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Polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans and polybrominated diphenyl ethers in sediments and fish species from the Murchison Bay of Lake Victoria, Uganda

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HIGHLIGHTS

- High concentrations of PCDD/Fs and PBDEs were found at wastewater discharge points.
- PCDD/F and PBDE concentrations were low to moderate compared to data in literature.
- The concentration difference between fish species was attributed to feeding habits.
- The fish were fit for human consumption with regard to PCDD/Fs and PBDEs.

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ABSTRACT

Polychlorinated dibenzo-*p*-dioxins/furans (PCDD/Fs) and polybrominated diphenyl ethers (PBDEs) were analyzed in sediments and fish from the Murchison Bay of Lake Victoria by high resolution gas chromatography/high resolution mass spectrometry (HRGC/HRMS). Average concentrations of total (Σ) PCDD/Fs and ΣPBDEs in sediments ranged from 68.8 to 479 pg g^{-1} dry weight (dw) and 60.8 to 179 pg g^{-1} dw, respectively. Contamination levels of sedimentary PCDD/Fs and PBDEs were low to moderate compared to other urbanized regions worldwide. The concentrations in different fish species (Nile perch; *Lates niloticus* and Nile tilapia; *Oreochromis niloticus*) were 5.32 to 49.0 pg g^{-1} wet weight (ww) for PCDD/Fs and 59.3 to 495 pg g^{-1} ww for PBDEs. Higher concentrations of the pollutants were found in *L. niloticus* than *O. niloticus*, which could be attributed to species differences in feeding habits and lifestyles. World Health Organization-toxic equivalents (WHO_{2005}-TEQs) for PCDD/Fs ranged from 0.08 to 0.33 pg TEQ g^{-1} dw and 0.001–0.14 pg TEQ g^{-1} ww in sediments and fish, respectively. The TEQ values were low compared to the data for fresh water fish reported in literature and within a permissible level of 3.5 pg g^{-1} ww recommended by the European Commission. Based on the Commission set value and minimum risk level criteria formulated by the Agency for Toxic Substances and Disease Registry, the fish from the Murchison Bay was fit for human consumption.

1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polybrominated diphenyl ethers (PBDEs) are a group of halogenated aromatic hydrocarbons with tendencies to persist in the environment, bioaccumulate in fatty tissues and undergo long range transport (Jones and de Voogt, 1999; van der Oost et al., 1996; Wania and MacKay, 1996). The sources of PCDD/Fs include waste incineration, industrial chemical processes and, burning of waste and vegetation (Antunes et al., 2012; Liu et al., 2013; Martínez et al., 2010; Moon et al., 2012; Tuppurainen et al., 1998; Xu et al., 2009). PBDEs originate from unregulated disposal of textiles, building materials and obsolete electronic waste (e-waste) (Ren et al., 2013; Wang et al., 2007). In the recent years, e-waste has emerged as a global environmental problem because of a high frequency of updating electronics to boost the rapid growing technology. The dumping points for e-waste from the developed countries are often in Africa and Asia, in a form of donations and sometimes for recycling due to less stringent environmental regulations

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in the receiver countries (Frazzoli et al., 2010; Lawhon, 2013; Ongondo et al., 2011).

In the environment, PCDD/Fs and PBDEs are transferred to sediment and water, taken up by aquatic organisms, and eventually biomagnified in top predators (Binelli and Provini, 2003; Kelly et al., 2007). The most probable routes of exposure to such pollutants in humans are dermal contact, inhalation and ingestion (Chan and Wong, 2013; Dirtu and Covaci, 2010; Ni et al., 2012). It has been estimated that more than 50% of total human exposure to dioxins is from food of animal origin (Dougherty et al., 2000; Kim et al., 2013; Martí-Cid et al., 2007). The compounds are of public concern because of the health implications attached to them such as lesions, suppression of the immune system, reproductive impairment and endocrine disruption (Mezcua et al., 2012; Reinen et al., 2010).

Despite the reports on PBDEs in the developing and developed world (Bervoets et al., 2005; Covaci et al., 2005; Hallanger et al., 2011), little is known about these pollutants in Uganda, while only two studies have been carried out on PCDD/Fs in sediments and fish from the Napoleon Gulf and Thurston Bay of Lake Victoria (Ssebugere et al., 2013a, 2013b). Another embayment of interest on Lake Victoria is the Murchison Bay, a receiver-end for industrial and municipal discharges from Kampala City, Uganda's capital (Banadda et al., 2009; Birungi et al., 2007; Haande et al., 2011). The aim of this study was to determine the concentration magnitude of PCDD/Fs and PBDEs in sediments and fish from the Murchison Bay and to compare the results with those obtained in studies elsewhere.

2. Methods and materials

2.1. Study area and sampling

The Murchison Bay is located in the northern waters of Lake Victoria and south-east of Kampala capital city. The Bay receives urban and industrial effluents from Kampala, the most highly polluted city in Uganda. The geographical locations of the Bay are shown in Fig. 1. A total of 24 surface sediments (<25 cm depth layer; 6 samples from each of sites 1, 2, 3 and 4) were collected using a tube sampler. Also, 48 Nile perch (*Lates niloticus*) a top-predator and 48 Nile tilapia (*Oreochromis niloticus*) a detritivorous species were collected using gill nets from the same locations as sediments. The two fish species were chosen because they are readily available and highly consumed within the Lake Victoria Basin (Nkalubo et al., 2014). The fish were transported on ice to the laboratory to minimize tissue decay. In the laboratory, the fish were dissected and muscle tissues taken. Four tissues from fish of the same species and location were pooled, and homogenized to obtain a composite sample. The sediments and homogenized fish were transferred into acetone rinsed glass bottles and transferred to the analysis laboratory in China where they were freeze-dried.

2.2. Analytical procedure

Sediment samples were divided into subsamples for POP analysis and total organic carbon (TOC) determination. The sediment TOC (%) was measured with a LECO C230 carbon analyzer after the removal of carbonates with hydrochloric acid. The extraction and clean-up procedure are described in our previous work (Wang et al., 2013). Briefly, 2 g of freeze-dried sediments and/or fish was ground into a fine powder using a pestle and mortar, and mixed with 10 g of anhydrous sodium sulfate. The mixture was spiked with ^{13}C-labeled surrogate standards (PBDE-LCS and US EPA defined EPA1613-LCS) and extracted on an accelerated solvent extractor device (ASE300, Dionex, USA) using a mixture of dichloromethane (DCM)/*n*-hexane (1:1 v/v) at 1500 psi and 150 °C for two static cycles (each cycle was 8 min). The resulting extract was concentrated on a rotary evaporator to 2 mL for sediments and to dryness for fish. The lipid content of the fish was then obtained by the difference in weight before and after evaporation to dryness. Prior to clean-up, the fish extract was reconstituted in *n*-hexane while activated copper granules were added to the concentrated sediment extract to remove elemental sulfur. The clean-up involved three sequential stages: a multi-layer silica gel column (packed from bottom to top with 1 g silica gel, 4 g basic silica gel, 1 g silica gel, 8 g acidified silica gel, 2 g silica gel and 2 g anhydrous sodium sulfate) followed by an alumina column (6 g basic alumina and 3 g anhydrous sodium sulfate at the top), and then a carbon column (1.5 g of 18% carbon dispersed in cilite, 3 g anhydrous sodium sulfate at the top). The first fraction containing PBDEs (on the alumina column) and the second fraction containing PCDD/Fs

