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PERSISTENT ORGANIC POLLUTANTS IN SEDIMENTS AND FISH FROM LAKE VICTORIA, UGANDA

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ABSTRACT

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This thesis describes the occurrence and sources of selected persistent organic pollutants (POPs) such as polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and hexachlorocyclohexanes (HCHs) in the northern watershed of Lake Victoria. Sediments and fish were collected from three highly polluted embayments (i.e. Murchison Bay, Napoleon Gulf and Thurston Bay) of the lake. The analysis for PCDD/Fs, PCBs and PBDEs was done using a high resolution mass spectrometer coupled to a gas chromatograph (GC), and a GC equipped with an electron capture detector was used for HCHs. Total (Σ) PCDD/Fs, PCBs and PBDEs in sediments ranged from 3.19 to 478, 313 to 4325 and 60.8 to 179 pg g^{-1} dry weight (dw), respectively. The highest concentrations of pollutants were found at sites close to industrial areas and wastewater discharge points. The maximum concentrations of PCDD/Fs, PCBs, PBDEs and HCHs in fish muscle homogenates were 49, 779, 495 and 45,900 pg g^{-1} wet weight (ww), respectively. The concentrations of the pollutants in Nile perch (*Lates niloticus*) were significantly greater than those in Nile tilapia (*Oreochromis niloticus*), possibly due to differences in trophic level and dietary feeding habits among fish species. World Health

Organization-toxic equivalency quotient (WHO$_{2005}$-TEQ) values in the sediments were up to 4.24 pg g^{-1} dw for PCDD/Fs and 0.55 pg TEQ g^{-1} dw for the 12 dioxin-like PCBs (dl-PCBs). 23.1% of the samples from the Napoleon Gulf were above the interim sediment quality guideline value of 0.85 pg WHO-TEQ g^{-1} dw set by the Canadian Council for Ministers of the Environment. The WHO$_{2005}$-TEQs in fish were 0.001-0.16 pg g^{-1} for PCDD/Fs and 0.001-0.31 pg g^{-1} ww for dl-PCBs. The TEQ values were within a permissible level of 3.5 pg g^{-1} ww recommended by the European Commission. Based on the Commission set TEQs and minimum risk level criteria formulated by the Agency for Toxic Substances and Disease Registry, the consumption of fish from Lake Victoria gives no indication of health risks associated to PCDD/Fs and PCBs. Principal component analysis (PCA) indicated that anthropogenic activities such as agricultural straw open burning, medical waste incinerators and municipal solid waste combustors were the major sources of PCDD/Fs in the watershed of Lake Victoria. The ratios of α-/γ-HCH varied from 0.89 to 1.68 suggesting that the highest HCH residues mainly came from earlier usage and fresh γ-HCH (lindane). In the present study, the concentration of POPs in fish were not significantly related to those in sediments, and the biota sediment accumulation factor (BSAF) concept was found to be a poor predictor of the bioavailability and bioaccumulation of environmental pollutants.

Key words: Lake Victoria, sediments, fish, POPs, PCDD/Fs, PCBs, PBDEs, HCHs, sources, bioaccumulation, TEQs, BSAFs, PCA.

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LIST OF PUBLICATIONS

This thesis is based on articles listed below which are referred to in the text by their roman numerals. Papers 1-V are reproduced with permission from Elsevier.

I. Ssebugere, P.; Kiremire, B.T.; Henkelmann, B.; Bernhöft, S.; Wasswa, J.; Kasozi, G.N.; Schramm, K.-W., PCDD/Fs and dioxin-like PCBs in surface sediments from Lake Victoria, East Africa. *Science of The Total Environment* **2013**, *454-455*, 528-33.

II. Ssebugere, P.; Kiremire, B.T.; Henkelmann, B.; Bernhöft, S.; Kasozi, G.N.; Wasswa, J.; Schramm, K.-W., PCDD/Fs and dioxin-like PCBs in fish species from Lake Victoria, East Africa. *Chemosphere* **2013**, *92*(3), 317-21.

III. Ssebugere, P.; Sillanpää, M.; Kiremire, B.T.;Kasozi, G.N.; Wang, P.; Sojinu, S.O.; Otieno, P.O.; Zhu, N.; Zhu, C.; Zhang, H.; Shang, H.; Ren, D.; Li, Y.; Zhang, Q.; Jiang,G., Polychlorinated biphenyls and hexachlorocyclohexanes in sediments and fish species from the Napoleon Gulf of Lake Victoria, Uganda. *Science of The Total Environment* **2014**, *481*, 55-60.

IV. Ssebugere, P.; Sillanpää, M.; Wang, P.; Li, Y.; Kiremire, B.T.; Kasozi, G.N.; Zhu, C.; Ren, D.; Zhu, N.; Zhang, H.; Shang, H.; Zhang, Q.; Jiang, G., Polychlorinated biphenyls in sediments and fish species from the Murchison Bay of Lake Victoria, Uganda. *Science of The Total Environment* **2014**, *482-483*, 349-57.

V. Ssebugere, P.; Sillanpää, M.; Wang, P.; Li, Y.; Kiremire, B.T.; Kasozi,G.N.; Zhu, C.; Ren, D.; Shang, H.; Zhang, Q.; Jiang, G., Polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans and polybrominated diphenyl ethers in sediments and fish species from the Murchison Bay of Lake Victoria, Uganda. *Science of The Total Environment* **2014**, *500-501*, 1-10.

Author´s contribution to the publications

Papers I-V: The author planned, designed, participated in sampling, carried out experiments, analysed data, interpreted results and prepared first drafts of all the manuscripts.

RELATED PUBLICATIONS

a) Zhu, N.; Fu, J.; Gao, Y.; Ssebugere, P.; Wang, Y.; Jiang, G., Hexabromocyclododecane in alpine fish from the Tibetan Plateau, China. *Environmental Pollution* **2013**, *181*, 7-13.

b) Wang, P.; Zhang, H.; Fu, J.; Li, Y.; Wang, T.; Wang, Y.; Ren, D.; Ssebugere, P.; Zhang, Q.; Jiang, G., Temporal trends of PCBs, PCDD/Fs and PBDEs in soils from an e-waste dismantling area in East China. *Environmental Science: Processes & Impacts* **2013**, *15*, 1897-1903.

c) Zhu, C.; Wang, P.; Li, Y.; Chen, Z.; Li, W.; Ssebugere, P.; Zhang, Q.; Jiang, G., Bioconcentration and trophic transfer of polychlorinated biphenyls and polychlorinated dibenzo-*p*-dioxins and dibenzofurans in aquatic animals from an e-waste dismantling area in East China. *Environmental Science: Processes & Impacts* **2015**, *17*, 693 - 699.

d) Zhu, C.; Li, Y.; Wang, P.; Chen, Z.; Ren, D.; Ssebugere, P.; Zhang, Q.; Jiang, G. Polychlorinated biphenyls (PCBs) and polybrominated biphenyl ethers (PBDEs) in environmental samples from Ny-Ålesund and London Island, Svalbard, the Arctic. *Chemosphere* **2015**, *126*, 40-46.

TABLE OF CONTENTS

ACRONYMS AND ABBREVIATIONS

ASE	Accelerated solvent extractor
BSAF	Biota-sediment accumulation factor
CCME	Canadian Council for Ministers of the Environment
d.w.	dry weight
DCM	Dichloromethane
DDT	1,1,1-trichloro-2,2-bis[*p*-chlorophenyl]ethane
dI-PCB	dioxin-like PCB
GC	Gas chromatography
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
HpCDD	Hexachlorinateddibenzo-*p*-dioxin
HpCDF	Hexachlorinateddibenzofuran
HRMS	High resolution mass spectrometry
i.d	inside diameter
l.w.	lipid weight
MAAIF	Ministry of Agriculture, Animal Industries and Fisheries
MS	Mass spectrometer
NEMA	National Environnent Management Authority
OCDD	Octachlorinateddibenzo-*p*-dioxins
OCDF	Octachlorinateddibenzofuran
PBDEs	Polybrominated diphenyl ethers
PCA	Principal Component Analysis

PCB	Polychlorinated biphenyl
PCDD/F	Polychlorinated dibenzo-*p*-dioxin/furan
PeCDD	Pentachlorinated dibenzo-*p*-dioxin
PeCDF	Penta-chlorinated dibenzofuran
POP	Persistent organic pollutant
TEF	Toxic equivalency factor
TEQ	Toxic equivalence quotient
US EPA	United States Environmental Protection Agency
WHO	World Health Organization

1. INTRODUCTION

1.1 Study background

Persistent organic pollutants (POPs) are substances that possess a combination of physical and chemical properties which include undergoing long-range atmospheric transport[1], accumulating in fatty tissue of living organisms[2, 3] and are difficult to degrade in the environment[4]. The twelve POPs listed for initial elimination and reduction in usage under the Stockholm Convention in 2001 included dieldrin, chlordane, aldrin, dichlorodiphenyltrichloroethane (DDT), endrin, heptachlor, mirex, toxaphene, hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs)[5]. Today, twelve more POPs have been added on the initial 2001 Stockholm Convention list (Table 1.1).

Table 1.1: A list of the Stockholm Convention POPs.

Annex A (elimination)	Chemicals
Industrial chemicals	Hexabromobiphenyl, hexa- and hepta bromodiphenyl ether, PCBs, tetra- and penta-bromodiphenyl ether
Pesticides	Aldrin, chlordane, dieldrin, endrin, heptachlor, toxaphene, mirex, lindane (γ-hexachlorocyclohexane)
Pesticides/Industrial chemicals	HCB, pentachlorobenzene
Pesticides/Unintentional by-products	α-hexachlorocyclohexane, β-hexachlorocyclohexane
Annex B (restriction)	
Pesticide	DDT
Industrial chemicals	Perfluorooctane sulfonic acid (PFOS) and salts, and perfluorooctane sulfonyl fluoride
Annex C (unintentional production)	
Unintentional by-products	PCDDs, PCDFs, HCB, pentachlorobenzene, PCBs

The sources of POPs include agriculture, waste incineration, industrial chemical processes, unregulated disposal of textiles, building materials and obsolete electronic waste (e-waste) and, burning of waste and vegetation [6-10]. Once released into aquatic systems, POPs except the PFOS tend to accumulate in sediments for a long period of time and are subject to partitioning, degradation and transport processes[11]. Relative to the water column, surface sediments are a more appropriate environmental compartment that can be related to the concentrations of POPs in aquatic organisms[12]. These chemicals have the potential to bio-accumulate across the food chain, building up in top predators through consumption of contaminated biota[13, 14]. The fish at the top of the aquatic food web are more likely to be affected by exposure to such pollutants[15, 16]. Different species of fish occupy different habitats in the same ecosystem and have different feeding behaviors. As a result, they are used as a good proxy to assess the influence of the environment and biological factors concerning the bioaccumulation of pollutants[17, 18]. Understanding the primary factors influencing the bioaccumulation of these compounds in fish from aquatic ecosystems like Lake Victoria is crucial in predicting and assessing risks to upper-trophic level consumers including humans.

In recent years the ecological health of Lake Victoria is being threatened, mainly by rapid urbanization and industrialization. These developments coupled with a population of over one million people in its catchment area have resulted into increased pollution[19, 20]. In addition, catchment wetlands which previously played the vital role of tertiary purification of effluent before discharging it into the lake have long been encroached on for settlement[20]. Persistent organic pollutants such as dieldrin, chlordane, aldrin, DDT, endrin, heptachlor, mirex, toxaphene and HCB have previously been documented in the lake[20]. However, a survey of literature shows no available data concerning the levels, sources, relationships and toxicological implications of PCDD/Fs, PCBs, PBDEs and HCHs in the aquatic environment of Lake Victoria.

1.2 Importance of Lake Victoria

Lake Victoria is the world's second largest freshwater lake after Lake Superior in North America. The lake covers a surface area of approximately 68,800 km^2 and is shared between Kenya (6%), Uganda (45%) and Tanzania (49%). Catchment area of the lake covers 180,959 km^2 of which 15.9% of this is constituted by Uganda. The rest of the watershed is shared by Tanzania (44%), Kenya (21.5%), Rwanda (11.4%) and Burundi (7.2%). Economically, millions of people depend on the lake as a source of drinking water and fish as a principal source of proteins to the local diet. The most highly caught and consumed fish are Nile perch (*Lates niloticus*), Nile tilapia (*Oreochromis niloticus*) and Silver fish (*Rastreneobola argentea*) (Table 1.2).

Table 1.2: Estimated annual catches (tones) and value (million dollars) from 2008 to 2011[21].

Fish species	2008		2010		2011	
	Catch	Value	Catch	Value	Catch	Value
L. niloticus	80,977	72.3	84,969	119	70,061	131
O. niloticus	19,557	11.1	16,971	14.8	19,350	21.7
R. argentea	70,001	7.62	58,717	6.46	88,808	18.4
Other species	2489	1.17	2270	1.79	3605	3.20
Total	173,024	92.2	162,929	142	183,824	174

Lake Victoria provides employment opportunities to approximately two million fishmongers, fishermen and people employed in fish processing. Therefore, the lake has indirect and direct impact on poverty reduction efforts in Uganda and her neighbors. The lake's fisheries contribute an annual foreign income of USD $124 million to Uganda's economy constituting the third foreign earner, after coffee and flowers[22]. The main export destinations for frozen fish include the United States of America, Europe, Hong Kong, Japan, Singapore, Israel, United Arab Emirates, Egypt and South Africa.

The bank of Lake Victoria is home to many perennial and migratory bird species and, fauna like hippopotamus and crocodiles. The lake's biodiversity combined with its scenic beauty, is one of the important contributors to increased tourist arrivals in Uganda and her neighbors. The lake also influences climatic conditions in the Basin. Furthermore, the lake provides the cheapest linkage for trade, water for power generation along the River Nile and, water for domestic and industrial purposes in most of the urban areas within Uganda and in the region.

1.3 Thesis objectives

The major objective of this study was to investigate the occurrence and bioaccumulation of selected POPs (i.e. PCDD/Fs, PCBs, PBDEs and HCHs) in sediments and fish from the northern waters of Lake Victoria. Moreover, the specific objectives were:

(i) to produce a baseline dataset for POP concentrations and their distribution in sediments and fish of Lake Victoria,

(ii) to identify the possible sources of the selected POPs,

(iii) to determine the potential human health risks through consumption of fish from Lake Victoria,

(iv) to evaluate the biota-sediment accumulation factors for these samples.

