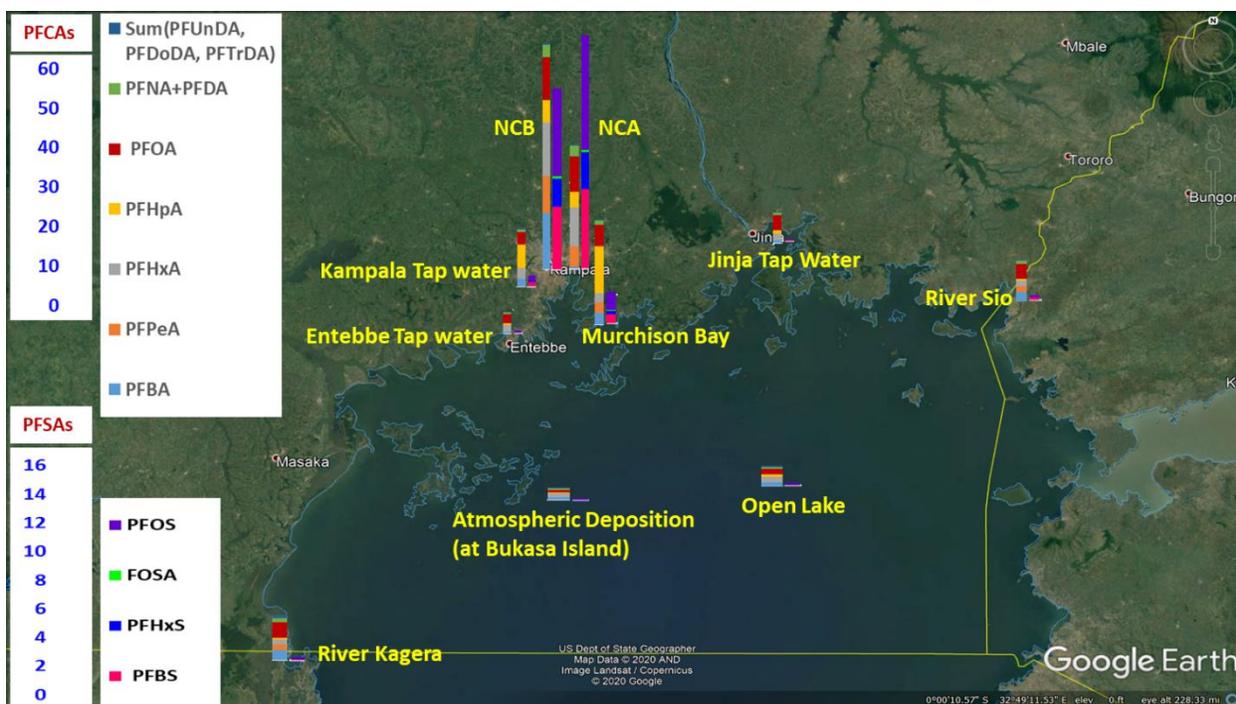
171 flow for the Nakivubo Channel of 0.63 m³/sec (Kayima et al., 2008) since we could not find recent
172 flow data for the channel. Apart from vaporization, River Nile is the only other outflow pathway
173 of water from Lake Victoria. Therefore, PFAS outflow estimates were based on a long term (1950-
174 2004) average flow of 1058 m³/sec for river Nile (EAC, 2006). In order to estimate PFAS exposure
175 via consumption of tap water associated with the samples used in this study, we assumed average
176 heights (for Uganda) of 166 cm for men and 157 cm for women (Roser et al., 2020). The equivalent
177 average body weights based on the Broca Index (Nahler, 2009) are 66 and 57 kg, respectively. An
178 average daily consumption of 2 L of tap water was assumed.

179 Descriptive statistics and outlier detection were done using Microsoft Excel while other statistical
180 analyses were done using SigmaPlot 13.0. Outliers were identified as values outside the
181 interquartile range (IQR) using an IQR multiplier of 1.5 ($X_i > Q_3 + 1.5 \cdot IQR$ or $X_i < Q_1 - 1.5 \cdot IQR$,
182 where X_i is the outlier data point). Data sets were compared using Pearson correlations, the t-test
183 and One-Way Analysis of Variance (ANOVA); the Shapiro-Wilk test was used to test normality
184 at an overall significance level of 0.05. If a dataset did not pass the normality test, Kruskal-Wallis
185 One-Way ANOVA on ranks and the Tukey Test (for comparison of many groups) were
186 additionally performed. During the t-test, if the dataset failed the normality or equal variance test,
187 the Mann-Whitney rank sum test was performed.

188 **3. Results and Discussion**

189 **3.1. PFAS concentrations and patterns in the different sample sets**

190 The overall summary of PFAS concentrations for all sample sets is given in Table S5 and Figure
191 2. Since all water samples were filtered prior to spiking with surrogate internal standards, all
192 concentrations given in this study refer to the dissolved phase only.