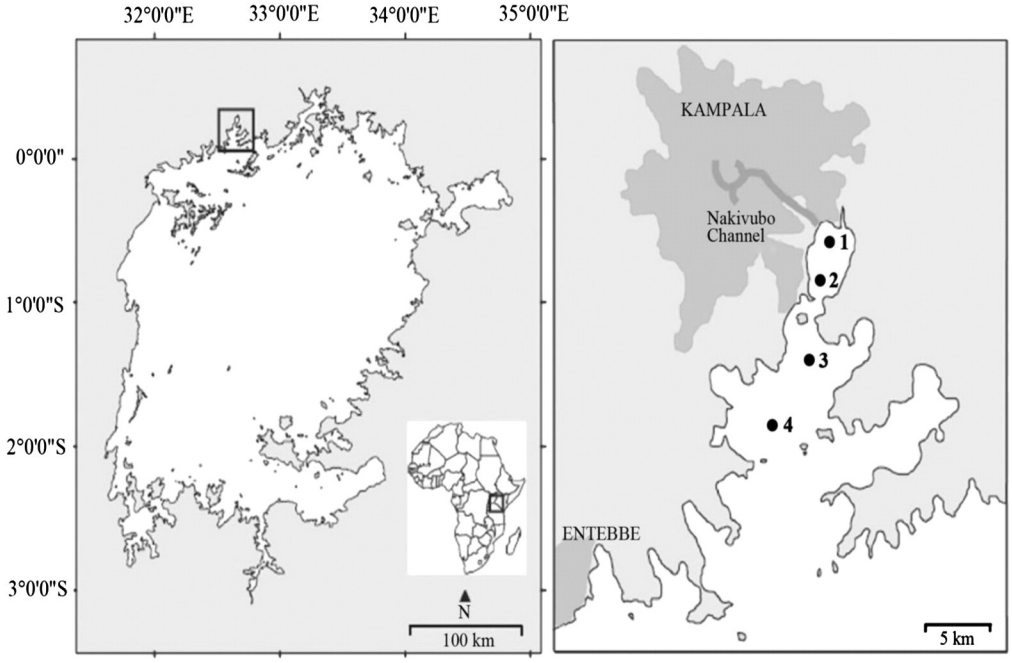

Fig. 1. Geographical location of the Murchison Bay.
Adapted from Haande et al. (2011).

(on the carbon column) were recovered using 50 mL of *n*-hexane and 80 mL of toluene, respectively. The fractions were separately concentrated to <2 mL and transferred to sample vials containing 20 μL nonane. The mixture was reduced to 20 μL using a gentle stream of nitrogen and spiked with ^{13}C-labeled internal standards (US EPA defined EPA1613-IS and 68A-IS) prior to instrumental analysis.

The analysis was performed on an AutoSpec Ultima (Waters Micromass, UK) high resolution mass spectrometer (HRMS) coupled to an Agilent 6890N series gas chromatograph (GC) (Wilmington, USA). The HRMS was operated in VSIR mode at a trap current of 500 μA, electron energy of 35 eV and an acceleration voltage of 8000 V. Chromatographic separation for PCDD/Fs was achieved on a fused-silica capillary column (DB5-MS 60 m × 0.25 mm i.d. × 0.25 μm film thickness). The GC oven temperature was initiated at 150 °C for 3 min, increased to 230 °C for 18 min at 20 °C min^{-1}, raised to 235 °C for 10 min at 5 °C min^{-1} and finally held at 330 °C for 3 min at 4 °C min^{-1}. Chromatographic separation for PBDEs was performed using a DB5-MS column (30 m × 0.25 mm i.d × 0.10 μm). The conditions for PBDE determination were as follows: the oven temperature was initially programmed at 100 °C for 2 min, increased to 230 °C for 1 min at a rate of 15 °C min^{-1}, elevated to 270 °C for 6 min at 5 °C min^{-1} and finally held at 330 °C for 8 min at 10 °C min^{-1}. Injection was carried out in splitless mode, using helium as carrier gas at a constant flow rate of 1 mL min^{-1}. The temperature of the ion source was set at 280 °C for PBDEs and 270 °C for PCDD/Fs. Quantification was performed using the ^{13}C isotopic dilution method.

2.3. Quality assurance/quality control

To ensure the quality of data, blanks, replicates and surrogate standards were included in the analysis. One laboratory blank was performed for each batch of 5 samples. For any analyte detected in the blank, the mean procedural blank value was used for subtraction. Concentrations of target analytes in the blanks were <5% of the minimum levels in the samples, suggesting that they were free from contamination. The recoveries for surrogate PCDD/Fs were 46–104% in the sediments and 50–100% in the fish. Those for PBDEs varied from 54 to 99% and 56 to 109%, respectively. The recoveries were within the range acceptable for EPA methods 1613B and 1614. Limit of detection (LOD) was calculated as three times the signal-to-noise ratio (S/N). The LODs

Table 1
Mean concentrations of PCDD/Fs and PBDEs (pg g^{-1} dw), TOC (%) and texture of the sediments from the Murchison Bay.