2. LITERATURE REVIEW

This chapter aims to briefly describe the sources, toxicology and occurrence of POPs included in articles I-V.

2.1 Polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs)

2.1.1 Physico-chemical properties, sources and toxicology of PCDD/Fs

PCDD/Fs (Fig. 2.1) are unintentional byproducts produced during metal smelting[23], forest fires[24], fossil fuel combustion, synthesis of chloro-phenols, production of chloro-organic pesticides and pulp bleaching in the process of paper production[25]. PCDD/Fs consist of 210 theoretical congeners (75 PCDDs and 135 PCDFs), commonly referred to as "dioxins". Some of physical-chemical properties of PCDD/Fs are given in Table 2.1.

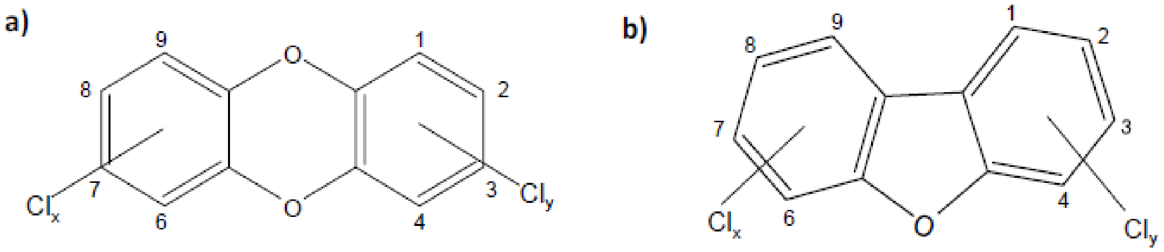


Fig 2.1: General structure of a) PCDDs and b) PCDFs.

Table 2.1: Physico-chemical properties for PCDD/Fs[26, 27].

Congener	Melting point at 25°C	Solubility in water at 22.7 or 25°C (mg L^{-1})	Vapour pressure at 25°C (Pa)	Log K_{ow}
2,3,7,8-TCDD	305-306	1.93×10^{-3}	2.0×10^{-7}	6.80
1,2,3,7,8-PeCDD	240-241	1.93×10^{-3}	5.8×10^{-8}	6.64
1,2,3,4,7,8-HxCDD	273-275	4.42×10^{-6}	5.1×10^{-9}	7.80
1,2,3,6,7,8-HxCDD	283-286	4.42×10^{-6}	4.8×10^{-9}	7.80
1,2,3,7,8,9-HxCDD	243-244	4.42×10^{-6}	6.5×10^{-9}	7.80
1,2,3,4,6,7,8-HpCDD	264-265	2.40×10^{-6}	7.5×10^{-10}	8.00
OCDD	325-326	0.75×10^{-7}	1.1×10^{-8}	8.20
2,3,7,8-TCDF	227-228	4.19×10^{-4}	2.0×10^{-6}	6.53
1,2,3,7,8-PeCDF	225-227	4.19×10^{-4}	2.3×10^{-7}	6.79
2,3,4,7,8-PeCDF	196-197	2.36×10^{-4}	3.5×10^{-7}	6.92
1,2,3,4,7,8-HxCDF	226-227	8.25×10^{-6}	3.2×10^{-8}	6.92
1,2,3,6,7,8-HxCDF	232-234	1.77×10^{-6}	2.9×10^{-8}	6.92
1,2,3,7,8,9-HxCDF	246-249	1.77×10^{-6}	2.4×10^{-8}	6.92
2,3,4,6,7,8-HxCDF	239-240	1.77×10^{-6}	2.6×10^{-8}	6.92
1,2,3,4,6,7,8-HpCDF	236-237	1.35×10^{-6}	4.7×10^{-9}	7.92
1,2,3,4,7,8,9-HpCDD	221-223	1.35×10^{-6}	6.2×10^{-9}	7.92
OCDF	258-260	1.16×10^{-6}	5.0×10^{-9}	8.78

PCDD/Fs are characterized by high boiling points and are thus semi-volatile, are persistent and therefore stable to chemical and biological transformations[28]. The congeners of the compounds are more or less lipophilic and bioaccumulative, but some of them are relatively easy to degrade. The position and degree of chlorination affects the rate and degree of metabolism. The 2,3,7,8-substituted PCDD/Fs (fully laterally chlorinated ones) lack adjacent hydrogen atoms and are thus highly resistant to metabolism[29]. Consequently, the 2,3,7,8-substituted PCDD/Fs are usually the only congeners present in living organisms[30]. The bioaccumulation potential of chlorinated aromatic hydrocarbons generally increases with degree of chlorination. However, in some cases, the most highly chlorinated molecules are bioaccumulated less than expected and this may be due to size-limitations in cell membrane diffusion processes and/or to low water solubility limiting passage over intestinal walls of the molecules[30].

Seventeen of the PCDD/F congeners (7 PCDDs and 10 PCDFs) are substituted with chlorine in the 2,3,7,8-positions, and are of public concern because they have been implicated in mutagenic and carcinogenic effects such as liver damage, malignant melanoma and preneoplastic and neoplastic changes[31, 32]. Other health concerns related to PCDD/Fs include gastrointestinal (gastric hyperplasia, ulceration, necrosis), respiratory (chronic bronchitis and coughs), dermal (chloracne, oedema, alopecia, hyperkeratosis of epithelium), neurotoxic (impaired behavioural responses, depressed motor activity, developmental deficits, numbness) and immunotoxic (lymphoid tissue atrophy, leukocyte and lymphocyte reduction, suppressed antibody responses), hepatotoxic (hepatomegaly, hyperplasia of the bile duct, necrosis, fatty degeneration, porphyria) and reproductive impairment (decreased sperm count and motility, increased miscarriages, decreased survival and mating success)[33].

Toxic actions of the 2,3,7,8-substituted congeners is mediated via mechanisms where the congeners bind to the aryl-hydrocarbon (Ah) receptor[34]. Studies have reported that PCDD/Fs can activate the Ah-receptor in the cytoplasm of most vertebrates and cause a range of biological and toxic effects[35, 36]. Due to the pronounced mode of action and toxicity via the Ah-receptor, PCDD/Fs and the dioxin-like PCBs have been assigned toxic equivalent factors (TEFs) that are used to calculate the total toxic equivalencies (TEQs). In this system 2,3,7,8-TCDD, which is more toxic is assigned a TEF value of unity and other congeners are given values depending on their toxicity (Table 2.2). The TEQ value can then be got by multiplying the concentration of the individual congener by its respective TEF.

Table 2.2: WHO-TEF values for PCDD/Fs[37].

Congener	WHO_{1998}-TEF	WHO_{2005}-TEF
2,3,7,8-TCDD	1	1
1,2,3,7,8-PeCDD	1	1
1,2,3,4,7,8-HxCDD	0.1	0.1
1,2,3,6,7,8-HxCDD	0.1	0.1
1,2,3,7,8,9-HxCDD	0.1	0.1
1,2,3,4,6,7,8-HpCDD	0.01	0.01
OCDD	0.0001	0.0003
2,3,7,8-TCDF	0.1	0.1
1,2,3,7,8-PeCDF	0.05	0.03
2,3,4,7,8-PeCDF	0.5	0.3
1,2,3,4,7,8-HxCDF	0.1	0.1
1,2,3,6,7,8-HxCDF	0.1	0.1
1,2,3,7,8,9-HxCDF	0.1	0.1
2,3,4,6,7,8-HxCDF	0.1	0.1
1,2,3,4,6,7,8-HpCDF	0.01	0.01
1,2,3,4,7,8,9-HpCDF	0.01	0.01
OCDF	0.0001	0.0003

2.1.2 Occurrence of PCDD/Fs in sediments and fish

Literature on the PCDD/F levels in sediments from water bodies in Africa is very scarce. In Egypt, El-Kady et al.[38] reported detectable background concentrations ranging from 240 to 775 pg g^{-1} dry weight (dw) in sediments from River Nile. In South Africa, Nieuwoudt et al.[39] found average values varying from 1.4 to 183 pg g^{-1} dw in sediments from the Vaal River. The PCDD/F values in the two studies were in the same range of data as those reported in sediments from Lake Awassa in Ethiopia[40] and the Winam Gulf of Lake Victoria, Kenya[41]. Related studies have also documented PCDD/Fs in sediments from water bodies in the United States of America[42-45], Europe [46-52] and Asia[8, 50, 53-58] (Table 2.3).

Table 2.3: PCDD/F concentrations in sediments from different locations in the world.

Area	Concentrations (pg g^{-1} dw)	References
Nile River, Egypt	240-775	El-Kady et al.[38]
Vaal River, South Africa	1.4-183	Nieuwoudt et al.[39]
Lake Awassa, Ethiopia	n.d. to 270	Urbaniak and Zalewski[40]
Winam Gulf of Lake Victoria, Kenya	38-860	Omwoma et al. [41]
Kentucky Lake, USA	580-1300	Loganathan et al.[42]
Kentucky Lake, USA	n.d. to 4830	Kannan et al.[43]
Guaymas Basin and Patzcuaro Lake, Mexico	12-44	Yunuén et al.[45]
Lake Superior	5-18,000	Shen et al.[44]
Lake Maggiore in Italy and Switzerland	n.d. to 400	Vives et al.[45]
Mondego estuary, Portugal	n.d. to 110	Nunes et al.[46]
Swedish coast of the Baltic Sea	n.d. to 200,000	Sundqvist et al.[48]
Polish rivers, Poland	146-1424	Niemirycz et al.[49]
Gdansk Basin, Baltic sea	11-181	Niemirycz et al.[49]
Lake Baikal, Russia	n.d. to 20	Ok et al.[51]
Oder River and Akona Basin, Western Baltic sea	2.5-820	Dannenberger et al.[52]
Can Gio, Southern Vietnam and Osaka, Japan	264-11,000	Kishida et al.[53]
Mai Po and Inner Deep Bay Ramsar site, Hong Kong, China	5000-7000	Müller et al.[50]
Xiangjiang River, China	876-497,759	Chen et al.[54]
Baiyin City, Northwest China	20-496	Hu et al.[55]
Lake Shihwa, Korea	1-1770	Moon et al.[8]
Busan port, Korea	186-383	Jeong et al.[56]
Coastal lagoons, Central Vietnam	192-2912	Piazza et al.[57]
Tokyo Bay area, Japan	3150-20,300	Sukarai et al.[58]

n.d. - non detectable

PCDD/Fs have also been reported in fish from rivers and lakes in Africa, Asia, United States and Europe. In Africa, Adu-Kumi et al.[59] reported PCDD/F concentrations in two fish species from Lake Volta, Lake Bosumtwi and Weija Lake in Ghana, finding them to occur at average concentrations of 5 to 26 pg g^{-1} wet weight (ww). A number of studies have reported PCDD/F levels in fish from other locations in the world. In Argentina and Uruguay, concentrations varying from non-detectable (n.d.) to 27.3 pg g^{-1} ww were reported in fish from the Río de la Plata estuary [60]. PCDD/Fs were also detected in fish collected from River Turia in Spain, at levels of 1.22 to 4.39 pg g^{-1} ww[61]. In Brazil, PCDD/F concentrations between 0.95 and 1.46 pg g^{-1} ww were reported in three fish species collected from Sepetibabay and Rio de Janeiro[62]. In Japan, concentrations of 0.40 to 2.46 pg g^{-1} ww were reported in five fish species from Lake Kasumigaura and surrounding waters[63]. The concentrations in the above four studies were lower than those in fish from the Masan Bay in South Korea[64], Tittabawassee and Saginaw Rivers in the United States of America[65] and Qiantangjiang River in China[66], but were in the same range of data as those in fish and shell fish from Spain[67] (Table 2.4).

Table 2.4: PCDD/F concentrations in fish species from water bodies in the world.

Location	Fish species	Concentrations (pg g^{-1} ww)	References
Lake Volta, Lake Bosumtwi and Weija Lake in Ghana	*Tilapia zillii* and *Clarias gariepinus*	5-26	Adu-Kumi et al.[59]
Río de la Plata estuary in Argentina and Uruguay	*Prochilodus lineatus*, *Cyprinus carpio* and *Mugil cephalus*	n.d. to 27.3	Colombo et al.[60]
River Turia in Spain	*Anguilla anguilla* and *Salmo trutta*	1.22-4.39	Bordajandi et al.[61]
Sepetibabay, Rio de Janeiro in Brazil	*Lepidopus caudatus*, *Micropogonias furnieri*, and *Mugil cephalus*	0.95-1.46	Ferreira [62]
Tittabawassee and Saginaw Rivers, Michigan, USA	*Cyprinus carpio* and *Clarias gariepinus*	20-440	Wan et al.[65]
Qiantangjiang River in China	*Carassius carassius* and *Parabramis pekinensis*)	1.14-7.88	Han et al.[66]
Masan Bay in South Korea	*Mytilus edulis* and *Cyrtopleura costata*	n.d. to 3418 (pg g^{-1} lw)	Im et al.[64]
Fresh water area, Japan	*Gobiidae*, *Hypomesus olidus*, *Odontesthes bonariensis*, *Cyprinus carpio*, *Opsariichthys uncirostris uncirostris*	0.40-2.46	Sakurai et al.[63]
Spanish markets	*Salmo salar*, *Sardina pilchardus*, *Thunnus Thynnus*, *Cassostrea madrasenssis*, *Chamelea gallina* Venus spp.	0.62-2.89	Gómara et al.[67]

n.d. - non detectable

2.2 Polychlorinated biphenyls (PCBs)

2.2.1 Physico-chemical properties, sources and toxicology of PCBs

PCBs were widely used from 1929, primarily as plasticizers in paints, and dielectric fluids in electric transformers and capacitors due to their low electrical conductivity, high thermal conductivity and resistance to thermal degradation[68, 69]. The industrial production and use of PCBs were banned in the 1980s due to their toxic nature, evidence of widespread environmental contamination and persistence[70, 71]. PCBs can still be formed as unintentional byproducts during waste incineration and industrial processes[72]. In the environment, PCBs have been reported to originate from the same technical mixtures, mostly classified according to their average degree of chlorination[69]. Commercial PCB mixtures consist of mixtures of 30-60 congeners and they show a unique combination of physico-chemical and biological properties (Table 2.5).