193

194 **Figure 2.** Spatial distribution of mean PFAS concentrations (ng/L); NCB and NCA represent
 195 samples collected before and after the inlet of the municipal WWTP effluent, respectively. For
 196 individual PFAS acronyms see Table S2.

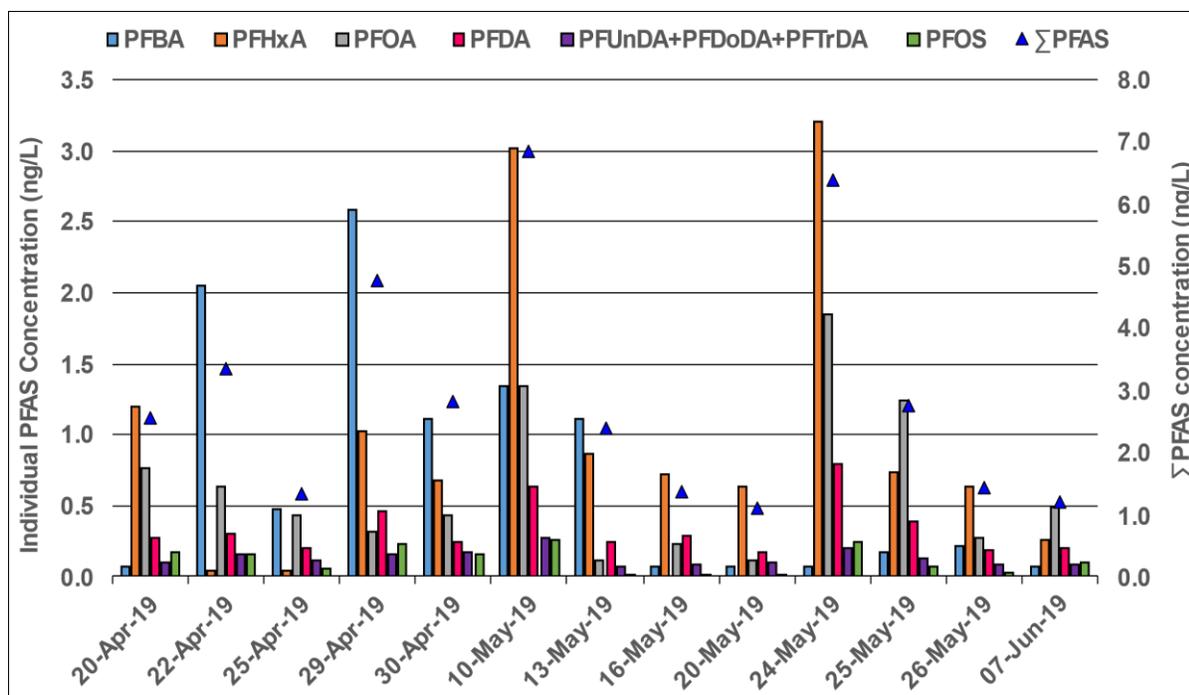
197 In general, the average concentration of total PFASs (Σ PFAS) in the sample sets decreased in the
 198 order Nakivubo Channel (66.7 ng/L) > Murchison Bay (26.6 ng/L) > Kampala tap water (14.3
 199 ng/L) > River Kagera (10.7 ng/L) > River Sio (10.0 ng/L) > Jinja tap water (7.27ng/L) > Open
 200 lake water (5.73 ng/L) > Entebbe tap water (5.17 ng/L) > Atmospheric precipitation (3.14 ng/L).

201 This order reflects the influence of proximity to urban emission sources on the water quality. The
 202 Nakivubo Channel and Murchison Bay are exposed to a cocktail of chemicals from industrial,
 203 artisanal and domestic sources in the Kampala City area via direct or indirect release into the
 204 channel. The high concentration of Σ PFAS in the Kampala tap water sample set (relative to Jinja
 205 and Entebbe sample sets) and the significant Pearson correlation of its average PFAS profile with
 206 the one of Murchison Bay sample set ($r = 0.938$, $p < 0.001$) suggests a legacy contaminant load

207 from the bay. This is plausible given that the Kampala municipal water is pumped from the
208 Murchison Bay, downstream of the inlet from Nakivubo Channel (Figure S1). The PFASs in the
209 Nakivubo Channel and Murchison Bay sample sets were one or two orders of magnitude higher
210 than in the other sample sets. This most likely indicated local emission sources in the Kampala
211 catchment. The similarity of the PFAS concentrations in sample sets collected from points before
212 and after the WWTP suggests negligible input from the plant.