	Site 1	Site 2	Site 3	Site 4
TOC (%)	5.23 (3.10–7.70)	4.03 (2.30–6.00)	3.35 (1.60–5.54)	2.33 (1.50–3.70)
Sediment texture	Clay	Sandy	Clay and sandy	Clay and plastic
PCDFs				
2,3,7,8-TCDF	n.d.	0.0700 (n.d.–0.0700)	1.42 (1.41–1.42)	0.760 (n.d.–0.760)
1,2,3,7,8-PeCDF	n.d.	0.100 (n.d.–0.100)	n.d.	n.d.
1,2,3,4,7,8-HxCDF	0.250 (n.d.–0.250)	0.120 (n.d.–0.120)	0.870 (n.d.–0.870)	0.440 (n.d.–0.440)
1,2,3,6,7,8-HxCDF	0.450 (n.d.–0.450)	0.500 (n.d.–0.500)	0.890 (0.750–1.03)	0.320 (n.d.–0.320)
2,3,4,6,7,8-HxCDF	n.d.	0.840 (0.380–1.29)	0.670 (0.380–0.960)	n.d.
1,2,3,7,8,9-HxCDF	n.d.	0.420 (n.d–0.420)	n.d.	n.d.
1,2,3,4,6,7,8-HpCDF	19.6 (6.58–28.1)	18.8 (15.8–21.1)	2.61 (2.54–2.69)	1.88 (1.70–2.13)
1,2,3,4,7,8,9-HpCDF	3.11 (n.d.–3.11)	1.80 (n.d.–1.80)	n.d.	0.110 (n.d.–0.110)
OCDF	341 (225–402)	219 (161–255)	5.79 (2.09–9.50)	18.8 (2.33–48.3)
ΣPCDFs	364 (249–430)	242 (181–271)	12.3 (6.10–9.50)	22.8 (5.65–50.4)
PCDDs				
2,3,7,8-TCDD	n.d.	0.300 (n.d.–0.300)	n.d.	n.d.
1,2,3,4,7,8-HxCDD	n.d.	0.100 (n.d.–0.100)	0.0600 (n.d.–0.0600)	n.d.
1,2,3,6,7,8-HxCDD	n.d.	0.130 (n.d.–0.130)	0.970 (0.470–1.46)	n.d.
1,2,3,7,8,9-HxCDD	n.d.	0.660 (n.d.–0.660)	0.330 (n.d.–0.330)	n.d.
1,2,3,4,6,7,8-HpCDD	6.76 (n.d.–6.76)	3.84 (3.47–4.07)	12.8 (5.58–18.3)	7.01 (6.67–7.35)
OCDD	108 (25.4–193)	68.3 (62.4–76.6)	85.6 (64.8–102)	39.0 (11.5–53.2)
ΣPCDDs	114 (25.4–193)	73.4 (65.9–80.6)	99.8 (71.9–121)	46.0 (11.5–59.8)
ΣPCDD/Fs	479 (442–518)	315 (247–351)	112 (81.4–129)	68.8 (61.9–67.7)
Ratios (ΣPCDFs/ΣPCDDs)	3.18 (1.29–16.96)	3.30 (2.75–3.74)	0.120 (0.0600–0.130)	0.50 (0.09–4.36)
WHO_{2005}-TEQs (PCDD/Fs)	0.14 (0.01–0.24)	0.17 (0.12–0.25)	0.33 (0.14–0.47)	0.08 (0.03–0.14)
ΣPCDD/Fs × % TOC	2505	1269	375	160
PBDEs				
Tri-BDE				
BDE 17	7.17 (1.26–17.5)	10.1 (6.68–15.7)	7.65 (0.440–19.7)	0.90 (n.d–1.26)
BDE 28	4.53 (1.60–6.15)	4.15 (2.64–5.61)	3.80 (1.71–6.09)	1.44 (1.36–1.60)
Tetra-BDE				
BDE 47	60.6 (16.6–97.2)	42.7 (22.6–63.1)	35.3 (8.98–58.6)	22.4 (16.5–34.1)
BDE 66	4.62 (1.13–6.97)	3.54 (2.97–4.45)	2.41 (0.73–3.38)	1.13 (0.73–1.52)
Penta-BDE				
BDE 85	3.74 (n.d–3.74)	2.02 (0.99–3.50)	0.71 (n.d–1.25)	1.69 (n.d–1.69)
BDE 99	48.2 (10.5–87.6)	30.1 (19.4–39.5)	20.9 (8.55–39.8)	21.1 (10.5–40.8)
BDE 100	15.5 (4.48–25.0)	9.54 (5.43–12.4)	7.83 (2.11–11.8)	5.49 (3.70–8.29)
Hexa-BDE				
BDE 138	1.83 (n.d–2.14)	n.d	0.480 (n.d–0.480)	n.d
BDE 153	9.07 (2.43–13.5)	8.54 (2.48–12.9)	5.10 (2.13–8.31)	2.38 (0.71–4.01)
BDE 154	8.68 (1.15–13.3)	4.97 (2.67–9.49)	4.08 (0.0400–6.70)	1.78 (0.61–3.58)
Hepta-BDE				
BDE-183	15.4 (2.99–34.8)	7.88 (4.31–10.2)	7.08 (1.91–11.3)	2.48 (1.58–2.88)
ΣPBDEs	179 (42.2–265)	124 (72.9–154)	95.4 (26.8–144)	60.8 (37.9–98.2)
ΣPBDEs × % TOC	936	450	320	142

The results are presented as mean concentration (ranges). The number of samples per station (N) = 6; n.d. — non-detectable or <LOD; TOC — total organic carbon.

Table 2
Comparison of sediment PCDD/F and PBDE concentrations (pg g^{-1} dw) in this study with the data from literature.

Area	PCDD/F concentrations	References
Murchison Bay of Lake Victoria, Uganda	69–479	This study
Napoleon Gulf and Thurston Bay of Lake Victoria, Uganda	3.36–50	Ssebugere et al. (2013a)
Nile River, Egypt	240–775	El-Kady et al. (2007)
Vaal River, South Africa	1.4–183	Nieuwoudt et al. (2009)
Lake Awassa, Ethiopia	n.d. to 270	Urbaniak and Zalewski (2011)
Kentucky Lake, USA	580–1300	Loganathan et al. (2008)
Kentucky Lake, USA	n.d.to 4830	Kannan et al. (2008)
Guaymas Basin and Patzcuaro Lake, Mexico	12–44	Yunuén et al. (2011)
Lake Superior	5–18,000	Shen et al. (2009)
Lake Maggiore in Italy and Switzerland	n.d. to 400	Vives et al. (2007)
Mondego estuary, Portugal	n.d. to 110	Nunes et al. (2011)
Can Gio, Southern Vietnam and Osaka, Japan	264–11,000	Kishida et al. (2010)
Xiangjiang River, China	876–497,759	Chen et al. (2012)
Lake Shihwa, Korea	1.0–1770	Moon et al. (2012)
Area	**PBDE concentrations (pg g^{-1} dw)**	**References**
Lake Victoria, Uganda	60.5–179	This study
Congo River Basin, DR Congo	n.d. to 1900	Verhaert et al. (2013)
Juksei River, South Africa	902–6800	Olukunle et al. (2012)
Niagara River, Canada	1700–124,600	Samara et al. (2006)
Ebro river basin, Spain	205–17,240	Lacorte et al. (2006)
Columbia River, USA	n.d.–15,100	Counihan et al. (2014)

n.d. — non-detectable.

for PCDD/Fs were 0.001–0.05 pg g^{-1} dry weight (dw) in the sediments and 0.002–0.07 pg g^{-1} wet weight (ww) in the fish. Those for PBDEs ranged from 0.08 to 1.27 pg g^{-1} dw and 0.05 to 0.92 pg g^{-1} ww, respectively. For samples where the concentrations of a congener were less than the LOD, the levels were reported as not detected (n.d.). Relative standard deviations of the method (n = 3) were in the range of 1–9%.

2.4. Statistical data analysis

Statistical analysis was conducted using Statistica 6.0 for windows. Non-parametric tests were performed in cases where the results were not normally distributed. The Mann–Whitney *U* test was used for comparing two data sets and the Kruskal–Wallis test when more than two groups were considered. In these two non-parametric tests, the level of significance was set at $p < 0.05$. Toxic equivalents (TEQs) were

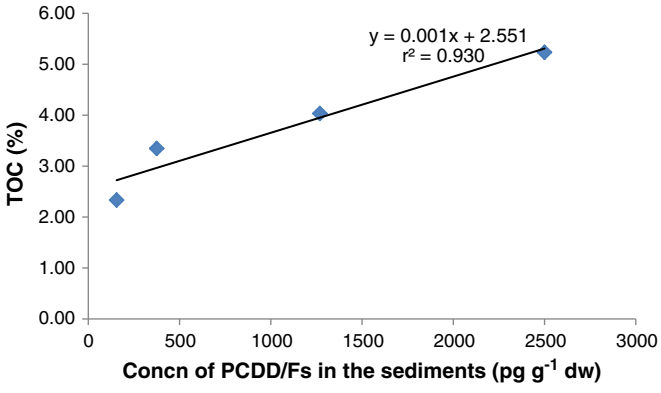

Fig. 2. Correlation between the sediment PCDD/F concentrations and percent TOC.