Table 2.5: Physical and chemical properties for the selected Aroclors[73-79].

Aroclor compound	Boiling point (°C)	Water solubility at 25°C (mg L^{-1})	Vapour pressure at 25°C (Pa)	Density at 25°C (g cm^{-3})
Aroclor 1016	325-356	4.2×10^{-1}	4.0×10^{-4}	1.33
Aroclor 1221	275-320	5.9×10^{-1}	6.7×10^{-3}	1.15
Aroclor 1232	290-325	4.5×10^{-1}	4.1×10^{-3}	1.24
Aroclor 1242	325-366	2.4×10^{-1}	4.1×10^{-3}	1.35
Aroclor 1248	340-375	5.4×10^{-2}	4.9×10^{-4}	1.41
Aroclor 1254	365-390	2.1×10^{-2}	7.7×10^{-5}	1.50
Aroclor 1260	385-420	2.7×10^{-3}	4.0×10^{-5}	1.58

PCBs constitute 209 theoretically possible congeners containing 1-10 chlorine substituents linked to two phenyl rings (Fig. 2.2). The solubility of PCBs in water is low[80] and they are highly lipophilic. It should be noted that for any PCB molecule, the water solubility decreases and lipophilicity increases with increasing number of chlorine atoms[81] [82].

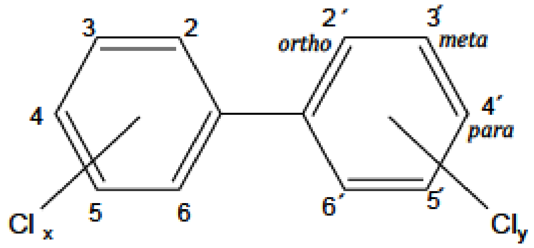


Fig 2.2: General chemical structure of and numbering system for PCBs.

PCBs are divided into three different groups (i.e non-*ortho*-, mono-*ortho*- and indicator-PCBs) depending on the number of chlorine atoms in the *ortho*-position. The non-*ortho*- and mono-*ortho*-PCBs are considered to be dioxin-like because they show similar chemical and toxicological properties to PCDD/Fs[83]. The dioxin-like PCBs (CBs 77, 81, 126, 105, 114, 118, 123, 156, 157, 167 and 189) have been assigned WHO-TEF values (Table 2.6). The TEQ values of most dioxin like-PCBs are considerably lower than those of the toxic dioxins. However, since the PCBs are generally present at higher concentrations, they can still contribute significantly to total (Σ) TEQs. The most toxic PCB congener is CB 126, which often makes the largest contribution to the ΣTEQs.

Table 2.6: WHO-TEF values for dioxin-like PCBs[37].

Non-*ortho* PCBs	WHO_{1998}-TEF	WHO_{2005}-TEF
3,3´, 4,4´-tetraCB (77)	0.0001	0.0001
3,4,4´,5-tetraCB (81)	0.0001	0.0003
3,3´,4,4´,5-pentaCB (126)	0.1	0.1
3,3´,4,4´,5,5´-hexaCB (169)	0.01	0.03
Mono-*ortho* PCBs		
2,3,3´,4,4´-pentaCB (105)	0.0001	0.00003
2,3,4,4´,5-pentaCB (114)	0.0005	0.00003
2,3´,4,4´,5-pentaCB (118)	0.0001	0.00003
2´,3,4,4´,5-pentaCB (123)	0.0001	0.00003
2,3,3´,4,4´,5-hexaCB (156)	0.0005	0.00003
2,3,3´,4,4´,5´-hexaCB (157)	0.0005	0.00003
2,3´,4,4´,5,5´-hexaCB (167)	0.00001	0.00003
2,3,3´,4,4´,5,5´-heptaCB (189)	0.0001	0.00003

Exposure to mono- and di-*ortho* PCB congeners may elicit toxic responses in animals, such as neurochemical[84, 85], neurobehavioural[86], carcinogenic[87] and endocrinological deficits [88].

Studies have suggested that the direction and magnitude of change in neurological function by PCBs depends on the developmental status of the animal at the time of exposure[85, 89]. Prenatal exposure to Aroclor 1016 leads to long-term dysfunctions in behavior of non-human primate[90, 91] and induces persistent increase in biogenic amine concentrations in rats[85]. Perinatal exposure of laboratory animals to PCBs has been associated to carcinogenic effects such as preneoplastic lesions, neoplastic nodules and hepatocellular carcinomas[92]. Parent coplanar and non planar PCBs have been reported to cause marked endocrine changes (such as decreased plasma thyroid hormone and vitamin A levels) in laboratory animals[93, 94]. Alterations in thyroid hormone and vitamin A concentrations may modulate tumour development and adult neurobehavioural changes[95]. Other adverse effects caused by these pollutants include reduction in reproductive capacity, induction of hyperactivity and decrease in learning capacity in mice and rats[96]. Further to this, PCBs alter bone morphology, composition and strength, causing cell apoptosis in male mice, and alternations in fish swimming behavior[97].

2.2.2 Occurrence of PCBs in sediments and fish

A number of studies have reported PCB concentrations in sediments from water bodies in Africa. In Egypt, El-Kady et al.[38] reported levels ranging from 1461 to 2244 pg g^{-1} dry weight (dw) in sediments from River Nile. The PCB concentrations in River Nile were comparable to the data from Ghar El Melh lagoon in Tunisia[98], Pangani River Basin in Tanzania[99], Congo River Basin in the Democratic Republic of Congo[100] and Port Elizabeth Harbour in South Africa[101], but were lower than the concentrations from the Winam Gulf of Lake Victoria in Kenya[41]. Several studies have also documented PCBs in sediments from lakes/rivers in Europe[46, 48, 102-105], the United States of America[106-108]and Asia[109-111] (Table 2.7).

Table 2.7: PCB concentrations in sediments from different water bodies in the world.

Area	PCBs (pg g^{-1} dw)	Congeners	References
Nile River, Egypt	1461-2244	18	El-Kady et al.[38]
Pangani River Basin, Tanzania	357-11,000	28	Hellar-Kihampa et al.[99]
Congo River Basin, Democratic Republic of Congo	n.d. to 1400	33	Verhaert et al.[100]
Ghar El Melh lagoon, Tunisia	n.d. to 3987	20	Ameur et al.[98]
Winam Gulf of Lake Victoria, Kenya	17-812	17	Omwoma et al. [41]
Port Elizabeth Harbour , South Africa	560-2350	6	Kampire et al. [101]
Mekong River delta, Vietnam	110-2000	13	Carvalho et al.[109]
Riverine and coastal waters of Surabaya, Indonesia	n.d. to 420,000	62	Ilyas et al.[110]
Water canals, Hanoi region	n.d. to 40,000	13	Nhan et al.[111]
Scheldt River, Belgium	n.d. to 200,000	27	Covaci et al.[102]
Remote lakes and coastal areas, Norwegian Arctic	180-13,000	15	Jiao et al.[105]
Mersey Estuary, United Kingdom	36,000-1,409,000	7	Vane et al.[103]
Lake Maggiore, Italy and Switzerland	n.d to 3000	12	Vives et al.[46]
Besòs River, Barcelona	6-14	30	Castells et al.[104]
Indiana Harbor and Ship Canal, Lake Michigan, United States of America	53,000-35,000,000	163	Martinez et al.[107]
Salton Sea, California, USA	116,000-304,0000	55	Sapozhnikova et al.[108]
Sea Lots, Port of Spain, Trinidad and Tobago	62,000-601,000	136	Mohammed et al.[106]

A survey of literature showed limited data about the levels of PCBs in fresh water fish from water bodies in Africa. In Burundi, Manirakiza et al.[112] reported PCB concentrations between 35,700 and 166,700 pg g^{-1} lw in fish collected from Lake Tanganyika. The authors noted that the

PCB levels increased with age of fish, indicating bioaccumulation. They concluded that fish length is directly related to the age and that the larger fish are exposed to POPs for longer periods. In Egypt, Said et al.[113] reported varying PCB concentrations of 3320 to 72,060 pg g^{-1} lw in fish from Lake Burullus. Similarly, PCB concentrations ranging from 695 to 853 pg^{-1} ww were documented in fish from River Nile in Egypt[38]. In Nigeria, Adeyemi et al.[114] collected three fish species from Lagos lagoon and analyzed them for PCB presence. They reported concentrations ranging from 560 to 2940 pg^{-1} ww. Studies outside Africa have also reported PCBs in fish. In Hawai'i, PCB levels ranging from 51,900 to 89,420 pg g^{-1} lw were reported in tilapia (*Oreochromis mossambicus*) collected from the Manoa stream and Ala Wai Canal of O'ahu [115]. In South China, Nie et al.[116] reported PCB concentrations ranging from 5150 to 226,000 pg g^{-1} lw in fish from Pearl River Delta. Several other studies have also documented PCBs in fish from the Baltic Sea[117], Danube River in Serbia[118] and rivers in Korea[119] (Table 2.8).

Table 2.8: Concentration of PCBs in fish from different parts in the world.

Location	Fish species	Concentration	Congeners	Reference
Lake Tanganyika, Burundi	*Boulengerochromis microlepis*, *Chrysichthys sianenna*, *Oreochromis niloticus*, *Stolothrissa Tanganikae*, *Limnothrissa miodon* *Lates angustifrons*, *Lates stappersii*	35,700-166,700 (pg g^{-1} lw)	12	Manirakiza et al. [112]
Lake Burullus, Egypt	*O. niloticus* and *Clarias gariepinus*	3320-72,060 (pg g^{-1} lw)	7	Said et al.[113]
River Nile, Egypt	*O. niloticus*	695-853 (pg^{-1} ww)	18	El-Kady et al.[38]
Lagos Lagoon, Nigeria	*Tilapia zilli* , *Ethmalosa fimbriata* and *Chrysichthys nigrodigitatus*	560-2940 (pg^{-1} ww)	8	Adeyemi et al. [114]
Lake Koka, Ethiopia	*Oreochromis niloticus*, *Clarias gariepinus*, *Barbus intermedius*, *Cyprinus carpio* and *Aplocheilichthyes antinorii*	50-330 (pg^{-1} ww)	7	Deribe et al.[120]
Manoa stream and Ala Wai Canal of O'ahu, Hawai'i	*Oreochromis mossambicus*	51,900-89,420 (pg g^{-1} lw)	19	Yang et al. [115]
Pearl River Delta, South China	*Tilapia mossambica*, *Siniperca kneri*	5150-226,000 (pg g^{-1} lw)	36	Nie et al.[116]
Baltic Sea	*Clupea harengus*, *Sprattus sprattus*, *Salmo salar*, *Gadus morhua*, *Platichthys flesus*	358,000-820,000 (pg g^{-1} lw)	7	Szlinder-Richert et al. [117]
Danube River in Serbia	*Silirus glanus*, *Esox lucius*, *Abramis brama*, *Carassius carassius*, *Stezostedion lucio perca*, *barbus barbus*, *Tinca tinca*, *Acipenser ruthenus* L., *Cyprinus carpio*, *Hypophthalmichthys nobilis*	2700-98,100 (pg g^{-1} ww)	6	Janković et al.[118]
Rivers in Korea	*Carassius auratus*	n.d. to 5410 (pg g^{-1} ww)	61	Moon et al. [119]

2.3 Polybrominated diphenyl ethers (PBDEs)

2.3.1 Physico-chemical properties, sources and toxicology of PBDEs

PBDEs were used since the early 1970s as brominated flame retardants in textiles, plastics and electronic equipment. They consists of 209 possible congeners, all are non-planar with 1-10 bromine atoms in the molecule (Fig. 2.3). The physical-chemical properties of PBDEs (Table 2.9) are in many ways similar to those of PCBs. PBDEs are highly hydrophilic with logarithms of octanol/water partition coefficients (log K_{ow}) varying from 5.08 to 8.70 for mono-BDE to deca-BDE[121, 122].

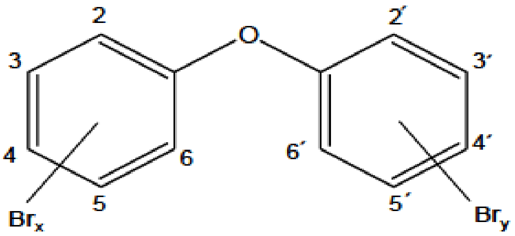


Fig 2.3: General structure of PBDEs.

Table 2.9: Physical and chemical properties of selected PBDE congeners.

Congener	Vapor pressure (mm Hg) [123]	Water solubility (µg L^{-1})[123]	Henry's Law constant (atm-m^3 mol^{-1}) [123]	Log K_{ow}[121]
BDE 3	1.94×10^{-3}	-	-	-
BDE 15	1.30×10^{-4}	130	2.07254×10^{-4}	-
BDE 17	-	-	-	5.74
BDE 28	1.64×10^{-5}	70	5.03331×10^{-5}	5.94
BDE 47	1.40×10^{-6}	15	1.48038×10^{-5}	6.81
BDE 66	9.15×10^{-7}	18	4.93461×10^{-6}	-
BDE 77	5.09×10^{-7}	6	1.18431×10^{-5}	-
BDE 85	7.40×10^{-8}	6	1.08562×10^{-6}	-
BDE 99	1.32×10^{-7}	9	2.26992×10^{-6}	7.32
BDE 100	2.15×10^{-7}	40	6.80977×10^{-7}	7.24
BDE 138	1.19×10^{-8}	-	-	-
BDE 153	1.57×10^{-8}	1	6.61238×10^{-7}	7.90
BDE 154	2.85×10^{-8}	1	2.36862×10^{-6}	7.82
BDE 183	3.51×10^{-9}	2	7.30323×10^{-8}	8.27
BDE 190	2.12×10^{-9}	-	-	-

The lower brominated compounds biomagnify in aquatic systems[124] and, thus cause toxic effects to humans and wildlife[125-127]. Studies in birds have shown that PBDEs cause oxidative stress, suppression of the immune system and its ability to fight infection and, decreased vitamins A and E[128, 129]. In mice, developmental neurotoxicity has been reported as the most critical endpoint for a range of PBDE congeners[100]. For example, neonatal exposure during development in the fetus affected the spontaneous behavior of the offspring and caused impaired learning and memory functions after they reached maturity. Further to this, PBDEs are potential endocrine disrupters because they affect thyroid hormone transport and metabolism[130]. Experiments on rats and mice have shown reduced levels of thyroid hormone in serum after exposure to PBDEs[131]. At high doses, BDE 209 induces tumors in rats and mice[100]. Furthermore, the low PBDEs (BDE 47 and 99) affect the reproduction and developments of harpacticoid copepod (*Nitocra spinipes*)[132].