213 **3.1.1. Atmospheric bulk deposition**

214 The average concentration of individual PFASs for all atmospheric samples ranged from <MDL –
215 1.00 ng/L with PFHxA being the most abundant (Table S5). Of all the frequently detected PFASs,
216 perfluorobutanoic acid (PFBA), which was the second most abundant PFAS, had no significant
217 Pearson correlation with any other PFAS (Table S6). This was a likely indication of a different
218 emission source of PFBA in the samples. The strongest and most significant Pearson correlations
219 were the ones between perfluorodecanoic acid (PFDA) and PFHxA ($r = 0.899$, $p < 0.001$) and
220 PFDA and PFOA ($r = 0.852$, $p < 0.001$). With the exception of PFBA, PFOS had significant
221 correlations to all the frequently detected PFASs. These significant correlations between short and
222 long chain PFASs potentially suggested common emission sources of these compounds in the
223 samples, except for PFBA. In atmospheric samples, PFAS occurrence is a result of multiple
224 emission pathways, such as direct release from manufacturing sites, spray from water bodies and
225 other wet surfaces or environmental dispersion of precursors (Prevedouros et al., 2006). Therefore,
226 significant correlations between PFAS profiles could indicate shared emission and/or dispersion
227 pathways.



228

229 **Figure 3.** Temporal concentration profiles of the most abundant PFASs in bulk atmospheric
 230 deposition samples.

231 Regarding temporal changes, PFBA was most abundant in samples collected in April (Figure 3).

232 Thereafter, PFHxA predominated with a subsequent increase of the PFOA fraction in late May

233 and early June samples. In order to have more insight into this profile, 72-hour backward air mass

234 movement trajectories were computed for each of the sampling dates (Figure S4). Samples in

235 which PFBA was predominant (22nd – 30th April 2019) were mostly associated with very slow air

236 mass movement characterized by long residence time within the lake’s basin. Conversely, the

237 samples in which PFHxA and PFOA predominated (and PFBA was diminished) were associated

238 with fast moving easterly and south-easterly air masses from as far as the Indian Ocean coast. It

239 has been indicated that the precipitation in East Africa is significantly influenced by convection

240 currents from the Indian Ocean (Vuille et al., 2005). It has also been suggested that volatile

241 precursor PFASs such as fluorotelomer alcohols and perfluoroalkane sulfonamides could undergo

242 atmospheric degradation to form PFCAs and PFSAs (Schenker et al., 2008), which are then
243 scavenged by precipitation. Therefore, these trajectories suggest that long range atmospheric
244 transport of PFAS-laden aerosols, coupled with possible atmospheric precursor degradation, from
245 as far as the Indian Ocean, was likely to be a major factor in the PFAS profiles. However, this was
246 least likely to be the case for the PFBA profile in which local emissions from the lake's basin,
247 including lake-spray aerosols from the lake surface, were suspected to play a bigger role. The
248 predominance of PFHxA in the atmospheric samples could also be a legacy of increased global
249 production and use of short-chain PFASs as substitutes for long chain PFASs (Li et al., 2020). The
250 air trajectories also suggest that emissions from regional urban centers such as Nairobi, Arusha
251 and Dar es Salaam (located east and south east of Lake Victoria) could be airlifted and
252 subsequently deposited in Lake Victoria and its watershed.

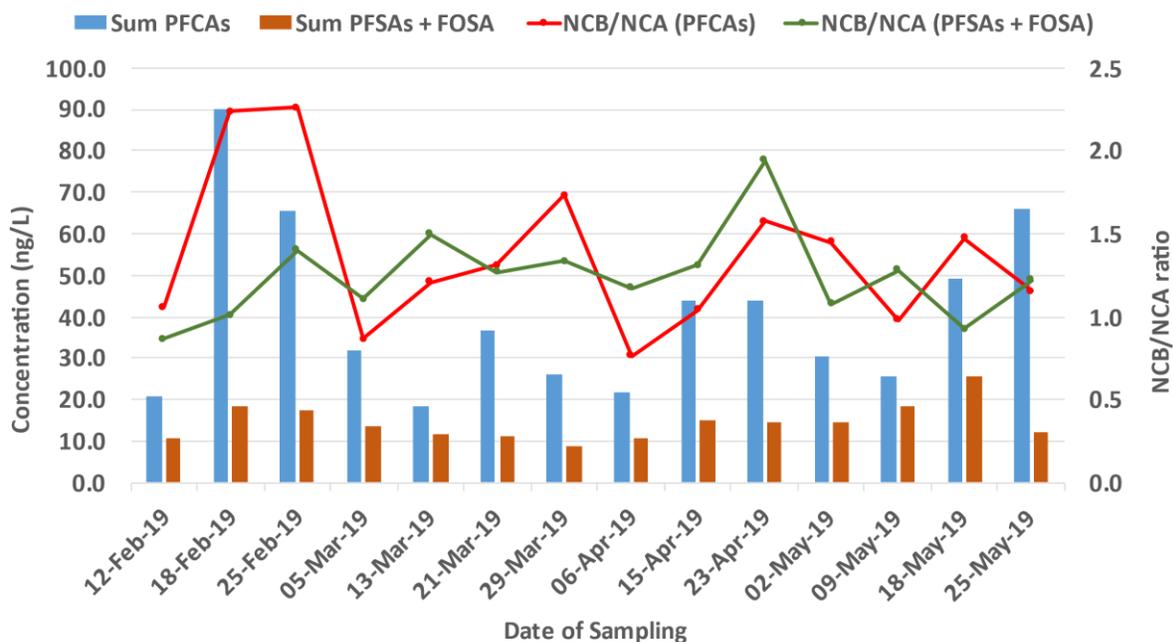
253 **3.1.2. Riverine concentration profiles**