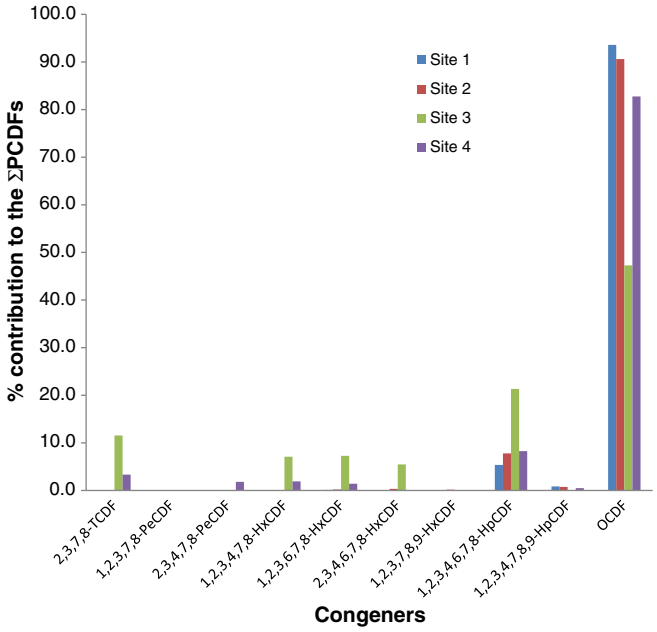

Fig. 3. Percent contribution of the congeners to the ΣPCDFs.

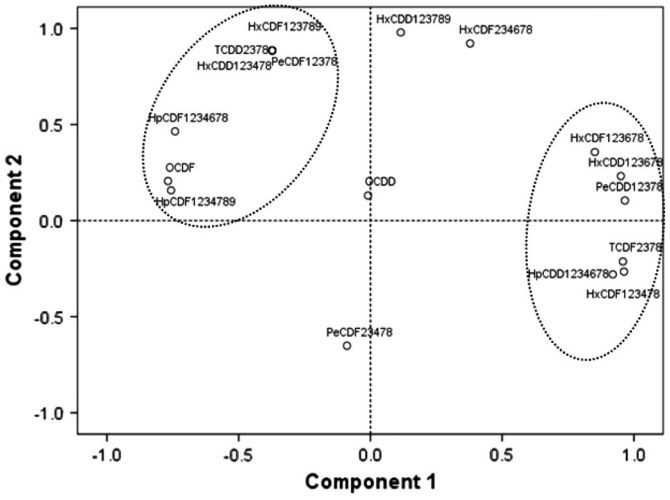

Fig. 4. PCA plot corresponding the PCDD/F concentrations to the known sources in literature.

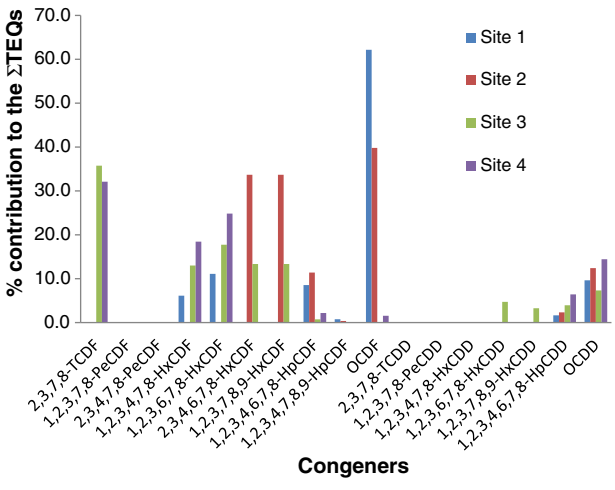

Fig. 5. Percent contribution of PCDD/F congeners to the ΣTEQs.

calculated using the proposed World Health Organization-toxic equivalent factors (WHO_{2005}-TEFs) for the corresponding chlorinated counterparts (Van den Berg et al., 2006). Biota-sediment accumulation factors (BSAFs) were calculated based on the formula $BSAF = C'_{avg} / Csoc_{avg}$ (Burkhard et al., 2005). Where C'_{avg} is a lipid normalized concentration of a chemical in the fish and $Csoc_{avg}$ is an organic carbon normalized concentration of the chemical in the sediment. Principal component analysis (PCA) was used to ascertain the major sources of PCDD/Fs. Principal components were considered when their Eigen values were >1. After identifying the correlations, a PCA analysis was conducted, using the Scree test to find the number of factors that can be used in the interpretation related to the estimated correlations and to justify the variance of the data (Jiang et al., 2011).

3. Results and discussion

3.1. Concentrations of PCDD/Fs in the sediments

Seventeen 2,3,7,8-substituted PCDD/F congeners were determined in the sediments from the Murchison Bay and their levels are shown in Table 1. The sum (Σ) PCDD/Fs at the different study sites varied widely with mean values ranging from 68.8 to 479 pg g^{-1} dw. The levels were in the same range with data from the Napoleon Gulf and Thurston Bay of Lake Victoria (Ssebugere et al., 2013b) and other fresh water bodies in Africa like the Nile River in Egypt (El-Kady et al., 2007), Vaal River in South Africa (Nieuwoudt et al., 2009) and Lake Awassa in Ethiopia (Urbaniak and Zalewski, 2011) (Table 2). The levels were low to

Table 3
Mean concentrations and ranges of PCDD/Fs and PBDEs (pg g^{-1} ww) in fish species from the Murchison Bay.