Some of the PBDE congeners are resistant to degradation[133]. The half-lives of the penta-BDE mixtures in humans have been estimated to be 2-3 years (for BDE-47, 99, 100 and 154) and 4-6 years for BDE-153[134]. Hepta- to deca-BDEs have relatively short half-lives of 16 days in humans[135]. The major sources of PBDEs in the environment include unregulated disposal of textiles, building materials and obsolete electronic waste (e-waste)[136, 137]. In 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 mainly Africa and Asia[138-140]. Commercial PBDE products are mixtures of congeners with different degrees of bromination i.e. penta-, octa- and deca-BDE[141]. Today, the use of all the three mixtures has been banned in both Europe and North America[141].

2.3.2 Occurrence of PBDEs in sediments and fish

Information regarding polychlorinated biphenyls (PCBs) in sediments from Africa is scarce. PBDE concentrations have been reported in sediments from the Congo River Basin in DR Congo (range n.d. to 1900 pg g^{-1} dw)[100] and Juksei River in South Africa (920 to 6760 pg g^{-1} dw)[142]. Several studies have also documented PBDEs in sediments outside Africa: Lake Superior[143], San Francisco Estuary[144], Lake Ontario and Erie in the United States[145], Coast of Qingdao in China[146], Kaohsiung and Tainan coasts in Taiwan[147], river basins in Portugal[148], Scheldt Estuary in the Netherlands[149], Ebro River Basin in Spain [150], Coastal locations of South Korea[151], Tokyo Bay in Japan[152], Niagara River in Canada[153], Danube Delta in Romania[154], South China Sea[155] (Table 2.10). BDE-209 has been reported to be in abundant amounts compared to other congeners in sediments around the world, such as the Swedish rivers (7,100,000 pg g^{-1})[156], Japanese rivers (11, 600,000 pg g^{-1})[157] and, the Pearl River Delta and adjacent South China (7,340,000 pg g^{-1})[155].

Table 2.10: PBDE concentrations in sediments from different studies in the world.

Location	PBDE concentrations (pg g^{-1} dw)	Congeners	References
Africa			
Congo River Basin, DR Congo	n.d. to 1900	7	Verhaert et al.[100]
Juksei River, South Africa	920-6760	16	Olukunle et al.[142]
United states of America			
San Francisco Estuary, USA	n.d. to 212,000	22	Oros et al.[144]
Lake Superior, USA	500-3000	9	Song et al.[143]
Lake Ontario and Erie, USA	137,830-246,850	10	Song et al.[145]
Niagara River, Canada	n.d. to 148,000	9	Samara et al.[153]
Europe			
River and coastal sediments, Portugal	30-18,000	40	Lacorte et al.[148]
Ebro River Basin, Spain	10-10,600	40	Lacorte et al.[150]
Scheldt Estuary, Netherlands	254,000-1,672,000	10	Verslycke et al.[149]
Danube Delta, Romania	n.d. to 100	7	Covaci et al.[154]
Asia			
Kaohsiung and the Tainan coasts, Taiwan	n.d. to 7730	19	Jiang et al.[147]
Coast of Qingdao, China	100-5500	21	Pan et al.[146]
Coastal locations, South Korea	270-493,900	20	Moon et al.[151]
Tokyo Bay, Japan	940-88,600	11	Minh et al.[152]
Yangtze River Delta, China	n.d. to 550	9	Chen et al.[158]
South China Sea	40-94,700	9	Mai et al.[155]

n.d. - non detectable or below limit of detection

Our survey of literature showed limited data on the PBDE levels in fish from Africa. In Ghana, Asante et al.[117] reported PBDE levels of up to 7300 pg g^{-1} lw in fish from Weija Lake, Volta Lake, Benya and Keta Lagoons. In South Africa, Wepener et al.[118] reported concentrations varying from 4100 to 5800 pg g^{-1} lw in fish from Lake Pongolapoort. Several authors have also documented quantifiable PBDE concentrations in fish from other parts of the world, like Asia[159, 160], Europe[161, 162] and USA[163, 164], (Table 2.11). In most of the above studies, the concentrations of BDE 47, 99 and 100, which are also major congeners in the technical penta-mix formulation were higher than those of other congeners. BDE 209, which is usually the predominant congener in sediments was not among the detected congeners in majority of the studies presumably due to its large molecular size that may impede its passage across tissue membranes in biota[165] and/or debromination of BDE 209 to more bioavailable metabolites (penta- to octa-BDEs) in the tissues of biota[166].

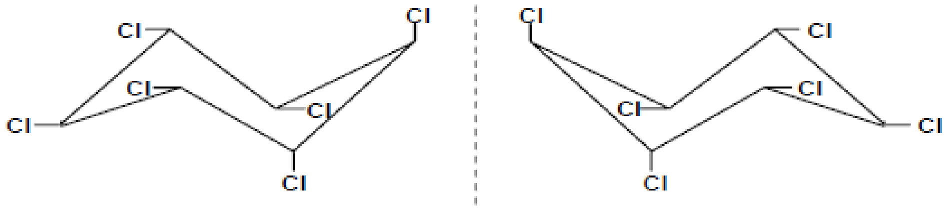
Table 2.11: Studies on the levels of PBDEs in fish from different regions in the world.

Area	Fish species	PBDE levels	References
Weija Lake, Volta Lake, Benya and Keta Lagoons, Ghana	*Oreochromis niloticus*, *Tilapia guineensis*, *Sarotherodon melanotheron*	n.d. to 7300 (pg g^{-1} lw)	Asante et al.[117]
Lake Pongolapoort, South Africa	*Hydrocynus vittatus*	4100-5800 (pg g^{-1} lw)	Wepener et al.[118]
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.[159]
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.[160]
Llobregat River, Spain	*Cyprinus carpio*	29,000-744,000 (pg g^{-1} lw)	Labandeira et al.[161]
Orge River, France	*Rutilus rutilus*, *Morone Americana, Couesius plumbeus*	12,000-18,000 (pg g^{-1} lw)	Teil et al.[162]
Lakes in Western Parks, USA	*Salvelinus* *Fontinalis, Oncorhynchus clarki, Lota lota, Prosopium cylindraceum*	180-5700 (pg g^{-1} ww)	Ackerman et al.[163]
Lake Michigan, USA	*Cyprinus carpio*	<1000-230,000 (pg g^{-1} ww)	Streets et al.[164]

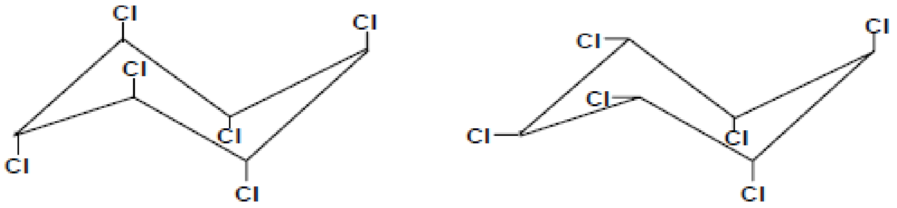
2.4 Hexachlorocyclohexanes (HCHs)

2.4.1 Physico-chemical properties, sources and toxicology of HCHs

Technical HCH was first synthesized in 1825, but its insecticidal properties were not identified until 1942. It is made up of eight isomers, of which α-HCH (60-70%), β-HCH (5-12%) and γ-HCH (10-15%) are the major components. The three major isomers (Fig. 2.4) have different axial-equatorial substitution patterns around the ring, but only α-HCH is chiral[167].



The enantiomers of α-HCH



β-HCH
(achiral)

γ-HCH
(achiral)

Fig 2.4: Core structures of HCH isomers (Adapted from Wilberg et al.[167]).

γ-HCH has the highest vapor pressure and shortest half-life due to its three *axial* chlorines that facilitate degradation[168]. β-HCH has all chlorines in the equatorial positions, therefore it is the most persistent and metabolically inactive isomer[169]. The degradation rates of HCHs increase with increasing number of *axial* Cls in the thermodynamically most stable conformation of an HCH isomer in agreement with a mechanism of anti-periplanar dehydrohalogenation where leaving H and Cl are both *axial* and in antiparallel position ("*trans* HCl elimination")[170].

Technical HCH was widely used in Uganda and other parts of the world as an insecticide on fruits and vegetables in the 1980s[171, 172]. It was also used medically on humans to treatment of lice[173]. The chemical has been banned in most Western countries because it is highly toxic and persistent[174]. HCH isomers are moderately hydrophobic with a log K_{ow} of approximately 4 (Table 2.12). The high volatility of HCHs has led to global transport, even in formerly pristine locations such as the Arctic. Health effects associated with HCHs in mammals include carcinogenicity, anemia and bone marrow changes[169]. γ-HCH stimulates the central nervous system of insects and causes convulsions that are followed by death within 24 hours[175]. Rosa et al.[176] found out that convulsions are mediated by the inhibition of γ-aminobutyric acid neurotransmission or stimulation related to neurotransmitter release. The γ-HCH isomer also induces hepatic nodules and hepatocellular carcinoma in rats[169]. A study on the enantiospecific toxicity of α-HCH reported that the (+) enantiomer was more effective than the (-) form in cytotoxic and growth stimulation bioassays of primary rat hepatocytes[177].

Table 2.12: Properties and parameters of five HCH isomers.

Isomer	α	β	γ	δ	ε
axial (*a*) and equatorial (*e*) position of the chlorine atom	*aaaaee*	*eeeeee*	*aaaeee*	*aeeeee*	*aeeaee*
Melting point (°C) [a]	159-160	309-310	112-113	138-139	
Log K_{ow} [b]	3.9	3.9	3.7	4.1	-
K_{oc} sediment[b]	3800	3800	3800-7000		
Henry's law constant (Pa m^3 mol^{-1})[c]	0.65	0.12	0.27	-	-
Solubility in water (mg L^{-1}, 20 or 25 °C)[d]	1.5	0.2	7.5-8.5	9	-
Vapour pressure (Pa, 20 or 25 °C)[d]	1.6×10^{-2}	4.2×10^{-5}	4.4×10^{-3}	2.1×10^{-3}	-

[a]Smith et al.[175], [b]Willett et al.[169], [c]Jantunen and Bidleman[178], [d]Mackay et al.[26].

2.4.2 Studies on HCH residues in fish and sediments

The various studies carried out have shown contamination by the pesticide. In Uganda, Ejobi et al.[179] reported HCH levels of up to 400 pg g^{-1} lw in *L. niloticus* from Ggaba landing site on the northern shoreline of Lake Victoria. The HCH levels in Uganda were in the same range of data as that documented in the Mekong Delta, South Vietnam[180], but were lower compared to the data from other tropical regions[181, 182]. In Ghana, HCH residues ranging from 700 to 1360 and 200 to 17,650 pg g^{-1} lw were reported in fish from Lake Bosomtwi[183] and Densu River[184], respectively. The results obtained in Ghana were comparable to those in fish from the Congo River Basin in DR Congo[100], Negro River in Argentina[129], but were lower than those reported in fish from Lake Tanganyika in Burundi[112] and, Tana and Sabaki Rivers in Kenya[185] (Table 2.13). In the majority studies, γ -HCH was the most predominant isomer, suggesting usage of lindane in these areas. Furthermore, the HCH concentrations were below the extraneous residue limit of 5,000,000 pg g^{-1} set by FAO/WHO Codex Alimentarius Commission[186].

Table 2.13: Literature survey on the HCH concentrations in fish.

Location	Fish species	Concentrations (pg g^{-1} lw)	References
Murchison Bay, Lake Victoria, Uganda	*Lates niloticus*	n.d. to 400	Ejobi et al.[179]
Congo River Basin, DR Congo	*Marcusenius sp.*, *Schilbe marmoratus*, *Schilbe grenfelli*, *Synodontis alberti*, *Brycinus imberi*, *Distichodus fasciolatus*	n.d. to 66,000	Verhaert et al.[100]
Lake Bosomtwi, Ghana	*Tilapia zilli*	700-1360	Darko et al.[183]
Densu River, Ghana	*Heterotis niloticus, Channa obscura, Hepsetus odoe, Tilapia zilli, Clarias gariepinus, Chrysichthys nigrodigitatus*	200-17,650	Fianko et al.[184]
Lake Tanganyika, Burundi	*Boulengerochromis microlepis, Chrysichthys sianenna, Oreochromis niloticus, Stolothrissa tanganikae, Limnothrissa miodon, Lates angustifrons* and *Lates stappersii*	n.d. to 288,000	Manirakiza et al.[112]
Tana and Sabaki Rivers, Kenya	*Tilapia zilli*	2100-131,250	Lalah et al.[185]
Mekong Delta, South Vietnam	*Parambassis wolffii*, *Parambassis paradiseus, Glossogobius aureus, Parambassis proctozysron, Cynoglossus sp*	n.d. to 200	Ikemoto et al.[180]
Negro River, Argentina	*Cyprinus carpio*	n.d. to 62,020	Ondarza et al.[129]

HCH residues have also been reported in sediments from African water bodies. In Tanzania, Kishimba et al.[187] examined the distribution of HCHs in sediments from areas with known large-scale pesticide use (Southern Lake Victoria and its basin, TPC sugar Plantations in Kilimanjaro region, Dar es Salaam coast, Mahonda-Makoba Basin in Zanzibar) and a former pesticide storage area at Vikuge farm in the coastal region. Total HCH concentrations were up to 132,000 pg g^{-1} dw. Low HCH levels were found in areas associated with agricultural pesticide use and high levels in the former storage areas. In DR Congo, Verhaert et al.[100] collected surface sediments from five different locations of the Congo River Basin (Itimbiri, Aruwimi, Lomami, Isangi and Kisangani) and analyzed them for HCH residues. Concentration of the ∑HCHs ranged from n.d. to 400 pg g^{-1} dw. The most dominant compound was α-HCH (its contribution to the ∑HCHs was 84%). In Ghana, Darko et al.[183] reported lindane (γ-HCH) residues in sediments from Lake Bosomtwi. The residue concentrations were up to 13,940 pg g^{-1}, and were detected in 68% of the samples. Still in Ghana, Ntow[188] reported average lindane concentrations of 2300 pg g^{-1} dw in sediments sampled from the Volta Lake, the largest lake in Ghana. In Egypt, Abbassy et al.[182] determined γ-HCH residues in 18 sediment samples from Lake Manzala. They reported concentrations ranging from 340 to 35,140 pg g^{-1} dw. In a similar study, Barakat et al.[189] reported HCH concentrations (range n.d. to 100,800 pg g^{-1} dw) in sediments from 34 locations in Lake Qarun in Egypt. Amongst the HCH compounds, γ-HCH was the dominant isomer, with a concentration range (90-100,800 pg g^{-1} dw), while α-HCH (n.d. to 1820 pg g^{-1} dw), β-HCH (n.d. to 0.44 pg g^{-1} dw), and δ-HCH (n.d. to 1400 pg g^{-1} dw) were present in small amounts. On average γ-HCH accounted for about 95% of ΣHCHs in sediments. The ratios of α/γ-HCH in the catchment of Lake Qarun ranged from zero to 0.67, indicating past usage of HCHs. Studies outside Africa have also reported HCH concentrations in sediments from Europe[190-193] and Asia[194-201, 202, 203, 204] (Table 2.14).