254 The PFCAs were generally higher than PFSAs in both Rivers Kagera and Sio in all samples (Figure
255 S5). The average \sum PFCA concentrations were about 30 and 58 times higher than \sum PFSAs for the
256 River Kagera and River Sio sample sets, respectively. PFOA dominated the PFAS profiles of the
257 riverine sample sets with overall average concentrations of 3.6 and 3.4 ng/L for Rivers Kagera and
258 Sio, respectively (Table S5 and Figures S5 and S6). In either riverine sample set, there was no
259 significant Pearson correlation between \sum PFCAs and \sum PFSAs ($r = -0.306$ and -0.290 for River
260 Kagera and River Sio sample sets, respectively), possibly indicating different PFAS input
261 pathways and fate processes. However, the two rivers had similar average concentration of \sum PFAS
262 and highly correlated PFAS profiles (Pearson coefficient $r = 0.996$, $p < 0.001$) despite having very
263 different catchment characteristics. River Kagera has a wide drainage basin from as far as Burundi,
264 through Rwanda, parts of south-western Uganda and parts of north-western Tanzania. It is thus

265 exposed to urban emissions and, possibly, significant atmospheric input from precipitation.
 266 Moreover, River Kagera has much higher water volume than River Sio. If atmospheric deposition
 267 is the major contributor of PFASs in the two rivers' catchments (see section 3.2.), then there is
 268 chance that this explains the similarity of the profiles and concentrations of these chemicals in the
 269 two sample sets. The general predominance of the PFCAs relative to the PFSA s suggests higher
 270 use of PFCA-containing or PFCA precursor-containing consumer products in the region or more
 271 PFCA precursors (e. g. volatile fluorotelomers) transported by long-range atmospheric transport
 272 into the region.

273 3.1.3. Concentration profiles in the Nakivubo Channel samples

274 The Nakivubo sample set had the highest concentrations of PFASs (Table S5). The temporal
 275 concentration data for this sample set is shown in Figures 4 and S7. The Σ PFCA concentration
 276 profile was dominated by short chain PFCAs, especially in February 2019 and May 2019.



277

278 **Figure 4.** Temporal concentration profiles of PFASs in the Nakivubo Channel sample set and their
279 NCB/NCA ratios. NCB and NCA represent samples collected before and after the inlet of the
280 municipal WWTP effluent, respectively.

281 With the exception of PFBA, the NCB/NCA ratios of individual PFCAs had a near-linear decrease
282 with an increase in chain length, but this was not the case with PFSA's whose NCB/NCA ratio was
283 fairly constant at about 1.2. This was likely due to matrix interference in NCA samples associated
284 with effluent from the WWTP. Based on the NCB/NCA ratio profile, the WWTP does not increase
285 the PFAS burden in the Nakivubo Channel, yet the latter has the highest PFAS concentrations of
286 all sample sets. Thus, other sources, including storm water and/or surface runoff, must likely
287 contribute to the relatively high concentrations in the Nakivubo Channel samples.

288 Rainfall data recorded in Kampala for three days building to the sampling date D, inclusively (i.e.
289 D, D-1, D-2), and the three-day average [i.e average (D, D-1, D-2)] was used to compute Pearson
290 correlations between PFAS concentrations in Nakivubo Channel samples and rainfall profiles
291 (Table S7). The sampling day rainfall profile was significantly correlated with profiles of PFOA
292 ($r = 0.680$, $p = 0.007$) and PFBS ($r = 0.617$, $p = 0.019$). The 3-day average rainfall profile was
293 significantly correlated with profiles of PFOA ($r = 0.544$, $p = 0.045$), PFHxA ($r = 0.630$, $p = 0.016$)
294 and PFHpA ($r = 0.704$, $p = 0.005$). Since the Nakivubo Channel drains storm water from the
295 Kampala city center, these significant correlations are likely to indicate rainfall influence on the
296 observed PFOA, PFBS, PFHxA and PFHpA profiles in samples from the channel, most likely as
297 surface runoff during rainfall events.