	Site 1		Site 2		Site 3		Site 4	
	L. niloticus	*O. niloticus*	*L. niloticus*	*O. niloticus*	*L. niloticus*	*O. niloticus*	*L. niloticus*	*O. niloticus*
Lipid content (%)	2.8	2.2	2.3	1.9	2.2	1.5	1.6	1.2
	2.3–3.4	2.0–2.5	1.9–2.6	1.7–2.1	1.8–2.5	1.2–1.8	1.3–1.8	0.9–1.4
PCDFs								
1,2,3,6,7,8-HxCDF	n.d.	0.14	n.d.	0.27	n.d.	n.d.	n.d.	n.d.
		n.d.–0.14		n.d.–0.27				
1,2,3,4,6,7,8-HpCDF	2.91	0.43	0.45	1.22	0.37	0.30	0.38	0.37
	2.19–3.63	0.29–0.56	0.34–0.55	n.d.–1.22	n.d.–0.37	0.23–0.37	0.37–0.39	n.d.–0.37
1,2,3,4,7,8,9-HpCDF	n.d.	n.d.	n.d.	0.53	n.d.	n.d.	n.d.	n.d.
				n.d.–0.53				
OCDF	23.2	0.77	n.d.	7.75	2.13	2.37	2.58	1.35
	11.6–34.7	n.d–0.77		3.99–11.5	n.d.–2.13	n.d.–2.37	2.13–3.02	n.d.–1.35
ΣPCDFs	26.1	1.34	0.45	9.77	2.50	2.67	2.96	1.72
	n.d.–38.4	n.d.–1.34	n.d.–0.55	n.d.–12.0	n.d.–2.50	n.d.–2.67	n.d.–3.41	n.d.–1.72
PCDDs								
1,2,3,4,6,7,8-HpCDD	2.91	1.87	3.06	1.08	0.94	0.51	0.57	0.44
	0.430–7.12	1.15–2.70	2.52–3.53	0.97–1.18	0.39–1.47	0.40–0.62	0.39–0.75	n.d–0.44
OCDD	20.0	15.1	19.0	6.16	8.58	3.08	3.72	3.16
	7.80–39.8	14.3–16.4	17.2–22.4	2.02–8.29	4.18–13.4	1.52–4.37	3.51–4.18	2.48–4.06
ΣPCDDs	22.9	17.0	22.1	7.24	9.52	3.59	4.29	3.60
	8.23–46.9	15.9–19.1	19.9–26.0	2.02–9.47	4.57–14.8	1.91–4.99	3.51–4.57	2.60–4.06
ΣPCDD/Fs	49.0	18.4	22.6	17.0	12.0	6.26	7.25	5.32
	27.5–46.9	16.5–19.1	20.3–26.0	9.15–15.0	7.07–14.8	3.37–5.22	3.51–7.07	2.60–4.63
ΣWHO_{2005}-TEQs	0.015	0.011	0.009	0.140	0.004	0.002	0.002	0.001
	0.011–0.019	0.006–0.020	0.008–0.010	0.003–0.033	0.003–0.005	0.001–0.002	0.001–0.003	0.001–0.002
(ΣPCDD/Fs)/(% lipid)	17.5	8.36	9.83	8.94	5.45	4.17	4.53	4.43
BSAF values	0.006	0.003	0.007	0.007	0.015	0.011	0.028	0.028
PBDEs								
BDE 17	1.54	3.53	13.0	7.09	2.14	2.07	1.12	1.17
	0.91–2.76	1.13–9.09	0.49–37.6	1.25–15.7	0.60–4.59	0.22–4.74	n.d.–1.26	0.92–1.66
BDE 28	11.3	2.79	6.73	3.73	2.25	1.86	1.82	1.11
	2.92–18.8	1.15–5.84	1.39–11.8	n.d.–5.61	1.33–3.99	0.88–2.56	n.d.–2.04	0.93–1.31
BDE 47	248	45.9	93.1	32.2	32.8	19.6	30.9	21.4
	149–342	20.7–97.2	17.9–138	12.8–42.8	11.9–66.5	9.04–36.9	16.6–45.0	10.3–29.5
BDE 66	4.95	3.33	4.74	2.82	1.54	1.05	1.49	1.43
	3.25–7.28	n.d.–6.97	1.26–11.4	n.d.–4.45	0.54–3.36	0.860–1.19	n.d.–1.86	0.79–2.02
BDE 85	4.27	1.88	1.33	1.57	4.11	n.d.	2.10	1.08
	n.d.–6.12	n.d.–3.74	0.64–2.02	n.d.–1.57	n.d.–4.11		n.d.–2.65	n.d.–1.29
BDE 99	44.0	36.0	33.1	20.8	39.2	19.0	30.0	21.7
	n.d.–76.1	14.7–87.6	9.78–77.6	11.9–31.5	4.27–17.1	7.37–37.6	10.5–50.4	11.9–29.4
BDE 100	16.9	10.8	22.4	6.33	9.89	6.56	8.18	5.29
	n.d.–16.9	4.96–25.0	2.66–34.9	3.29–10.8	1.54–23.1	1.30–15.1	n.d.–11.9	2.99–7.03
BDE 138	0.76	2.14	n.d.	n.d.	n.d	n.d	5.20	0.31
	n.d.–0.76	n.d.–2.14					n.d.–5.20	n.d.–0.31
BDE 153	17.4	5.78	12.7	5.50	6.06	4.23	3.69	3.16
	6.76–37.9	2.13–13.5	2.02–21.2	0.85–12.9	2.40–12.5	0.850–10.6	2.43–4.76	0.930–4.82
BDE 154	117	4.96	20.4	5.81	3.16	1.49	2.47	2.34
	8.49–231	2.19–11.5	0.98–52.6	n.d.–9.49	1.86–5.67	0.600–1.76	1.15–3.32	1.16–3.26
BDE 183	28.9	5.96	20.2	5.70	4.77	3.45	2.99	6.46
	n.d.–55.2	2.31–10.7	n.d.–38.8	n.d.–9.11	1.05–3.88	0.980–7.07	n.d.–2.99	2.22–13.4
ΣPBDEs	495	123	228	91.6	106	59.3	90.0	64.8
	275–695	59.9–264	40.8–361	37.6–144	29.3–229	22.7–118	42.2–121	32.1–87.9
(ΣPBDEs)/% lipid	177	55.9	99.1	48.2	48.2	39.5	56.3	54.0
BSAFs	0.071	0.022	0.078	0.038	0.128	0.105	0.352	0.338

Number of fish homogenates per species per station (N) = 3; n.d. — non detectable.

moderate compared to the data reported in sediments from industrialized areas in the United States of America (Kannan et al., 2008; Loganathan et al., 2008; Shen et al., 2009; Yunuén et al., 2011) and Europe (Nunes et al., 2011; Vives et al., 2007). The results were lower than those reported in sediments from Asia: Can Gio in Vietnam and Japan (Kishida et al., 2010); Xiangjiang River in China (Chen et al., 2012) and Lake Shihwa in Korea (Moon et al., 2012).

In the present study, the PCDD/F concentrations in the sediments from site 1 were statistically greater ($p < 0.05$, Mann–Whitney U test) compared to sites 3 and 4. However, the levels between sites 1 and 2 both located in the inner Murchison Bay were insignificant. Site 1 is close to the mouth of Nakivubo channel which traverses through urban and industrial areas of Kampala capital city, bringing in wastewater effluents into the bay. Statistical analysis showed that the PCDD/F concentrations for site 2 were also significantly higher than those at stations 3 and 4. Site 2 is located near Ggaba where local conditions are influenced by industrial activities. It should be noted that the use of wood and charcoal stoves is also typical in areas surrounding site 2, and could be sources of PCDD/Fs and/or related compounds. In the outer Murchison Bay, the concentrations of the ΣPCDD/Fs at sites 3 and 4, both situated far from industrial and urban activities were an order of magnitude lower compared to the ones in the inner Murchison Bay. The PCDD/F levels at site 3 were higher compared to those at site 4, although no significant difference was observed between the two locations. The low levels at site 4 could be due to its far off shore distance from the point sources. The concentrations of PCDD/Fs were strongly correlated to the total organic carbon ($r^2 = 0.930$) (Fig. 2), suggesting that organic matter could have an influence on the levels of the pollutants. The result is in agreement to that by van der Oost et al. (1996), who reported a significant correlation between the sediment organic content and persistent organic pollutant (POP) levels from six freshwater sites in Amsterdam. It should be noted that there was no relationship between the PCDD/F concentrations and sediment texture.