Table 2.14: Literature survey on the HCH concentrations in sediments from different parts of the world.

Location	Concentration (pg g^{-1} dw)	References
Africa		
Different parts of Tanzania	n.d. to 132,000	Kishimba et al.[187]
Congo River Basin, Dr Congo	n.d. to 400	Verhaert et al. [100]
Lake Bosomtwi	n.d. to 13,940	Darko et al.[183]
Volta Lake, Ghana	Average value, 2300	Ntow[188]
Lake Manzala, Egypt	340-35,140	Abbassy et al.[182]
Lake Qarun, Egypt	n.d. to 100,800	Barakat et al.[189]
Europe		
River Lambro, Northern Italy	500-27,600	Bettinetti et al.[190]
Ebro River, Spain	1-38	Fernández et al.[191]
Remote lakes and coastal areas, Norwegian Arctic	201-7000	Jiao et al.[192]
Guba Pechenga and adjacent areas, Russia	50-680	Savinov et al.[193]
Asia		
Lower Mekong River Basin, Laos and Thailand	5-9900	Sudaryanto et al.[194]
Estuarine and marine sediments, Vietnam	n.d. to 1000	Hong et al.[195]
Hugli Estuary, West Bengal, northeast India	110-400	Guzzella et al.[196]
Bengal Bay, India	170-1560	Rajendran et al.[197]
Danshui River Estuary and coastal areas, Taiwan	260-1640	Hung et al.[198, 199]
Baiyangdian Lake, China	4700-679,000	Hu et al.[200]
Minjiang River Estuary, China	2990-16,210	Zhang et al.[201]
Yangtze Estuary, China	500-17,500	Liu et al.[202, 203]
Pearl River Delta, China	n.d. to 82,300	Fu et al.[204]

The presented data on POPs (i.e. PCDD/F, PCB, PBDE and HCH) in sediments and fish is not all-inclusive; however, studies have reported reduced levels overtime in developed countries than the developing ones[205-207]. The reason for the low levels could be pollution controls (both technical and regulatory) which have been put in place for long periods of time in the developed nations. All the above studies revealed the presence of POP residues in the environment. However, a survey literature shows limited data on POPs in Africa (i.e. particularly the Lake Victoria Basin) compared to the developed world. Therefore, the present study will address this gap.

2.5 Fate, transport and distribution of POPs

2.5.1 Factors that determine the fate of POPs in the environment

Physical-chemical properties such as vapour pressure, water solubility, Henry's law constant, octanol-water partition coefficient (K_{ow}), and the organic carbon-water partition coefficient (K_{oc}) play an important role in the transport of POPs. The properties are also of significance in determining the partitioning of pollutants between air, water, soil, sediment and biota[75, 208]. Temperature strongly affects the vapour pressure, water solubility, Henrys law constant and thus, the partitioning of POPs. Several authors have documented the effect of temperature on the direction and magnitude of air-water gas exchange for PCBs and HCHs in the Great Lakes[209, 210]. They reported that greater volatilization during summer is a result of warming of surface water and, that the net direction of exchange for compounds in the ocean is related to differences in atmospheric concentration and surface water temperature.

Temperature also affects the deposition and distribution of POPs in areas away from the source[211]. The distribution of POPs is inversely proportional to vapour pressure, and thus to temperature. Lower temperatures favour greater partitioning of these substances from the vapour phase to suspended particles in the atmosphere and this increases the likelihood of their removal and transport to the earth's surface in the cold, while in rainy areas wet deposition will deposit the pollutants on land or water by particle sedimentation or rain-out[212]. The rate of release from the surface will be slower in colder polar or high altitude regions. Hence, it has been proposed that on a global scale, POPs can potentially migrate from warmer to colder areas and become 'fractionated' on latitudinal or altitudinal gradients[213].

2.5.2 Influence of the environment on persistence and transport of POPs

Chemical properties like low vapor pressure and water solubility, and high fat solubility are hallmarks in the environmental persistence of POPs[214]. Possible biodegradation has been suggested for some POPs, such as lindane[215]. The degradation of PCDD/Fs, PCBs and PBDEs in the natural environment is slow[216] and is inversely correlated to carbon chain length and chlorine content, and log K_{ow}[217].

The degradation pathways for POPs include hydrolysis, photolysis, adsorption onto particulate matter and volatilization into the atmosphere. Photolytic degradation of POPs on particulate surfaces is highly variable and is dependent on the surface type and, the wavelength and intensity of light. In a laboratory experiment, large differences were found between the rates of photolysis of PCDDs (half-life = 88,000 hours) and PCDFs (half life = 3-14 hours) adsorbed on silica gel and fly ash[218]. The degradation rates during hydrolysis are influenced by factors such as temperature, pH, ionic strength and, the presence of metal ion catalyses. However, if the degradation rate is catalyzed by acid or base then pH will have a strong effect on half-life of the compound[219].

2.5.3 Transport of POPs in the tropics and temperate

POPs have been found at low levels in sediments from tropical regions; however, the low levels are not necessarily an indication of low exposure to or usage of POPs in the tropics[220, 221]. Volatilization dominates the environmental distribution and partitioning of semi-volatile POPs in the tropics[222, 223]. In South India, the flux of HCH residues into the atmosphere of the Vellar River and its watershed was approximated to 99.6% of the HCH applied in the rice growing areas of this watershed. Only 0.4% was transported by welter to the estuary over the year and about 75% of the water-borne flux to the estuary was estimated to be lost to the air by volatilization. Thus, only about 0.1% of the applied HCH was estimated to ultimately be drained to the sea via the water into the Vellar River[186].

In a global monitoring survey of air and surface seawater, HCH concentrations were greater in the Northern hemisphere and in the cold welter deposition areas than in the tropical regions, while PCBs showed a more uniform global distribution[224]. The results suggest, that POPs having intermediate volatility such as HCHs will rapidly be transported and deposited, while the less volatile ones like PCBs migrate slowly. The volatilization of POPs combined with their low aqueous solubility and elevated year-round ambient temperatures leads to higher atmospheric concentrations and lower aquatic ecosystem concentrations in tropical regions relative to temperate regions[222]. In addition to higher potential for volatilization and subsequent atmospheric dispersal, POPs may be subject to faster rates of degradation in tropical regions due to increased metabolic activity in biota, further reducing their potential levels[225].

2.6 Exposure of humans and wildlife to POPs

Living organisms can be exposed to POPs and other hazardous substances mainly via intake of contaminated food[226], particularly fatty fish, meat and dairy products[227]. The body burdens caused by POPs are dependent on human dietary habits, i.e. whether they consuming a variable diet or from the top of the food chain. The later is the case for fishermen and their families[228]. Human beings have adipose tissues that accumulate and retain lipophilic contaminants[229]. Therefore, the contamination of food, including breast milk and fish by POPs is of worldwide concern[230]. Other important routes of exposure are dermal contact and inhalation[231]. These normally occur for populations living in contaminated areas and/or with a record of production of the chemical(s)[229].

Acute exposure to pesticides and dioxins can occur during herbicide production, industrial accidents, chemical fires or through the burning of waste[232]. Furthermore, exposure to chlorinated pesticides can occur via poor handling or application processes[233]. Accidental and occupational high level exposure to POPs is of concern for both acute and chronic worker exposure. The risk of exposure is high in developing countries where the use of POPs in tropical agriculture, livestock and vector control has resulted in a number of injuries and deaths[234]. In many countries worker exposure to POPs during waste management, in addition to other exposure routes is a significant source of occupational risk[235].

Organochlorine pesticides and PCBs have shown to elicit a wide range of toxic and biochemical effects in both laboratory animals and wildlife. Adverse health effects including illness and death, reproductive failures, tumor induction, endocrine disruption and cancers can occur once humans and wildlife are exposed to POPs[233]. They pose a serious health risk, especially for infants, since their enzymatic and metabolic systems are not fully active[236]. With regard to maternal exposure to pesticides in early pregnancy, several epidemiological studies suggest that maternal employment in agriculture may be a risk factor for birth defects[237, 238]. The International Agency for Research on Cancer classified PCBs as probable carcinogens to living organisms and are thought to be among the leading causes of human breast cancer [239, 240].

3. MATERIALS AND METHODS

3.1 Study areas

Three contaminated embayments, namely the Murchison Bay, Napoleon Gulf and Thurston Bay, all located on the northern watershed of Lake Victoria were selected for this study.

3.1.1 Murchison Bay

Murchison Bay is located in the northwestern part of Lake Victoria and is southeast of Kampala city (Fig. 3.1). The Bay covers an area of 60 km^2, and includes semi-enclosed inner and outer parts. Murchison Bay is the main recipient of sewage and effluents from the central district of Kampala, via the Nakivubo channel. The channel passes through dense residential settlements and commercial areas. In the past, the channel ended in wetland areas, allowing wastewater to be drained into papyrus swamps before entering the Bay. However, in recent years the wetlands which played the role of secondary purification have been degraded for settlement and commercial activities.

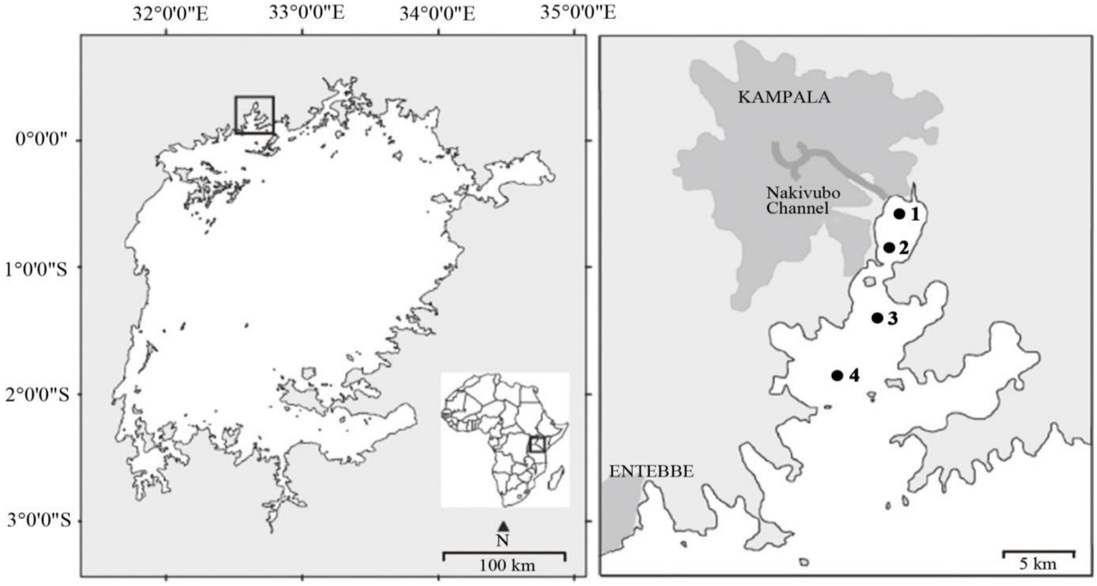


Fig 3.1: Map of the Murchison Bay showing the study stations. Adapted from Haande et al.[241]

3.1.2 Napoleon Gulf

Napoleon Gulf is located south east of Jinja Municipality where River Nile (the only outlet of L. Victoria) begins its journey to the Mediterranean Sea (Fig. 3.2). The Gulf covers an area of about 200 km^2 and supports approximately half a million inhabitants. However, in recent years it has been subjected to strong anthropogenic pressures. The major pollution sources being untreated wastewater from sewage systems, industrial plants, waste oil from parking lots and car repair garages. The major industries in the vicinity of the Gulf include a water treatment plant and copper smelting plant.

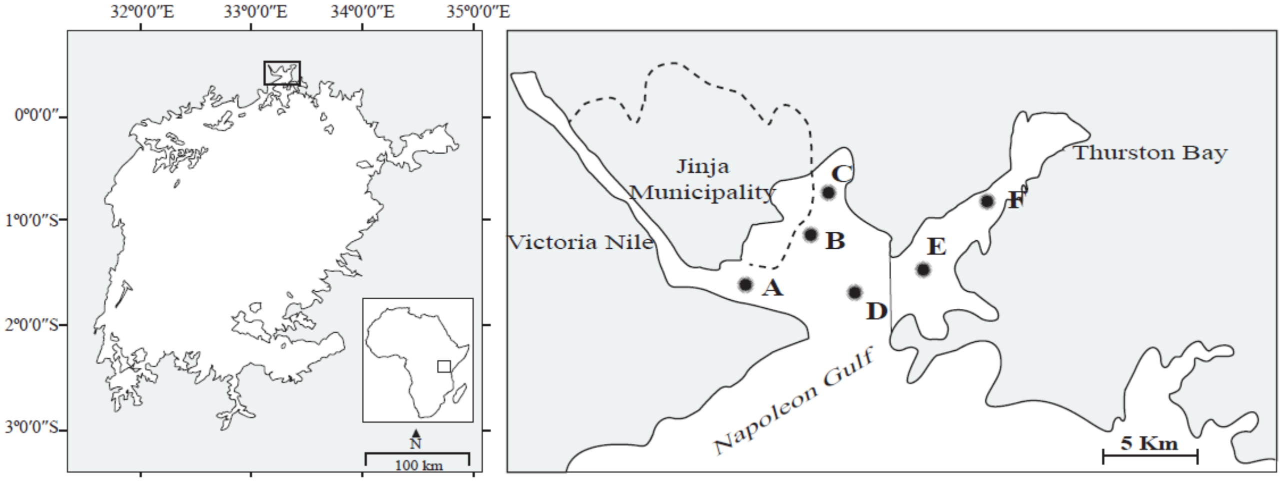


Fig 3.2: Map of the Napoleon Gulf showing the sampling points. The shaded section is the Thurston Bay.