298 **3.1.4. Concentrations in lake water samples**

299 The Murchison Bay samples had the highest concentrations among the samples taken from the
300 lake (Table S5, Figure S8). Samples from points P1, P5 and Murchison Bay had significantly
301 elevated PFHpA. Point P1 is impacted by inflow from River Sio while P5 and Murchison Bay are
302 impacted by emissions from Kampala City. The PFAS profiles of P5 and Murchison Bay had the
303 highest Pearson correlation ($r = 0.935$, $p < 0.001$) (Table S8). Point 5 is located at the gateway of
304 Murchison Bay into the open lake (Figure 1). Thus, the lower concentrations at Point 5 could have
305 been due to dilution relative to Murchison Bay levels. All the lake sample profiles were
306 significantly correlated to the average atmospheric deposition profile and like in atmospheric
307 deposition samples, PFBA, PFHxA, PFOA and PFDA were the most abundant PFASs. This was
308 more so at Point 4, the furthest into the open lake, which had the highest correlation with the
309 atmospheric sample profile ($r = 0.967$, $p < 0.001$). This is a strong suggestion of significant
310 influence of atmospheric deposition in the open lake PFAS profile (see discussion above and in
311 section 3.2). In general, for the open lake (excluding Point 5 and Murchison Bay) the average
312 concentration of \sum PFCA (5.02 ng/L) was significantly higher than for \sum PFSA (0.10 ng/L). Also
313 this is expected in light of the discussion of atmospheric deposition (section 3.1.1) and influx
314 profiles (section 3.2).

315 **3.1.5. PFAS profiles in Kampala, Jinja and Entebbe municipal water grids**

316 The overall average concentrations of the individual PFASs and their temporal profiles in tap water
317 from Entebbe, Jinja and Kampala municipal grids are given in Table S5 and Figure S9,
318 respectively. In general, Kampala samples had the highest average concentrations of individual
319 PFASs (<MDL-5.3, <MDL-3.56 and <MDL-1.96 ng/L for Kampala, Jinja and Entebbe,
320 respectively). As already indicated in Section 3.1, Kampala tap water is pumped from the
321 Murchison Bay, which is heavily impacted by emissions from Kampala city centre via the

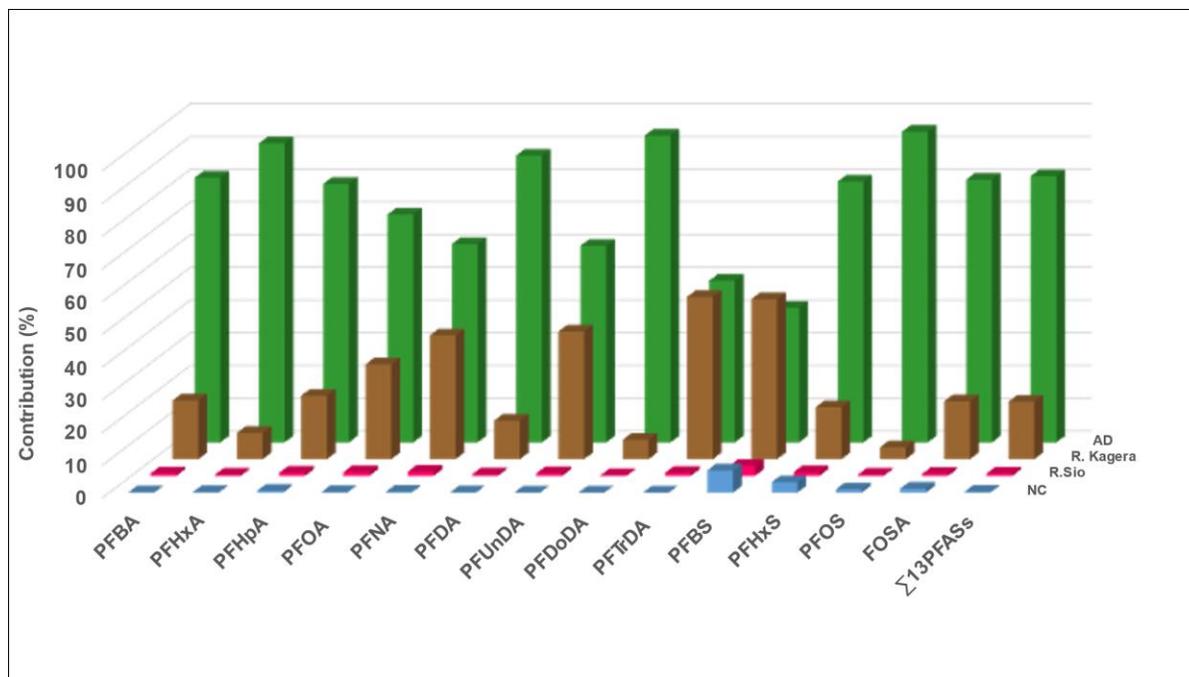
322 Nakivubo Channel and via direct surface runoff. To a lesser extent, this also happens for Jinja
323 whose tap water is pumped from the Napoleon Gulf. This gulf is the gateway to the only outflow
324 route (River Nile) for the entire lake. It is exposed to PFAS loads from the open lake, Murchison
325 Bay and part of Jinja urban center, albeit with much more dilution than in the Murchison Bay. The
326 Entebbe Bay is less impacted by urban activity and is not as naturally sheltered from the open lake
327 as are Murchison Bay and Napoleon Gulf (see Figures 2 and S1). It is, therefore, conceivable that
328 the above general PFAS profile in tap water is related to the relative PFAS exposure risks at the
329 pumping stations in the lake. There was a general increase in the PFAS concentrations in the
330 Kampala samples during the sampling period, peaking in April where PFHpA was more than twice
331 the February levels. This indicated variability in the PFAS concentrations in tap water over time.
332 In the Entebbe samples, the PFHxA concentration increased as the PFOA concentration decreased
333 from March to May. During this time, the average monthly rainfall also increased (1.4, 5.6 and 6.5
334 mm for March, April and May, respectively). In light of (i) the similarity of the average \sum PFAS
335 concentration between Entebbe tap water and open lake water (see section 3.1), (ii) the highly
336 significant correlation of the PFAS profiles between Entebbe samples and either of open lake water
337 and atmospheric deposition samples ($r = 0.978$ and 0.904 , respectively, $p < 0.001$) and (iii) the
338 increase in the proportion of PFHxA, from April to May, in atmospheric samples (see section
339 3.1.1), it is likely that the Entebbe samples are a reflection of the lake profile with insignificant
340 PFAS removal by the water treatment process.

