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Per- and polyfluoroalkyl substances (PFASs) in water, soil and plants in wetlands and agricultural areas in Kampala, Uganda

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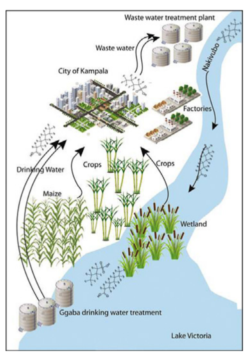
HIGHLIGHTS

- PFOA and PFBS were detected in the drinking water reservoir Lake Victoria.
- PFNA, PFDA, PFUnDA, PFOS and EtFOSAA had high log K_d and log K_{oc} values.
- PFHpA, PFOA, PFNA and PFBS were taken up by yam root, maize cob and sugarcane stem.

GRAPHICAL ABSTRACT



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ABSTRACT

Occurrence and concentrations of 26 per- and polyfluoroalkyl substances (PFASs) were evaluated in wastewater, surface water, soil and crop plants (yam (*Dioscorea* spp.), maize (*Zea mays*) and sugarcane (*Saccharum officinarum*)) in Nakivubo wetland and Lake Victoria at Kampala, Uganda. ∑PFAS concentrations in effluent from Bugolobi wastewater treatment plant (WWTP) were higher (5.6–9.1 ng L^{-1}) than in the corresponding influent (3.4–5.1 ng L^{-1}), indicating poor removal of PFASs within the WWTP. ∑PFAS concentrations decreased by a factor of approximately five between Nakivubo channel (8.5–12 ng L^{-1}) and Lake Victoria (1.0–2.5 ng L^{-1}), due to dilution, sorption to sediment and uptake by plants in the wetland. ∑PFAS concentrations were within the range 1700–7900 pg g^{-1} dry weight (dw) in soil and 160 pg g^{-1} dw (maize cobs) to 380 pg g^{-1} dw (sugarcane stems) in plants. The dominant PFASs were perfluorohexanesulfonate (PFHxS) in wastewater, perfluorooctanoate (PFOA) in surface water, perfluorooctanesulfonate (PFOS) in soil and perfluoroheptanoate (PFHpA) and PFOA in different plant tissues, reflecting PFAS-specific partitioning behaviour in different matrices. Soil-water partitioning coefficient (log K_d) in wetland soil under yam was lowest for short-chain PFHxA (1.9–2.3 L kg^{-1}) and increased with increasing chain length to 2.8–3.1 L kg^{-1} for perfluoroundecanoate (PFUnDA) and 2.8–3.1 L kg^{-1} for perfluoroctanesulfonate (PFOS). The log K_{oc} values ranged between 2.2 and 3.6 L kg^{-1}, with the highest log K_{oc} estimated for long-chain perfluorocarbon PFASs (i.e. PFUnDA 3.2–3.5 L kg^{-1} and PFOS 3.2–3.6 L kg^{-1}). The concentration ratio (CR) between plants and soil was <1 for all PFASs and plant species, with the highest CR estimated for PFHpA (0.65–0.67) in sugarcane stem and PFBS (0.53–0.59) in yam root.

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Overall, this investigation demonstrated PFASs entry into the terrestrial food chain and drinking water resources in Kampala, Uganda. Source identification, assessment of impacts on human health and the environment, and better wastewater treatment technologies are needed.

1. Introduction

Per- and polyfluoroalkyl substances (PFASs) are highly fluorinated organic compounds and have been widely used since the 1950s in a variety of different applications, such as dirt repellents in textiles, carpets, leather and aqueous film-forming foams (AFFFs) (Buck et al., 2011). Some PFASs are thermally, chemically and biologically persistent, and are ubiquitously found in the environment (Ahrens et al., 2016). Primary sources of PFASs in the environment are discharge from wastewater treatment plants (WWTPs), leachate from landfill, industry and use of specific PFAS-containing products (Busch et al., 2010; Ahrens, 2011). Wastewater in particular is a major point source (Ahrens et al., 2009). Concerns have been raised about the transfer of PFASs to humans via contaminated drinking water (Chang et al., 2016; Glynn et al., 2012) or ingestion of contaminated edible crops (Bizkarguenaga et al., 2016) or fish (Ahrens et al., 2016). Several studies have shown uptake of PFASs (e.g. perfluorooctanesulfonate (PFOS), perfluorooctanoate (PFOA), perfluorononanoate (PFNA) and perfluorohexanoate (PFHxA)) in different plant species (e.g. wheat (*Triticum* spp.), strawberry (*Fragaria* spp.), carrot (*Daucus carota sativus*) and lettuce (*Lactuca sativa*), following soil irrigation with treated wastewater or fertilisation with contaminated biosolids (Bizkarguenaga et al., 2016; Blaine et al., 2014; Zhao and Zhu, 2017).

There has been intensive research on the occurrence and fate of PFASs in the environment in industrialised countries such as the US, Europe, Canada, Japan and Australia (Batt et al., 2007; Martin et al., 2004; McLachlan et al., 2007; Papageorgiou et al., 2016; Plahuta et al., 2017; Rosal et al., 2010; Schenker et al., 2008; Thompson et al., 2011; Tomy et al., 2009; Trudel et al., 2008; Vierke et al., 2012; Wilhelm et al., 2010; Wilhelm et al., 2008; Yamashita et al., 2004). However, only a few studies have examined the occurrence and fate of PFASs in environmental samples in Africa and these mainly focus on PFOS and PFOA in wastewater influent and effluent or in Lake Victoria basin in Kenya (Chirikona et al., 2015; Orata et al., 2009) and in Kakume River water in Ghana (Essumang et al., 2017; Orata et al., 2009). Thus, information regarding PFASs pollution of water resources, soil and plants in developing countries is still scarce, partly due to the high cost of analysis and lack of laboratory infrastructure and analytical methods.