In this study, octachlorodibenzofuran (OCDF) was the most dominant congener compared to other PCDF homologues (based on mean values it constituted 47–94% to the ΣPCDFs at all stations) (Fig. 3). OCDF was followed by 1,2,3,4,6,7,8-HpCDF (accounting for 5–21% of the ΣPCDFs). Martínez et al. (2010) pointed out that significant concentrations of OCDF and HpCDF in environmental samples are mostly indicative of thermal processes, urban waste incineration, ferrous- and non-ferrous metal operations, and sintering plants as potential sources. Of the PCDD congeners, OCDD was the principal contributor (accounting for >84% to the ΣPCDDs in majority samples). High concentrations of OCDD compared to those of other PCDD congeners were also reported in surface sediments from the Vaal River of South Africa (Nieuwoudt et al., 2009). One reason for the domination of OCDD could be that it is emitted at higher rates in the atmosphere than other PCDDs. Generally, OCDD is associated with emissions from uncontrolled low temperature sources, such as inefficient bio-mass combustion and/or open-burning of wood and domestic waste (Anderson and Fisher, 2002; Wevers et al., 2004). Ratios of the ΣPCDFs/ΣPCDDs can be used to identify the main source(s) of dioxins to the environment (Huang and Buekens, 1995). The authors pointed out that if the ratio (PCDFs/PCDDs) is >1, then combustion is a possible major source of PCDD/Fs. The calculated ratios in the present study were >1 in 58% of the sediments suggesting combustion as a possible source of PCDD/Fs and/or related compounds.

Principal component analysis was done to compare the percent congener profiles with known sources of PCDD/Fs reported in literature such as medical waste incinerators, municipal solid waste combustors and thermal industrial processes (Baker and Hites, 2000; Martinez et al., 2010). The result from PCA indicates that CP-1 which was related to pentachlorodibenzofuran (PeCDF), explained 45% of the variance and CP-2 which was related to OCDF, explained 36% of the variance, with these two components explaining 81% of the variance (Fig. 4). In the ordination space generated by the PCA, two groups were formed: the first one (containing OCDF and PeCDF) grouped the sources related to local combustion processes, including agricultural straw open burning, medical waste incinerators and municipal solid waste combustors, and the second one (containing PeCDD, HxCDD and HpCDD) corresponded to ovens.

The WHO_{2005}-TEQs for PCDD/Fs in sediments of this study ranged from 0.08 to 0.33 pg TEQ g^{-1} dw. In our earlier work, we reported

Table 4
Comparison of PCDD/F and PBDE concentrations in fish from the Murchison Bay compared with literature data.

Location	Fish species	PCDD/Fs	References
Murchison Bay	*L. niloticus* and *O. niloticus*	5.32 to 49 pg g^{-1} ww	This study
Napoleon Gulf and Thurston Bay of Lake Victoria, Uganda	*L. niloticus* and *O. niloticus*	0.06–0.59 pg g^{-1} ww	Ssebugere et al. (2013b)
River Turia, Spain	*Anguilla anguilla* and *Salmo trutta*	1.22–4.39 pg g^{-1} ww	Bordajandi et al. (2003)
Sepetibabay, Rio de Janeiro, Brazil	*Lepidopus caudatus*, *Micropogonias furnieri*, and *Mugil cephalus*	0.953–1.455 pg g^{-1} ww	Ferreira (2013)
Lake Volta, Lake Bosumtwi and Weija Lake, Ghana	*Tilapia zillii* and *Clarias gariepinus*	5–26 pg g^{-1} ww	Adu-Kumi et al. (2010)
Río de la Plata estuary, Argentina and Uruguay	*Prochilodus lineatus*, *Cyprinus carpio* and *Mugil cephalus*	n.d. to 27.3 pg g^{-1} ww	Colombo et al. (2000)
Tittabawassee and Saginaw Rivers, USA	*Cyprinus carpio* and *Clarias gariepinus*	20–440 pg g^{-1} ww	Wan et al. (2010)
Masan Bay, South Korea	*Mytilus edulis* and *Cyrtopleura costata*	n.d. to 3418 pg g^{-1} lw	Im et al. (2004)
Qiantangjiang River, China	*Carassius carassius* and *Parabramis pekinensis*	1.14–7.88 pg g^{-1} ww	Han et al. (2007)
Location	**Fish species**	**PBDEs**	**References**
Murchison Bay, Lake Victoria, Uganda	*L. niloticus* and *O. niloticus*	59.3–495 pg g^{-1} ww (48.2 to 177 pg g^{-1} lw).	This study
Weija Lake, Volta Lake, Benya and Keta Lagoons, Ghana	*Oreochromis niloticus*, *Tilapia guineensis*, *Sarotherodon melanotheron*	10–52,000 pg g^{-1} lw	Asante et al. (2013)
Lake Pongolapoort, South Africa	*Hydrocynus vittatus*	4100–5800 pg g^{-1} lw	Wepener et al. (2012)
Pearl River Estuary, South China	*Pseudosiaena crocea*, *Pampus argenteus*, *Platycephalus indicus*, *Cynoglossus robustus*, *Harpodon nehereus*	34,100–1,068,000 pg g^{-1} lw	Xiang et al. (2007)
Baiyangdian Lake, North China	*Pelteobagrus fluvidraco*, *Parasilurus asotus*, *Carassius auratus*, *Aristichthys nobilis*, *Ctenopharyngodon idella*, *Channa argus*, *Monoperus ablus*, *Misgurnus anguillicaudatus*	4100–114,000 pg g^{-1} lw	Hu et al. (2010)
Llobregat River, Spain	*Cyprinus carpio*	29,000–744,000 pg g^{-1} lw	Labandeira et al. (2007)
Orge River, France	*Rutilus rutilus*, *Morone Americana*, *Couesius plumbeus*	12,000–18,000 pg g^{-1} dw	Teil et al. (2012)
Lake Michigan, USA	*Cyprinus carpio*	<1000–230,000 pg g^{-1} ww	Streets et al. (2006)
Columbia River, Oregon and Washington in USA	*Catostomus macrocheilus*	n.d. to 21,600 pg g^{-1} ww	Nilsen et al. (2014)

n.d. — non-detectable.

WHO_{2005}-TEQ values for sediments from the Napoleon Gulf and Thurston Bay of Lake Victoria as 0.98–4.24 and 0.08–0.42 pg TEQ g^{-1} dw, respectively (Ssebugere et al., 2013b). The data herein was in the same range as that for sediments from the Thurston Bay but lower than the data from the Napoleon Gulf. A number of studies have also reported TEQs for sediments elsewhere in the world. In South Africa, values ranging from 0.12 to 32 pg WHO_{2005}–TEQ g^{-1} dw were reported from Rivers Klip, Vaal and Suikerbosrand (Nieuwoudt et al., 2009). In Korea, estimated values between 0.1 and 1590 pg WHO_{2005}-TEQ g^{-1} dw were reported from Lake Shihwa (Moon et al., 2012). In general, our findings are lower compared to those reported in South Africa and Korea. Site 3 had the highest ΣTEQs (mean value 0.33 pg g^{-1} dw) compared to other stations. PCDFs accounted for more than 70% to the ΣTEQs at all sites. The toxicity pattern at sites 1 and 2 was dominated by OCDF while 2,3,7,8-TCDF was dominant at sites 3 and 4 (Fig. 5).