341 **3.2. Relative importance of PFAS input pathways**

342 The estimated influxes for a sum of 13 target PFASs (\sum_{13} PFAS) were 355, 76.7, 3.11 and 0.97
343 kg/yr for atmospheric deposition over the lake, River Kagera, River Sio and Nakivubo Channel,
344 respectively (Table S9 and Figure 5). The highest combined influx estimates were 125, 103, 102

345 and 44 kg/yr for PFHxA, PFOA, PFBA and PFDA. Atmospheric deposition had the highest fluxes
346 for individual PFASs with the exception of PFBS and PFTrDA for which River Kagera was the
347 major influx pathway. The PFOS influx from the Nakivubo Channel (0.14 kg/yr) was significantly
348 higher than that of rivers, further indicating that urban emissions were likely to be more significant
349 PFOS contributors to the lake than riverine discharge. Assuming River Nile to be the only
350 significant outflow pathway for the PFASs, the estimated influx for 11 target PFASs detected in
351 the open lake ($\sum_{11} \text{PFAS}$) was about twice their outflow, representing a net retention of just over
352 50% of the incoming PFAS load. With the exception of PFUnDA, PFTrDA and PFBS, the rest of
353 the eight PFASs were estimated to be retained, PFOS having the highest retention estimate (Table
354 S9). These results suggest a rapid increase of PFASs in Lake Victoria. In light of this trend, it is
355 worth noting that the net retention of some PFASs could be due to uptake into other compartments
356 of the lake's aquatic ecosystem, such as bioaccumulation in aquatic organisms or sedimentation.
357 For instance, PFOS, PFDA and PFUnDA have been shown to dominate the PFAS profile in fish
358 from the same lake (Arinaitwe et al., 2020) while the predominance of PFOS in sediment of other
359 water bodies has been reported elsewhere (Seo et al., 2019; Wang et al., 2019). Uncertainties
360 associated with these flux estimates include: (i) Atmospheric fluxes could have been overestimated
361 if the measured concentrations involved significant lake-spray ("environmental recirculation"),
362 especially for the short chain PFASs. (ii) Excluding the Nakivubo Channel, this study has
363 investigated Rivers Kagera and Sio which account for about 34% of the surface water flow into
364 the lake (Commission and GRID-Arendal, 2017). The rest of the catchment drainage, particularly
365 from the Tanzanian and Kenyan parts of the catchment, along with urban emissions from there,
366 could be significant input pathways of additional PFAS loads that have not been catered for in this
367 study. (iii) As mentioned earlier, it is possible that the total catchment drainage influxes are