Kampala, the capital city of Uganda, borders two important ecosystems with great socio-economic value: Nakivubo wetland and Lake Victoria. Today, the Nakivubo wetland system covers one-sixth of Kampala (31 km^2), but it is under considerable pressure due to demographic and ecological transformations, including rapid urbanisation, industrial development and establishment of informal settlements nearby (Kayima et al., 2008; Mbabazi et al., 2010). In fact, Nakivubo wetland now receives domestic and industrial wastewater from half a million people and surface water from the city of Kampala (Fuhrimann et al., 2015). The water in Nakivubo channel is used for cultivation of maize (*Zea mays*) and sugarcane (*Saccharum officinarum*) in the surrounding fields, while the wetland itself is used for cultivation of yam (*Dioscorea* spp.) (Emerton, 2005; Kayima et al., 2008; Mbabazi et al., 2010). The remaining Nakivubo water flows down into Lake Victoria, which is another important ecosystem of great socio-economic significance for riparian status in the area, as it is the major source of drinking water and water for agricultural and industrial purposes (Juma et al., 2014; Orata et al., 2009). The lake also provides a large quantity of fish for East African countries and for export markets in the USA, Australia, European Union countries and Israel (Juma et al., 2014).

The aim of this study was to investigate multimedia contamination with PFASs in Nakivubo wetland and the surrounding agricultural fields and in the waters of Lake Victoria at Kampala, Uganda. Specific objectives were (i) to evaluate the occurrence and concentrations of 26 PFASs in water and wastewater resources in Nakivubo wetland and in lake water, and (ii) to assess uptake of PFASs from soil and water by three edible plant species (maize, sugarcane, yam) in Nakivubo wetland downstream of Bugolobi WWTP and upstream of Lake Victoria.

2. Materials and methods

2.1. Target analytes

In total, 26 PFASs were included as target analytes, comprising 13 perfluorooalkyl carboxylates (C_4-C_{14}, C_{16}, C_{18} PFCAs) (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFHxDA, PFOcDA,), four perfluoroalkylsulfonates (C_4, C_6, C_8, C_{10} PFSAs) (PFBS, PFHxS, PFOS, PFDS), three perfluorooctanesulfonamides (FOSAs) (FOSA, MeFOSA, EtFOSA), two perfluorooctanesulfonamidoethanols (FOSEs) (MeFOSE, EtFOSE), three perfluorooctanesulfonamido acetic acids (FOSAAs) (FOSAA, MeFOSAA, EtFOSAA) and 6:2 fluorotelomersulfonate (FTSA) (see Table S1 in Supporting information for list of all abbreviations and full names). All standards were purchased from Wellington Laboratories (ON, Canada). In addition, 16 mass-labelled internal standards (IS) were included, consisting of the following compounds: $^{13}C_4$-PFBA, $^{13}C_2$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_5$-PFNA, $^{13}C_2$-PFDA, $^{13}C_2$-PFUnDA, $^{13}C_2$-PFDoDA, $^{18}O_2$-PFHxS, $^{13}C_4$-PFOS, $^{13}C_8$-FOSA, d_3-MeFOSAA, d_5-EtFOSAA, d_3-MeFOSA, d_5-EtFOSA, d_7-MeFOSE and d_9-EtFOSE, which were purchased from Wellington Laboratories (ON, Canada) (for details, see Ahrens et al. (2016)).

2.2. Description of the study area

The study area was divided into four sampling systems along the main wastewater chain of Kampala: (i) Nakivubo channel, (ii) Nakivubo wetland, (iii) agricultural fields bordering Nakivubo channel and (iv) Inner Murchison Bay in Lake Victoria (Fig. 1). Nakivubo channel is a 12.3 km long drainage channel and is influenced by stormwater and illegally discharged wastewater from communities, markets, industries and by secondary-treated effluent from Bugolobi WWTP before it discharges into Nakivubo wetland. Nakivubo wetland is located to the south-east of Kampala (00°18′N, 32°38′E), at an altitude of 1135 m above sea level, covers ~5.29 km^2 and has a total catchment area of over 40 km^2 (Emerton, 1998). The wetland is divided by an old railway line and consists mainly of drained farmland in the north and submerged (floating) wetland in the south. Both areas are cropped with yams and sugarcane. The wetland acts as a buffer zone before water reaches Lake Victoria (Kansiime and Nalubega, 1999).

2.3. Sample collection

Wastewater, surface water, soil and plant samples were collected from the study area during two sampling events, on 29 April and 5 May 2015 (Fig. 1). Grab samples of wastewater were collected in duplicate from the influent and effluent of Bugolobi WWTP during both sampling events (in total $n = 8$). Surface water grab samples (50 cm below the water surface) were collected manually (using a rope and bucket) in duplicate in Nakivubo channel and Nakivubo wetland near the yam