3.1.3 Thurston Bay

Thurston Bay is located far from Jinja (Fig. 3.2). A great deal of bio-mass burning occurs along the banks of the Thurston Bay where sugarcane growing is also significant. The shorelines of the Napoleon Gulf and Thurston Bay which were previously fringed by papyrus (*Cyprus papyrus*), have been extensively degraded by a rapid rising human population, mounting pressure on the lake, in terms of water quality and sedimentation[242].

3.2 Sampling

A total of 180 surface sediments (<25 cm depth) were taken using a sediment corer between March 2011 and May 2013. Two hundred fish of different species namely: *L. niloticus* a top-predator and *O. niloticus* a detritivorous species[243-245] were collected using gill nets (mesh sizes 25.4 to 101.6 mm) within the same locality as the sediments. The two fish species were chosen because they are readily available and highly consumed within the Lake Victoria Basin[246]. Fish specimens were measured for total length and weight to the nearest 0.1 cm and 0.01 g, respectively. Muscle tissues were dissected between the pectoral fin and vent of the fish, minced into pieces and subsamples taken. Subsamples of 4 fish from the same location and, of similar length and species were pooled and homogenized to minimize the cost that would be spent on a large number of samples. The sediments and homogenized fish were transferred into acetone rinsed glass bottles and were freeze-dried.

3.3 Analytical approaches

The various analytical procedures, from sample extraction to instrumental analysis for sediments and fish are described below. Sediment samples were divided into two subsamples for POP analysis and total organic carbon (TOC) determination. The sediment TOC was measured using a LECO C230 carbon analyzer after removal of carbonates with hydrochloric acid. Standard procedures were followed for this, with a combustion temperature of 1020 ^{o}C, using acetanilide as a standardizing agent. Prior to extraction, the samples (sediments and fish) were allowed to thaw and a known mass was spiked with ^{13}C-labelled PCDD/F, PCB and PBDE surrogate standards. The spiked samples were then mixed with hydromatrix™ (Varian) to remove any moisture. Extraction was carried out using an accelerated solvent extractor device (Dionex 200, Sunnyvatem CA, USA) at a temperature of 120 ^{o}C and pressure of 120 bar, using a 3:1 v/v of *n*-

hexane/acetone as extraction solvent. The resulting organic extract was passed through a drying funnel of anhydrous sodium sulfate and eluted with 100 mL of *n*-hexane. The extraction volume was concentrated to 2 mL on a rotary evaporator and kept for purification to remove interferences by two sequential liquid chromatography steps. The first purification step was performed by adsorption chromatography using a multilayered sandwich glass column (packed successively from bottom to top with 2 g silica gel, 5 g of 33% silica gel-sodium hydroxide, 2 g silica gel, 5 g of 44% silica gel-sulfuric acid, 10 g of 22% silica gel-sulfuric acid and 5 g sodium sulfate). The silica gel was heated overnight at 200 °C before use, to reduce the background levels of analytes. The column was washed with 60 mL *n*-hexane before it was connected to a reversible carbon column (Carboxen 1016, Supelco) that had been rinsed by an equivalent volume (25 mL) of toluene and *n*-hexane, respectively. The concentrated extract was then applied to the column and eluted with 100 mL of *n*-hexane. PCDD/Fs and non-*ortho* PCBs were retained in the carbon column, while the PBDEs and mono-*ortho* PCBs passed through both columns. The sandwich column was removed and the carbon column was further eluted with 30 mL of 10% dichloromethane (DCM) in *n*-hexane, and the resultant solution concentrated. The carbon column was reversed, eluted with 100 mL toluene and the resultant solution was concentrated to <2 mL. The second purification step was achieved by using a chromatographic column filled with 5 g of alumina B super 1 as described in Çok et al. (2009). The purified fractions were transferred to sample vials followed by addition of internal standards containing ^{13}C-labelled PCDD/Fs and PCBs prior to instrumental analysis. The analysis of 18 PCB congeners, 17 PCDD/Fs and 12 PBDEs (Table 3.1), 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).

Table 3.1: Persistent organic pollutants in this study.

POPs	Congeners and HCH-isomers
PCDDs	2,3,7,8-TCDD, 1,2,,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD and OCDD
PCDFs	2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF and OCDF
PCBs	CB 28, 52, 101, 138, 153, 180, 77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 167 and 189
PBDEs	BDE 17, 28, 47, 66, 85, 99, 100, 138, 153, 154, 183 and 206
HCHs	α-HCH, β-HCH, γ-HCH, δ-HCH and ε -HCH

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 and PCBs was achieved on a fused-silica capillary column (DB5-MS 60 m × 0.25 mm i.d. × 0.25 µm film thickness). For PCDD/Fs, 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}. For PCBs, the oven temperature of the GC was held at 120 °C for 1min, then increased to 150 °C at a rate of 30 °C min^{-1} and finally to 300 °C at 2.5 °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.

For HCHs, the samples were extracted and analyzed as described by Javedankherad et al. (2013). Briefly, the samples were freeze-dried and 5 g was spiked with HCH internal standards. The spiked sample was transferred into a thimble and extracted in a soxhlet apparatus for 4 h with 100 mL of 3:1 v/v *n*-hexane/acetone. The resultant extract was concentrated to 2 mL using a rotary evaporator and divided into subsamples for fish (0.3 and 1.7 mL). The 0.3 mL was used for lipid determination, while the 1.7 mL was cleaned up using a column filled from bottom to top with 8 g of acidified silica gel and 2 g of anhydrous sodium sulfate. The column was eluted with 15 mL *n*-hexane followed by 10 mL of DCM. The resulting eluate was concentrated under a gentle stream of nitrogen to 100 μL and kept for analysis. The analysis was performed using a GC equipped with an electron capture detector (ECD) and a DB-5 capillary column (60 m × 0.25 mm i.d. × 0.25 μm). Helium was used as the carrier gas at a flow rate of 2 mL min^{-1}. The operating conditions were split injection mode. The temperature program was as follows: 100 °C (1 min), 10 °C min^{-1} to 240 °C (1 min), 3 °C min^{-1} to 260 °C (1 min), 20 °C min^{-1} to 300 °C (10 min). The injection port temperature and detector temperature were 250 and 300 °C, respectively.

3.4 Quality assurance and quality control

To ensure quality 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. Recovery assays were replicated three times. Recoveries for surrogate PCDD/Fs were 46 to 104% in the sediments and 50 to 100% in the fish (articles I, II and V). Those for PBDEs varied from 54 to 99% and 56 to 109%, respectively (paper V). The recoveries were within the range acceptable for EPA methods 1613B and 1614. The recoveries for PCB surrogate standards ranged between 45% and 108% (papers 1, 11, 111 and 1V). Majority of the PCB recoveries met the requirements of US EPA methods 1668A (were in the range of 75-127%) except for CBs 28, 118, and 138 whose recoveries were <75% (range: 56-74%). The recoveries resulting from triplicate determinations (N = 3) of the PCB certified reference material were between 82% and 106%. The recoveries for HCH isomers (α, β and γ) in fish varied from 50.1 to 84.2%, 70.3 to 90.4% and 60.3 to 80.1%, respectively (paper III).

Limit of detection (LOD) was calculated as three times the signal-to-noise ratio (S/N). The LODs 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. The LOD for PCBs in the sediments varied from 0.04 to 0.96 pg g^{-1} dw and 0.02 to 0.80 pg g^{-1} ww in the fish. HCHs had their LODs between 1 and 4 pg g^{-1} ww in fish. For samples where the concentration of a congener was less than the LOD, the levels were reported as not detected. Relative standard deviations of the method (n = 3) were in the range of 1-9%.

3.5 Statistical analysis of data

Statistical analysis was performed using SPSS 17.0 software (SPSS Inc., Chicago, IL). The Kolmogorov-Smirnov test was used to verify the normality of data. For concentration values that were not normally distributed, non-parametric statistics was applied. The Mann-Whitney U test was used for comparing two data sets and the Kruskall-Wallis test when more than two groups were considered. In all cases, the level of significance was set at $p < 0.05$. One-way analysis of variance (ANOVA) followed by Tukey's HSD post hoc tests, were conducted for multiple comparison to test for significant differences between POP profiles of different media.

3.6 Determination of the Toxic equivalents (TEQs)

TEQs for 17 PCDD/Fs and 12 dioxin-like (dl) PCBs were calculated by multiplying the concentration of each congener by its WHO-TEF and then summing all the values [37]. The TEF concept is based on the assumption that PCDD/Fs and dl-PCBs all exert their toxicity via a common mechanism, which involves binding to the aryl hydrocarbon (Ah) receptor.

3.7 Calculation of Biota-Sediment Accumulation Factors (BSAFs)

BSAFs are used to estimate the degree to which chemicals accumulate in biota relative to sediments. BSAFs were calculated based on the formula BSAF = $C'_{avg}/Csoc_{avg}$ (Burkhard, 2009), where C'_{avg} is lipid normalized concentration in the tissue and $Csoc_{avg}$ is the sediment organic carbon normalized concentration. The BSAF model assumes that (1) the exposure time was long enough to approach equilibrium between uptake and elimination, (2) both the organism and its food are exposed to sediments, (3) the sediment used for measuring/estimating the sediment concentration is the same sediment to which the organism is exposed and (4) there is no transformation in either the sediments, water, or organism[247]. The model is suggested as a useful first-level screening tool for predicting bioaccumulation and is used by regulatory agencies to evaluate the risk of organic contaminants in the aquatic environment[248].

3.8 Principal Component Analysis (PCA)

PCA was used to identify the possible sources of PCDD/Fs. Principal components were considered when their Eigen values were >1. After identifying the correlations, 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[147].

4. RESULTS AND DISCUSSION

4.1 Concentrations and sources of PCDD/Fs in the sediments

Total PCDD/F concentrations (Σ_{17} PCDD/Fs) varied widely from one location to another with mean values ranging from 3.19 to 478 pg g^{-1} dry weight (dw) (Table 4.1). The PCDD/F concentrations in the sediments from station 1 located in the inner Murchison Bay were statistically greater (Kruskall-Wallis test, $p < 0.05$) than those from the Napoleon Gulf and Thurston Bay sites. Station 1 is near the mouth of Nakivubo channel which traverses through urban and industrial areas of Kampala city (i.e. the most industrialized city in Uganda[249]), bringing in waste-water effluents into the Murchison Bay.

Statistical analysis showed that the PCDD/F concentrations in sediments from station 2 were also significantly higher compared to the data from the Napoleon Gulf and Thurston Bay. Station 2 is located near Ggaba where local conditions are influenced by industrial activities. The PCDD/F levels at stations 3 and 4 were comparable, but statistically insignificant compared to other locations in the Napoleon Gulf and Thurston Bay. The general discussion of PCDD/F concentrations in sediments from the study sites and comparison with literature values are reported in papers 1 and V. The ΣPCDDs varied from 3.19 to 114 pg g^{-1} dw while the ΣPCDFs ranged between n.d. to 364 pg g^{-1} dw. Statistical analysis between the ΣPCDDs and ΣPCDFs showed no significant differences (Mann-Whitney U test, $p > 0.05$).

In this study, octachlorodibenzodioxin (OCDD) was the major contributor to the ΣPCDDs (it contributed >29.7% at all locations). The reason for the domination of OCDD could be that it is emitted at higher rates into the environment than all other congeners. OCDD is usually produced in high amounts as a by-product during combustion of domestic and fuel oil mixtures[250], and open fire of garden waste[251].

Table 4.1: PCDD/F concentrations (pg g^{-1} dw) in the sediments.

	Murchison Bay				Napoleon Gulf				Thurston Bay	
	ST 1	ST 2	ST 3	ST 4	ST A	ST B	ST C	ST D	ST E	ST F
% TOC	5.23 (2.32)	4.03 (1.86)	3.35 (2.01)	2.33 (1.19)	2.37 (1.08)	1.53 (0.51)	1.70 (0.20)	0.93 (0.40)	1.30 (0.10)	1.40 (0.26)
Texture	Clay	Sandy	Clay/ sandy	Clay/ plastic	Sandy	Clay	Sandy	Loamy	Loamy	Sandy
ΣPCDFs	364 (112)	242 (54.0)	11.8 (6.77)	22.8 (5.28)	45.9 (26.8)	6.06 (3.90)	11.3 (3.11)	16.4 (7.80)	n.a	5.21 (2.00)
ΣPCDDs	114 (83.8)	73.4 (7.69)	99.8 (26.1)	43.7 (27.9)	5.11 (1.99)	34.2 (18.2)	36.2 (4.23)	35.1 (15.8)	3.19 (0.34)	41.4 (26.4)
ΣPCDD/Fs	478 (196)	315 (61.7)	112 (32.9)	66.5 (33.2)	51.0 (28.8)	40.3 (22.1)	47.5 (7.34)	51.5 (23.6)	3.19 (0.34)	46.6 (28.4)
ΣPCDD/Fs ×% TOC	2500	1269	375	155	121	61.7	80.8	47.9	4.15	65.2
ΣWHO-TEQs	0.14 (0.11)	0.17 (0.07)	0.33 (0.17)	0.08 (0.06)	0.98 (0.71)	1.32 (1.26)	0.71 (0.02)	4.24 (2.23)	0.08 (0.01)	0.42 (0.31)

Results are presented as mean values (standard deviation); ST – station; n.a. – not available; number of samples per station (N) = 6.

Of the PCDFs, octachlorodibenzofuran (OCDF) was the dominant congener (based on mean values it contributed >22% to the ΣPCDFs at all stations except the Thurston Bay). Martínez et al.[252] pointed out that significant concentrations of OCDF in environmental samples is mostly indicative of ferrous- and non-ferrous metal operations and sintering plants as potential sources. 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[107, 253]. The result from PCA indicate 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. 4.1). In the ordination space generated by 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. It is note-worthy that stations 1, 2 and A which were enriched with OCDF and PeCDF, indicate similar origins. In addition, the grouping of stations 3, B, C and E with PeCDD and HxCDD also suggests a common source.