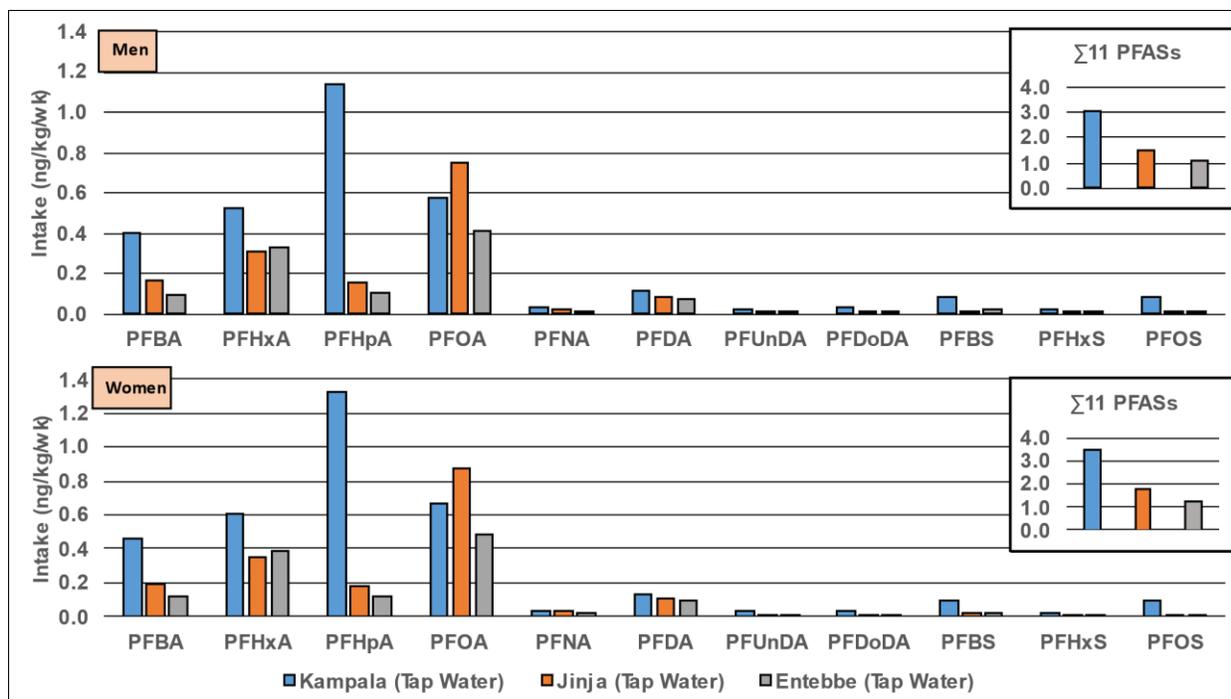
368 underestimated, especially for long chain PFASs, due to filtration of the samples and thus removal
369 of particle-bound PFASs.



370
371 **Figure 5.** Relative contribution (%) of input pathways to the estimated total PFAS flux. AD =
372 Atmospheric Deposition; NC = Nakivubo Channel.

373 3.3. Public exposure risk through tap water consumption

374 There are no advisories for PFASs in drinking water for Uganda. Internationally, drinking water
375 health advisories for PFASs have mainly focused on PFOA and PFOS. The USEPA has recently
376 published PFOA and PFOS drinking water health advisories of a lifetime exposure of 70 parts per
377 trillion (70 ng/L) down from 2009 provisional health advisories of 400 and 200 ng/L for PFOA
378 and PFOS, respectively (USEPA, 2009; USEPA, 2016). Comparatively, the average ΣPFAS
379 concentration in the Uganda tap water samples (Table S5) is less than this lifetime health advisory.



380

381 **Figure 6.** Estimated weekly exposure (ng/kg bw/wk) to PFASs via drinking municipal water for
 382 an average Ugandan.

383 The estimated weekly intake values (ng/kg bw/ wk) for PFASs are presented in Figure 6. The
 384 estimated Σ PFAS intake was 3.27, 1.65 and 1.17 ng/kg bw/ wk (or 0.47, 0.24 and 0.17 ng/kg bw/d)
 385 from Kampala, Jinja and Entebbe tap water, respectively. These estimates are lower than the
 386 reference doses instituted by the USEPA and other US provincial departments for PFOA and PFOS
 387 towards various toxicological endpoints (Cordner et al., 2019). These estimates were also less than
 388 the tolerable weekly intake thresholds of 6 and 13 ng/kg bw/wk, recommended for PFOA and
 389 PFOS, respectively, by the European Food Safety Agency (EFSA, 2018). The estimated Σ_4 PFAS
 390 intake (for PFOA, PFNA, PFHxS and PFOS) in men (0.71, 0.80 and 0.45 ng/kg bw/wk) and
 391 women (0.83, 0.92 and 0.52 ng/kg bw/wk) for Kampala, Jinja and Entebbe tap water, respectively,
 392 were also below the tolerable weekly intake threshold of 8 ng/kg bw/wk recently proposed for
 393 these 4 PFASs by the same agency (EFSA, 2020). These results did not indicate a current prevalent

394 adverse health risk associated with consumption of the tap water. However, if levels of PFASs in
395 the Lake Victoria environment are increasing as suggested in the discussion above, health advisory
396 limits for drinking water may be exceeded in the future.