3.2. Concentrations of PCDD/Fs in fish species

The ΣPCDD/Fs varied largely between fish species, with mean values ranging from 5.32 to 49.0 pg g^{-1} ww (4.17 to 17.5 pg g^{-1} lw) (Table 3). The concentrations were higher than those in fresh water fish from River Turia in Spain (Bordajandi et al., 2003), the Napoleon Gulf and Thurston Bay of Lake Victoria (Ssebugere et al., 2013a) and Sepetibabay, Rio de Janeiro in Brazil (Ferreira, 2013). The results of the present study were in the same range of data as for fresh water fish from the Río de la Plata estuary in Argentina and Uruguay (Colombo et al., 2000) and Lakes Volta, Bosumtwi and Weija in Ghana (Adu-Kumi et al., 2010). The PCDD/F levels were however lower than those in fish from Masan Bay in South Korea (Im et al., 2004), Tittabawassee and Saginaw Rivers in the United States of America (Wan et al., 2010) and Qiantangjiang River in China (Han et al., 2007) (Table 4).

The ΣPCDD/F concentrations in *L. niloticus* were greater than those in *O. niloticus* at all study sites. The high concentrations in the former fish than the latter could be due to species differences in feeding habits and lifestyles. The *O. niloticus* depend on phytoplankton and detritus, while *L. niloticus* feed on smaller fish, insects, mollusks, aquatic crustaceans and zooplankton (Dadebo et al., 2005; Ogari and Dadzie, 1988). Furthermore, *L. niloticus* showed high lipid contents compared to *O. niloticus* from the same sites (Table 3). In an earlier study, fish with high lipid content were reported to accumulate POPs than the ones with less lipid content (Ssebugere et al., 2009). The authors also established that the lipid content is directly correlated to the weight and length of fish. In the present study, the weights of *O. niloticus* ranged from 289 to 652 g (mean 389 g) while those of *L. niloticus* varied from 1628 to 2979 g (mean 2378 g). The mean lengths (±standard deviation) were 29 ± 3 and 68 ± 5 cm, respectively. Statistical analysis of the PCDD/F concentrations in *L. niloticus* showed a significant difference ($p < 0.05$, Mann–Whitney *U* test) for sites 1 and 4 while the concentrations for *O. niloticus* were insignificant.

The maximum concentrations of the ΣPCDFs were 12.0 and 38.4 pg g^{-1} ww for *O. niloticus* and *L. niloticus*, respectively. The highest mean concentration of the ΣPCDFs was found in fish from site 1 (26.1 pg g^{-1} ww). A decreasing trend in concentrations was then observed for other sites as the distance away from the mouth of the Nakivubo Channel increased. The ΣPCDDs ranged from 4.57 to 46.9 pg g^{-1} ww for *L. niloticus* and from 2.02 to 19.1 pg g^{-1} ww for *O. niloticus*. Of the PCDD congeners, only OCDD and 1,2,3,4,6,7,8-HpCDD were detected, and their levels could be attributed to the inability of the enzyme system of fish to biodegrade the congeners (Wu et al., 2001).

In the present study, only five congeners (1,2,3,6,7,8-HxCDF, 1,2,3,4,6,7,8-HpCDF, OCDF, 1,2,3,4,6,7,8-HpCDD and OCDD) contributed to the ΣTEQs in fish. OCDD was more predominant compared to the other four congeners (it contributed >38% to the ΣTEQs at all locations in both fish species). Recently, European Community authorities have set a maximum value of 3.5 pg WHO-TEQ g^{-1} ww for PCDD/Fs in muscle meat of fish and fishery products (EC, 2011). The value is far over the TEQs found for *L. niloticus* (range 0.001–0.019 pg WHO-TEQ g^{-1} ww) and *O. niloticus* (0.001–0.033 pg WHO-TEQ g^{-1} ww), suggesting that the fish was fit for human consumption with regard to PCDD/Fs.

The literature on the TEQ levels of PCDD/Fs in fresh water from Africa is very scarce. In Uganda, Ssebugere et al. (2013a) estimated TEQ values in fish (*L. niloticus* and *O. niloticus*) from the Napoleon Gulf and Thurston Bay of Lake Victoria, ranging from 0.01 to 0.16 pg TEQ g^{-1} ww. In Ghana, an average value of 0.3 pg TEQ g^{-1} ww was reported in fish (*Tilapia zillii and Clarias gariepinus*) from Lakes Volta, Bosumtwi and Weija (Adu-Kumi et al., 2010). The TEQ values in both studies are in the same range of the data like in the present work.

3.3. Levels of PBDEs in sediments and fish species

The levels of 11 BDE congeners (BDEs 17, 28, 47, 66, 85, 99, 100, 138, 153, 154 and 183) in sediments from the Murchison Bay are summarized in Table 1. It should be noted that BDE 209 a commonly detected congener in sediments was not eluted by our column and therefore its concentrations are not reported. The ΣPBDEs varied widely between stations, with mean values ranging from 60.8 to 179 pg g^{-1} dry weight (dw). The concentrations in this study were lower than the data in sediments from the Congo River Basin in the Democratic Republic of Congo (Verhaert et al., 2013), Juksei River in South Africa (Olukunle et al., 2012), Niagara River in Canada (Samara et al., 2006), Ebro river basin

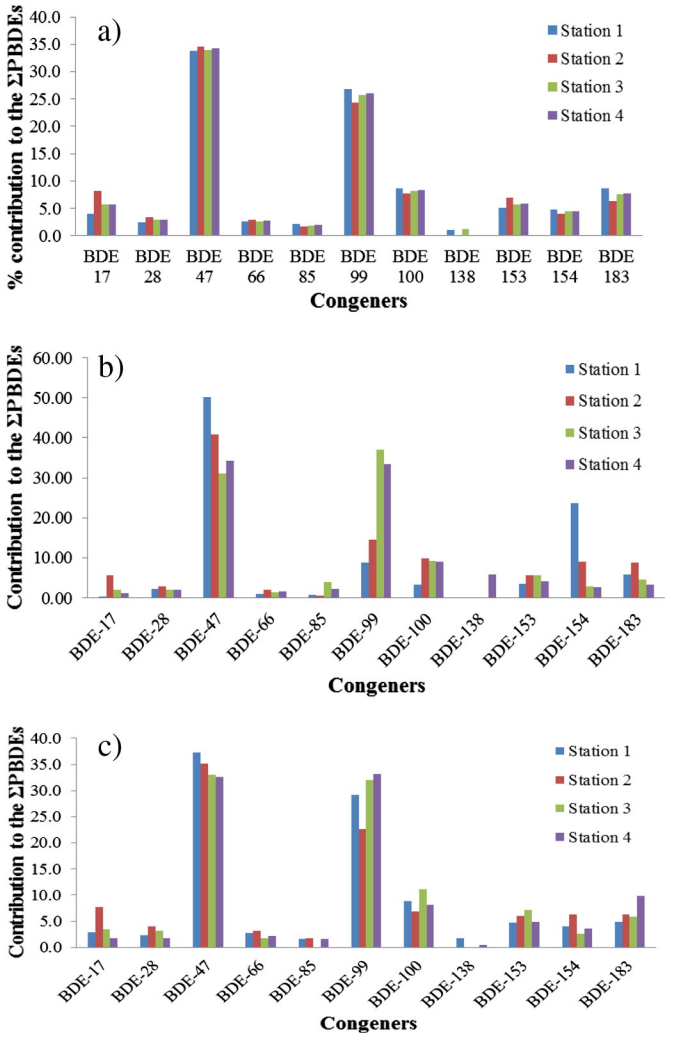

Fig. 6. Contribution of the different congeners to the ΣPBDEs. a) In sediments. b) In *L. niloticus*. c) In *O. niloticus*.