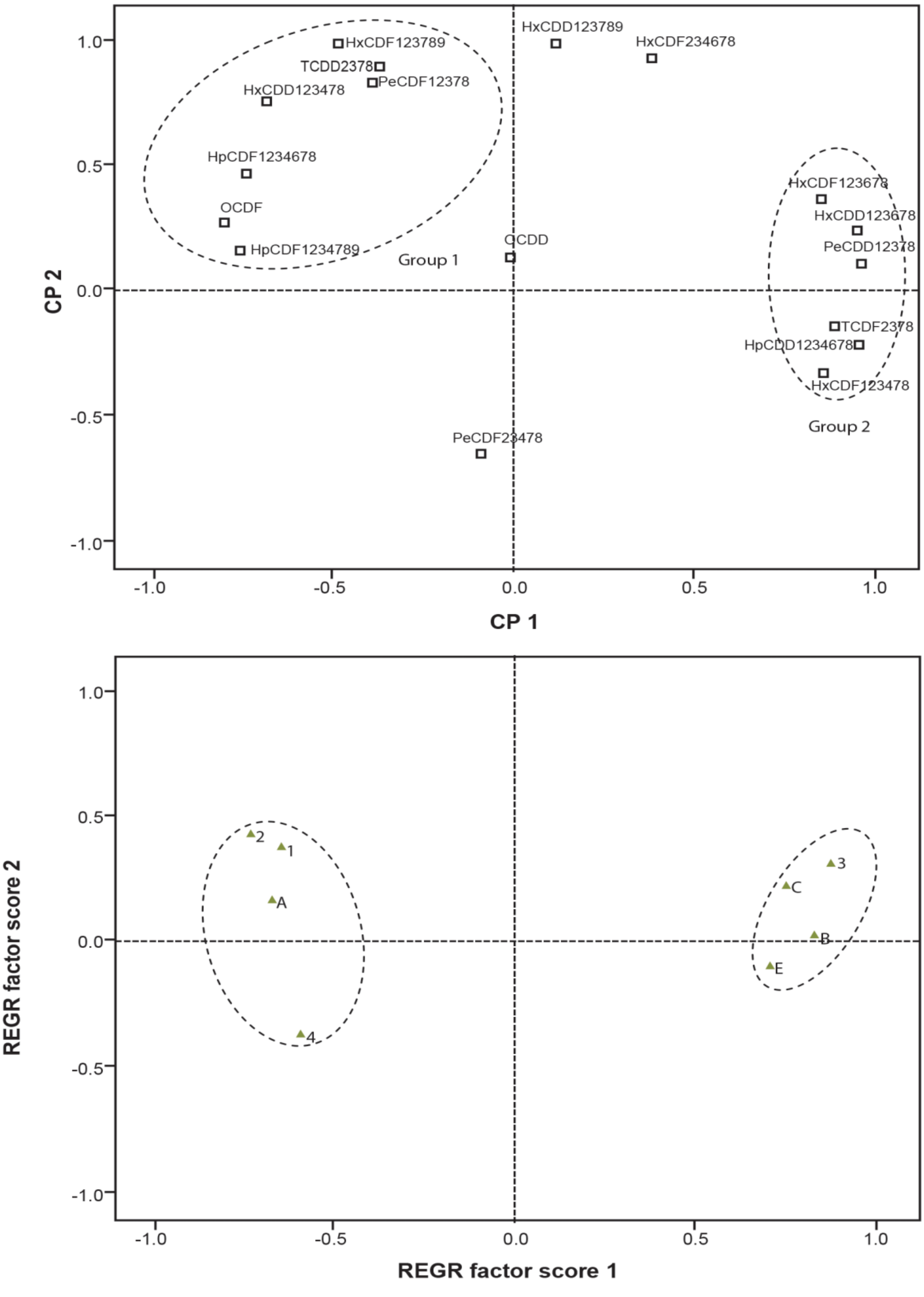


Fig 4.1: Loading (a) and score (b) plots of principal component analysis of 17 toxic PCDD/Fs emissions. The circles represent similar cluster conditions and scores.

4.2 Toxic equivalents for PCDD/Fs in the sediments

The World Health Organization-toxic equivalents (WHO_{2005}-TEQs) for PCDD/Fs in sediments ranged from 0.08 to 4.24 pg TEQ g^{-1} dw. PCDFs accounted to a larger extent to the ΣTEQs than PCDDs (>70%) at all sites. Canadian Council for Ministers of the Environment set a maximum value of 0.85 pg WHO-TEQ g^{-1} dw as an interim sediment quality guideline[254]. 23.1% of the samples from the Napoleon Gulf were above the set value. The current PCDD/F levels do not pose any toxic related problems to fish and birds associated within Murchison Bay and Thurston Bay of Lake Victoria.

4.3 Levels of PCDD/Fs in fish muscle homogenates

The PCDD/F results on a wet-weight (ww) basis for fish muscle homogenates are presented in Table 4.2. The ΣPCDD/Fs varied largely between fish species, with mean values ranging from 0.12 to 49.0 pg g^{-1}. Comparison of the levels with literature values is reported in papers 11 and V. The ΣPCDD/F concentrations in *L. niloticus* on a wet weight basis were statistically greater (Mann-Whitney U test, $p < 0.05$) than those in *O. niloticus*. The difference in concentrations could be attributed to the fact that former fish species feeds at a higher trophic level than the later. Campbell et al.[255] previously used stable nitrogen and carbon isotope ratios ($\delta^{15}N$ and $\delta^{13}C$, respectively), and found that *L. niloticus* was the top trophic predator (n = 9, $\delta^{15}N = 8.9 \pm 1.3$ ‰, $\delta^{13}C = -19.0 \pm 0.9$ ‰) while *O. niloticus* was littoral and feeding at lower trophic levels (n = 8, $\delta^{15}N = 5.7 \pm 0.6$‰, $\delta^{13}C = -19.5 \pm 1.0$‰). Other possible reasons for the observed biomagnification could be the efficiency of chemical transfer from prey to predator upon ingestion, and the degree of elimination via excretion and biotransformation. Biomagnification of POPs in aquatic biota depends on the route of exposure, in particular on the composition and contamination of the underlying food web[256]. This is because the predominant route of exposure for fish to bioaccumulate POPs is via the diet and/or ingestion of contaminated prey.

Table 4.2: PCDD/F concentrations (pg g^{-1} ww) in fish from Lake Victoria.

	Murchison Bay				NG	TB
	ST 1	ST 2	ST 3	ST 4		
L. niloticus						
% lipid	2.8 (0.4)	2.3 (0.5)	2.2 (0.6)	1.6 (0.4)	1.9 (0.3)	1.5 (0.6)
ΣPCDFs	26.1 (17.4)	0.45 (0.15)	2.50 (1.44)	2.96 (0.64)	0.84 (0.27)	0.47 (0.23)
ΣPCDDs	22.9 (20.9)	22.1 (3.46)	9.52 (5.15)	4.29 (0.57)	0.10 (0.07)	0.15 (0.08)
ΣPCDD/Fs	49.0 (38.3)	22.6 (3.61)	12.0 (6.59)	7.25 (1.21)	0.94 (0.34)	0.62 (0.31)
ΣPCDD/Fs/ % lipid	17.5	9.83	5.45	4.53	0.49	0.41
BSAF values	0.007	0.008	0.014	0.029	0.006	0.012
ΣWHO$_{2005}$-TEQs	0.015 (0.004)	0.009 (0.001)	0.004 (0.001)	0.002 (0.001)	0.09 (0.07)	0.03 (0.02)
O. niloticus						
% lipid	2.2 (0.4)	1.9 (0.6)	1.5 (0.4)	1.2 (0.3)	1.1 (0.4)	0.8 (0.2)
ΣPCDFs	1.33 (0.19)	9.77 (5.32)	2.58 (0.19)	1.72 (0.16)	0.33 (0.16)	0.12 (0.08)
ΣPCDDs	17.0 (1.86)	7.24 (3.74)	3.59 (1.61)	3.60 (0.76)	n.d.	n.d.
ΣPCDD/Fs	18.3 (2.05)	17.0 (9.06)	6.17 (1.80)	5.32 (0.92)	0.33 (0.16)	0.12 (0.08)
ΣPCDD/Fs/ % lipid	8.32	8.95	4.11	4.43	0.30	0.15
BSAF values	0.003	0.007	0.011	0.029	0.004	0.004
ΣWHO$_{2005}$-TEQs	0.011 (0.007)	0.016 (0.010)	0.002 (0.001)	0.001 (0.001)	0.01 (0.01)	0.02 (0.01)

Results are presented as mean values (standard deviation); ST – station; n.d. – non detectable; number of fish muscle homogenates (N) = 3.

In the present study, the ΣPCDDs in *L. niloticus* varied from 0.10 to 22.9 pg g-1 ww while the ΣPCDFs were between 0.45 and 26.1 pg g-1 ww. The concentrations of the later and former pollutants in *O. niloticus* were 0.12-9.77 and n.d. to 17.0 pg g-1 ww, respectively. Statistical analyses between the ΣPCDDs and ΣPCDFs in *L. niloticus* showed no significant difference (Mann-Whitney U test, $p > 0.05$). Similarly, the ΣPCDDs in *O. niloticus* were not statistically correlated to the ΣPCDFs. Of the PCDD congeners, only OCDD and 1,2,3,4,6,7,8-HpCDD were detected in the fish. The high prevalence of OCDD as well as HpCDD in both species could be indicative of a pentachlorophenol (PCP) source[257]. However, our survey of literature showed no reports about the presence of PCPs in the study area. 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 other congeners (it contributed >38% to the ΣTEQs at all locations in both fish species).

4.4 Concentrations and sources of PCBs in the sediments

The analyzed categories were the indicator (IUPAC numbers 28, 52, 101, 138, 153 and 180), non-*ortho* (CBs 77, 81, 126 and 169) and mono-*ortho* PCBs (CBs 105, 114, 118, 123, 156, 157, 167 and 189). Total PCB concentrations (Σ_{18}PCBs) in the three embayments ranged from 313 to 4325 pg g^{-1} dw (Table 4.3). Comparison of the present data with literature values is reported in papers 1,111 and 1V. In this study, a significant difference (Kruskall-Wallis test, $p < 0.05$) was observed between the Σindicator PCBs, and the Σnon-*ortho* PCBs and Σmono-*ortho* PCBs. However, a statistical test between the Σnon-*ortho* PCBs and Σmono-*ortho* PCBs showed no significant difference (Mann-Whitney *U* test, $p > 0.05$).

Table 4.3: PCB concentrations (pg g^{-1}dw) in the sediments and % TOC.

	ST 1	ST 2	ST 3	ST 4	ST A	ST B	ST C	ST D	ST E	ST F
% TOC	5.23 (2.32)	4.03 (1.86)	3.35 (2.01)	2.33 (1.19)	2.37 (1.08)	1.53 (0.51)	1.70 (0.20)	0.93 (0.40)	1.30 (0.10)	1.40 (0.26)
Σindicator PCBs	3516 (1106)	2209 (1151)	945 (379)	651 (283)	640 (113)	848 (471)	393 (97)	362 (65)	196 (9)	292 (14)
Σnon-*ortho* PCBs	88 (23)	61 (8)	14 (5)	11 (2)	32 (18)	17 (10)	n.a.	n.a.	31 (5)	21 (7)
Σmono-*ortho* PCBs	721 (350)	305 (95)	173 (83)	115 (69)	266 (128)	173 (104)	n.a.	n.a.	149 (63)	n.a.
Σ_{18}PCBs	4325 (1479)	2575 (1254)	1132 (467)	777 (354)	937 (259)	1038 (585)	393 (97)	362 (65)	385 (77)	313 (21)
Σ_{18}PCBs x % TOC	22620	10377	3792	1810	2221	1588	668	338	501	438
WHO_{2005}-TEQs for the dl-PCBs	0.55 (0.17)	0.25 (0.07)	0.11 (0.06)	0.10 (0.04)	0.17 (0.03)	0.09 (0.08)	n.a.	n.a.	0.03 (0.02)	0.21 (0.04)

Results are presented as mean values (standard deviation); ST - station; n.a. – not available

The proportion of the 6 indicator PCBs (Σ_6PCBs) to Σ_{18}PCBs in this study were between 51 and 100% (with a mean value of 83%). CBs 138 and 153 were the predominant congeners at majority of the sites, collectively accounting for >23% to the Σ_{18}PCBs on average. Barakat et al.[189] also reported the predominance of CBs 138 and 153 in surface sediments from Lake Maryut in Egypt. Boon et al. [258] reported that the rates of metabolism of PCBs depends mainly on structure and the degree of chlorination of the molecule. PCBs with vicinal non-substituted carbons are readily eliminated via metabolism while the highly chlorinated congeners (e.g. CBs 138, 153 and 180) with only isolated non-chlorinated carbons show the longest half-lives, and therefore the greatest accumulation[258-261]. The different Cl-substitution patterns in the *ortho-meta* and *meta-para* positions, which can influence persistency of the congeners, are shown in

Table 4.4. The congeners in Group I are persistent due to lack of vicinal hydrogen atoms (H), and groups II and III have vicinal H-atoms only in *ortho-meta* positions. Group II congeners may be metabolized through inhibition due to steric hindrance by di-*ortho* Cl-substitution, whereas group III may be metabolized by CYP1A mediated enzymes due to non- or mono-*ortho* Cl-substitution (*om*-congeners). Metabolism of Group IV is due to vicinal H-atoms in *meta-para* positions (*mp*-congeners) with two or less *ortho* Cl-substitutions.

Table 4.4: Summary of the different substitution pattern of the PCB congeners [262].

PCBs	Positions of the vicinal H-atoms
NO-PCBs	CB 77, 81, 126 and 169. Non-*ortho* Cl-substituted PCBs.
MO-PCBs	CB 28, 105, 118, 156, 157 and 189. Mono-*ortho* Cl-substituted PCBs.
DI-PCBs	CB 47, 52, 99, 101, 110, 128, 137, 138, 141, 153, 170 and 180. Di-*ortho* Cl-substituted PCBs.
Group 1	CB 153, 169, 180 and 189 (no vicinal H-atoms).
Group 11	CB 138 (vicinal H-atoms only in *ortho–meta* positions, ≥2 Cl in *ortho*-position).
Group 111	CB 28, 77, 81, 105, 126, 118, 156 and 157 (vicinal H-atoms only in *ortho-meta* positions, <2 Cl in *ortho*-position).
Group 1V	CB 52 and 101 (vicinal H-atoms in *meta-para* positions, ≤2 Cl in *ortho*-position).