397 **3.4. Comparison with similar studies elsewhere**

398 There remain very limited measurements of PFASs from the Lake Victoria basin and Africa in
399 general. We compared results from this study with a few recently published studies from
400 elsewhere, including a couple of studies from the East African region (Table S10). The PFOA and
401 PFOS concentrations in our open lake samples were generally lower than previously measured
402 from lake waters of Winam Gulf (Orata et al., 2009). Since this is a bay of Lake Victoria impacted
403 by urban emissions, the difference in concentrations could be due to dilution in the open lake. The
404 PFOA and PFOS concentrations in Winam Gulf waters were generally lower than observed in our
405 Murchison Bay samples, suggesting elevated emissions to the latter bay or in more recent years.
406 Our Nakivubo Channel samples and the Winam Gulf tributary river samples (Orata et al., 2009)
407 had similar PFOS and PFOA concentrations. Both are heavily impacted by urban emissions. It is
408 noteworthy that concentrations in the Winam Gulf could have likely significantly changed by now,
409 but the above comparison is in light of absence of recent data. Our Murchison Bay samples had
410 relatively higher concentrations of short chain PFASs than recently reported for the same bay
411 (Dalahmeh et al., 2018). The concentrations of short chain PFASs (PFBA, PFPeA, PFHxA and
412 PFHpA) in our riverine and open lake samples are comparable to levels recently reported in water
413 in Germany while long chain PFASs, along with PFBS and PFHxS, in our samples are relatively
414 lower (Joerss et al., 2019; Shafique et al., 2017). This is likely due to differences in local emissions
415 of the latter group of PFASs. Dilution in Lake Victoria leads to open lake PFAS concentrations as
416 low as the ones reported in some less urban-impacted Canadian Arctic lakes (such as Char Lake,

417 Small Lake, North Lake, 9 Mile Lake) (Lescord et al., 2015). In general, our samples show much
418 lower concentrations of PFASs and long chain PFCAs than recent measurements from areas with
419 more urban development (Bai and Son, 2021; Groffen et al., 2018; Sharma et al., 2016; Wang et
420 al., 2020; Zhao et al., 2020). This is likely due to lower use of PFAS containing consumer products
421 in the Lake Victoria Basin than in these more urbanized areas. Average concentrations of PFCAs
422 in our Uganda tap water samples were higher than recently measured in tap water from Turkey
423 (Ünlü Endirlik et al., 2019). With the exception of PFOA and PFBS, the concentrations in our tap
424 water samples were also higher than recently reported levels for tap water samples from Germany
425 (Shafique et al., 2017). It is possible that this is due to less efficient PFAS cleanup of municipal
426 grid water in Uganda. In general, tap water concentrations in our study were either similar or lower
427 than measurements reported for more urbanized countries such as USA (Bradley et al., 2020),
428 Brazil, France and Spain (Schwanz et al., 2016) and China (Li et al., 2019), possibly due to
429 elevated emissions of PFASs to the water stream in these countries. PFAS concentrations in our
430 bulk deposition samples were comparable to general concentration ranges recently reported from
431 measurements in Europe (Johansson et al., 2018) and USA (Gewurtz et al., 2019) but lower than
432 levels measured in China (Johansson et al., 2018).

433 **4. Conclusions**

434 This study is the first study to simultaneously assess open lake concentrations, catchment release
435 and human exposure to PFASs through drinking water in Lake Victoria's basin. The study has
436 shown that for the nearshore urban population, local emissions to nearshore areas from which
437 municipal water is pumped could lead to chronic exposure to PFASs. Although the estimated
438 human exposure to PFASs in this study indicated low risk of adverse health effects, deliberate
439 efforts to specifically target PFAS removal during drinking water treatment are recommended so

440 as to prevent elevated chronic exposure. The results indicate likely contribution of environmental
441 airborne PFAS precursor transformation to the exposure of the lake to PFASs. Atmospheric
442 deposition is the major source of PFASs into Lake Victoria from local, regional and long-range
443 emission source contribution. This reinforces the need for transboundary synergy in reduction of
444 PFAS emissions into the environment. The study also indicates a possibility of high PFAS
445 retention in the lake which could lead to increased human exposure to PFASs via fish consumption
446 following bioaccumulation. The proportions of short chain PFASs in our samples, such as PFHxA
447 and PFHpA, were elevated in the Nakivubo Channel samples. This is consistent with increase of
448 short chain PFASs in consumer products following phase out and/or reduced production of long
449 chain PFASs. It is noteworthy that the data reported here is for the dissolved phase since the water
450 samples were filtered.

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460 **Supporting Information**

461 More details of sampling sites, target analytes, sample preparation, instrumental analysis, quality
462 control parameters, statistics, graphs and a comparison of our results with data published elsewhere
463 are given in the Supporting Information.

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