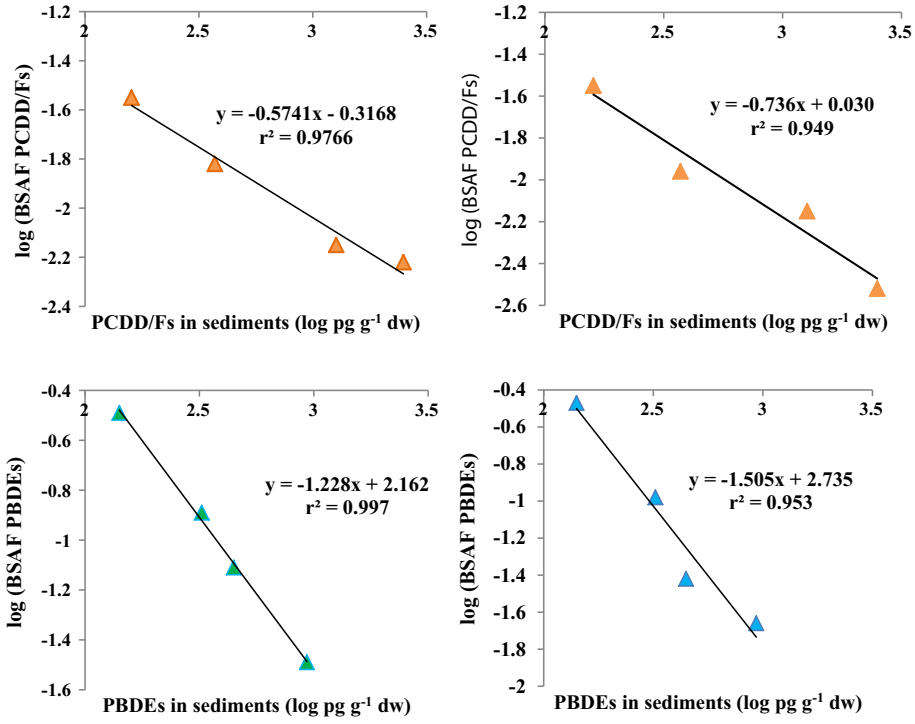

Fig. 7. Relationship between sediment concentrations and BSAF for PCDD/Fs and PBDEs. a) Left hand side — *L. niloticus*. b) Right hand side — *O. niloticus*.

in Spain (Lacorte et al., 2006) and the Columbia River in the United States (Counihan et al., 2014).

Tetra-BDE 47 was the most abundant congener (Fig. 6). The high levels of this congener could be due to the fact that it is resistant to microbial degradation in the environment (Hale et al., 2006). The contribution by BDE 47 was followed by penta-BDE 99 (24.4–26.9% of the ΣPBDEs) and then penta-BDE 100 (7.7–8.6% of the total amount). The use of mixtures of penta- and deca-BDEs in the tannery and textile industries is the major source of BDE 47, 99 and 100 in the environment (Eljarrat and Barceló, 2012). In both applications, deca-BDE formulations are used in a water-based binder dispersion system, which would lead to waste water emissions in the aquatic environment (Guzzella et al., 2008). Congeners such as BDEs 17, 28, 66, 85, 138, 153, 154 and 184 were also detected, but at lower concentrations. Just like the PCDD/Fs, the PBDE concentrations at site 1 were higher than the data at other sites. However, a statistical test between the different sites showed no significant differences in PBDE concentrations.

The present study also determined PBDEs in fish at concentration ranges of 59.3 to 495 pg g^{-1} ww (48.2 to 177 pg g^{-1} lw). The PBDE levels were lower than the data reported in fish from Ghana (Asante et al., 2013), South Africa (Wepener et al., 2012), China (Hu et al., 2010; Xiang et al., 2007), Spain (Labandeira et al., 2007; Lacorte et al., 2006), France (Teil et al., 2012) and the United States (Nilsen et al., 2014; Streets et al., 2006) (Table 4). BDE 47 the most predominant congener in the sediments was also abundant in fish (Fig. 6). Similarly, BDE 99 another highly contributing congener in sediments (24.4 to 26.9% of the ΣBDE) was also abundant in fish.

To establish the relationship between the pollutant concentrations in sediments and fish, the biota-sediment accumulation factors (BSAFs) were calculated. The BSAF values at the different study locations varied from 0.003 to 0.028 for PCDD/Fs and 0.022 to 0.352 for PBDEs. No significant differences in BSAF values between different fish species were found. To evaluate the applicability of the BSAF concept for risk assessment, correlations between sediment concentrations and BSAFs were analyzed using log-transformed data (Bervoets et al., 2005). A significant inverse relationship was found between the sediment PCDD concentrations and BSAF values in *L. niloticus* ($r^2 = 0.976$) and *O. niloticus* ($r^2 = 0.949$) (Fig. 7). The r^2 values for PBDEs were 0.997 and 0.953, respectively. Therefore, the BSAF values indicate no association between the fish body burden and the contaminants derived from sediments.

The Agency for Toxic Substances and Disease Registry has determined minimum risk levels (MRLs) for oral intake of POPs (ATDSR, 1996). With the levels, the maximum amount of fish which can be consumed without risk for an average person of 70 kg is calculated with the observed concentration of PCDD/Fs and PBDEs in fish from the Murchison Bay (Table 5). A person of 70 kg who consumes more than 70 g day^{-1} of either *L. niloticus* or *O. niloticus* was below the MRL of 0.02 and 7000 ng kg^{-1} body weight day^{-1} for PCDD/Fs ng kg^{-1} and PBDEs, respectively, indicating no health risks associated with the consumption of fish.

Table 5
Maximum amounts which are recommended to eat without risk of pollution for an average person of 70 kg based on MRLs (ATDSR, 1996) and mean concentrations of PCDD/Fs and PBDEs found in fish from Lake Victoria.

	ΣPCDD/Fs	ΣPBDEs
MRL (ng kg^{-1} body weight day^{-1})	0.02	7000
MRL (ng/day) for a person of 70 kg	1.40	490,000
Mean concentration in *L. niloticus* (ng g^{-1} ww)	0.02	0.23
Mean concentration in *O. niloticus* (ng g^{-1} ww)	0.01	0.08
Maximum edible amount of *L. niloticus* per day for a person of 70 kg	70	2,130,434
Maximum edible amount of *O. niloticus* per day for a person of 70 kg	140	6,125,000

4. Conclusion

The present study constitutes the first report on the levels of PCDD/Fs and PBDEs in sediments and fish from the Murchison Bay of Lake Victoria. Based on the European Commission set TEQ value and minimum risk level criteria formulated by the Agency for Toxic Substances and Disease Registry, the fish from the Murchison Bay was of no harm to human consumers with regard to PCDD/Fs and PBDEs. The

concentrations in sediments and fish were lower to moderate compared to those of literature. However, even though the pollutant levels are still low, the results provide evidence for decision makers to establish priorities to reduce any further entry of the pollutants into Lake Victoria, because the effect of long-term exposure of these chemicals to humans and the aquatic ecosystem cannot be ascertained.

Conflict of interest

There is no conflict of interest.

Acknowledgments

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