The PCB levels at station 1 were significantly higher (Kruskal-Wallis test, $p < 0.05$) than those at other sampling locations except station 2. High concentrations of PCBs were also observed at station 2 (mean value of the Σ_{18}PCBs in sediments was 2576, range 1753-3970 pg g^{-1} dw). Station 2 is situated close to Ggaba landing site and a local market. Besides the market a lot of commercial, domestic and industrial activities occur near the station and could be releasing domestically-produced commercial PCB mixtures into the lake. The PCB levels in sediments from the outer part of the Murchison Bay (stations 3 and 4) and Thurston Bay (sites E and F), all located far from point sources, were an order of magnitude lower compared to those in the inner Bay. The low PCB levels in the outer Murchison Bay and Thurston Bay are likely due to the far offshore distance from the pollution sources. It was observed that even though sites A and B are in the close vicinity of Jinja Municipality, their concentrations were in the same range as those of stations 3 and 4. Statistical analysis showed a significant difference between station B located in the Napoleon Gulf and sites of the Thurston Bay.

4.5 Concentrations of PCBs in the fish

Average concentrations of the Σ_{18}PCBs ranged from 48 to 779 pg g^{-1} ww (Table 4.5). Comparison between the PCB concentrations with other studies is reported in papers 11-1V. The Σ_{18}PCBs in *L. niloticus* at station 1 was one-fold higher than that at station 2 and about three times higher than that at station 4. The Σ_{18}PCBs at stations 3, 4, Napoleon Gulf and Thurston Bay were quite similar. For *O. niloticus*, still station 1 showed the highest levels of the Σ_{18}PCBs (mean value 229 pg g^{-1}) compared to other stations. The concentrations at other stations (2, 3, 4, Napoleon Gulf and Thurston bay) were in the same range.

Table 4.5: Concentrations of PCBs (pg g^{-1} ww) in fish.

Stations	ST 1	ST 2	ST 3	ST 4	NG	TB
L. niloticus						
% lipid content	2.8 (0.4)	2.3 (0.5)	2.2 (0.6)	1.6 (0.4)	1.9 (0.3)	1.5 (0.6)
Σ indictor PCBs	636 (368)	397 (208)	242 (121)	203 (117)	223 (24)	213 (16)
Σ non-*ortho* PCBs	7 (2)	0.2*	3 (2)	3 (1)	2 (0.34)	0.36 (0.08)
Σ mono-*ortho* PCBs	136 (77)	41 (24)	29 (7)	44 (33)	n.a.	2*
Σ_{18}PCBs	779 (447)	438 (232)	274 (130)	250 (150)	225 (24)	215 (16)
Σ_{18}PCBs/ % lipid	278	190	125	156	118	143
BSAF values	0.01	0.02	0.03	0.09	0.10	0.30
ΣWHO$_{2005}$-TEQ	0.31 (0.15)	0.29 (0.41)	0.20 (0.17)	0.13 (0.05)	0.001 (0.0002)	0.01 (0.005)
O. niloticus						
% lipid content	2.2 (0.4)	1.9 (0.6)	1.5 (0.4)	1.2 (0.3)	1.1 (0.4)	0.8 (0.2)
Σ indictor PCBs	158 (82)	94 (58)	82 (43)	67 (35)	99 (11)	48 (6)
Σ non-*ortho* PCBs	4 (3)	n.d.	1 (1)	2 (1)	0.4 (0.3)	n.a.
Σ mono-*ortho* PCBs	66 (54)	10 (8)	10 (3)	10 (4)	n.a.	n.a.
Σ_{18}PCBs	229 (139)	104 (66)	93 (47)	79 (40)	99 (11)	48 (6)
Σ_{18}PCBs/ % lipid	104	55	62	66	90	60
BSAF values	0.005	0.005	0.016	0.036	0.075	0.127
ΣWHO$_{2005}$-TEQ	0.05 (0.01)	0.13 (0.16)	0.08 (0.05)	0.12 (0.09)	0.001 (0.003)	n.a.

Results are presented as mean values (standard deviation); number of fish muscle homogenates per station (N) = 3; ST – station; n.d. – non detectable; asterisk (*) – only one sample.

On the whole, the $\Sigma_6$indictor PCBs in fish of the present study were an order of magnitude higher than the $\Sigma_7$mono-*ortho*-PCBs and $\Sigma_4$non-*ortho*-PCBs. However, the $\Sigma_6$indicator PCB levels (48-636 pg g^{-1}) in this study were within the limit of 75,000 pg g^{-1} ww set for fish by European Commission[263], implying that the fish were fit for human consumption in regard to indicator PCBs. Indicator PCB congeners such as CB 138 and CB 153 which were predominant in the sediments were also major contributors in *L. niloticus* (Fig. 4.2). Similarly, CBs 138 and 153 were abundant in *O. niloticus* at 75% sites. The high prevalence of the congeners could be attributed to the fact that superhydrophobic POPs, such as the heavily chlorinated PCBs (i.e. CBs 138 and 153) will not accumulate to the same extent as those with somewhat less hydrophobicity given reduced mass transfer of these chemicals in the gastrointestinal tract[256, 264]. For the coplanar PCBs, the mono-*ortho* PCBs were predominant (their contribution to the Σ_{12}PCBs ranged from 85 to 100%). CB 118 was the most abundant congener, accounting for 45 to 66% of the Σ_{12}PCBs, in comparison to other coplanar compounds. The high levels of CB 118 in the two fish species could be due to the inability of the species to metabolize the congener after accumulation.

It is likely that the PCBs came from the residues of former applications like old engines and power transformers, heat exchangers, hydraulic systems and other industrial uses. The use of these applications containing PCBs was common in Uganda before their commercial production was banned worldwide in 1979[265]. Other potential sources of PCBs could be paint additives, vaporization or leaching from PCB-containing formulations, disposal of waste PCBs or PCB-containing fluids, municipal waste plants and medical waste incinerators[266-270].

The PCB concentrations in *L. niloticus* of the present study were markedly higher (Mann Whitney U-test, $p < 0.05$) than those in *O. niloticus*. Differences in feeding habits and trophic levels could be possible explanations for such inter-specific differences in concentrations. *Lates niloticus* are exclusively piscivorous, consuming all available fish species including its own siblings whereas *O. niloticus* relies on zooplankton and macro-invertebrates[243, 271].

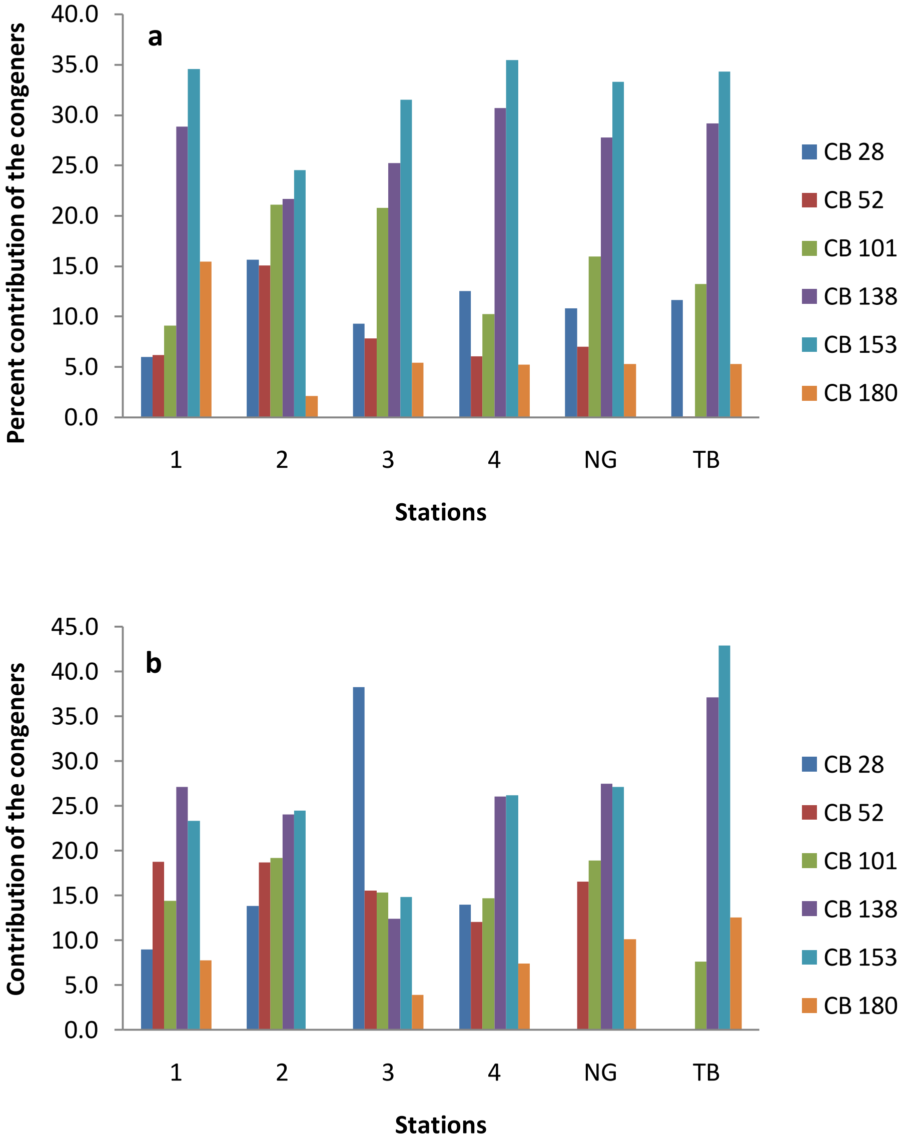


Fig 4.2: Contribution of the congeners to the $\Sigma_6$indicator PCBs at the different stations. a) for *L. niloticus*. b) for *O. niloticus*.

4.6 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 presented in Table 4.6. BDE 209 a commonly detected congener in sediments was not eluted by our column and therefore its concentration are not reported. Also, the determination of PBDEs in sediments from the Napoleon Gulf and Thurston Bay is on-going and the results are not reported herein. The ΣPBDEs varied between stations, with mean values ranging from 60.8 to 179 pg g^{-1} dw. The concentrations in this study were lower than the data in sediments from the Congo River Basin in DR Congo[100], Juksei River in South Africa[142], Niagara River in Canada[153], Ebro River Basin in Spain[150] and the Columbia River in the United States[272]. Just like the PCDD/Fs, the PBDE concentrations in the sediments at site 1 of the present study were higher than the data at other sites. However, a statistical test between the different sites showed no significant differences in PBDE concentrations.

Table 4.6: Mean concentrations of PBDEs (pg g^{-1} dw) and TOC (%) 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)
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.

In this study, tetra-BDE 47 was the most abundant congener in the sediments (Fig. 4.3). The high levels of this congener could be due to the fact that it is resistant to microbial degradation in the environment[273]. 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). This typical PBDE congener profile has been reported worldwide due to the former use of mixtures of penta- and deca-BDEs in the tannery and textile industries, mainly as flame retardant in consumer products [274]. In both applications, deca-BDE formulations are used in a water-based binder dispersion system, which would lead to wastewater emissions in the aquatic environment [275]. Congeners such as BDEs 17, 28, 66, 85, 138, 153, 154 and 184 were also detected, but at lower concentrations. The low concentrations could be attributed to low bioavailability of the compounds in the study areas.

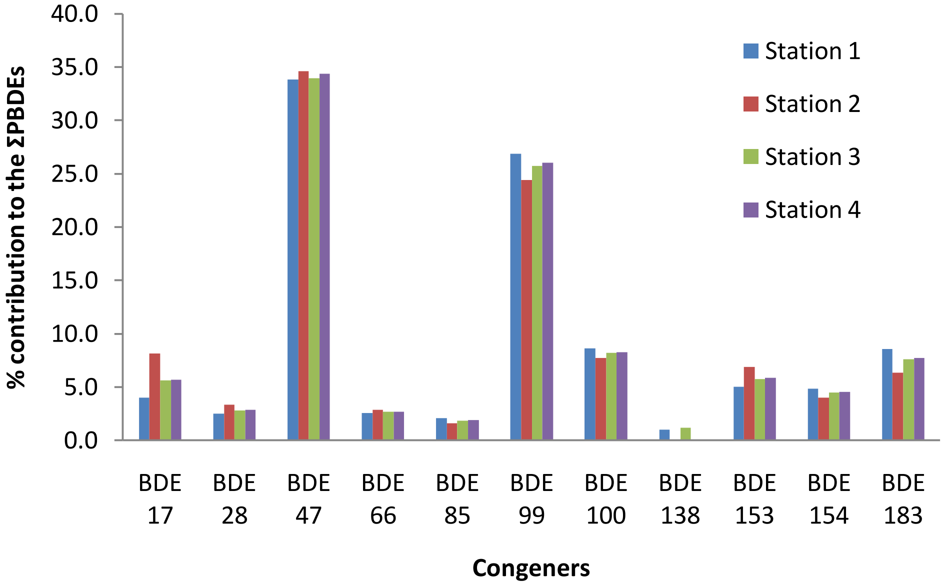


Fig 4.3: Contribution of the different congeners to the ΣPBDEs in the sediments.

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). A statistical test showed no significant difference between the PBDE levels in sediments and fish from the different study stations (ANOVA followed by Tukey's post hoc test, $p > 0.05$). The PBDE levels in the fish of the present study were lower than the data reported in fish from Ghana[276], South Africa[277], China[159, 160], Spain[150, 161], France[162] and the United States[164, 278]. BDE 47 and 99 the most predominant congeners in the sediments were also abundant in *O. niloticus* (Fig. 4.4). The result suggests that *O. niloticus* lacks the capability to metabolize the congeners and are able to preserve congener composition initially present in the sediments. On the other hand, the contribution of BDE 138 to the ΣPBDEs was low in both fish species (<2.1%). The low levels could be due to elimination of the congener in fish through biotransformation to less brominated congeners[279].

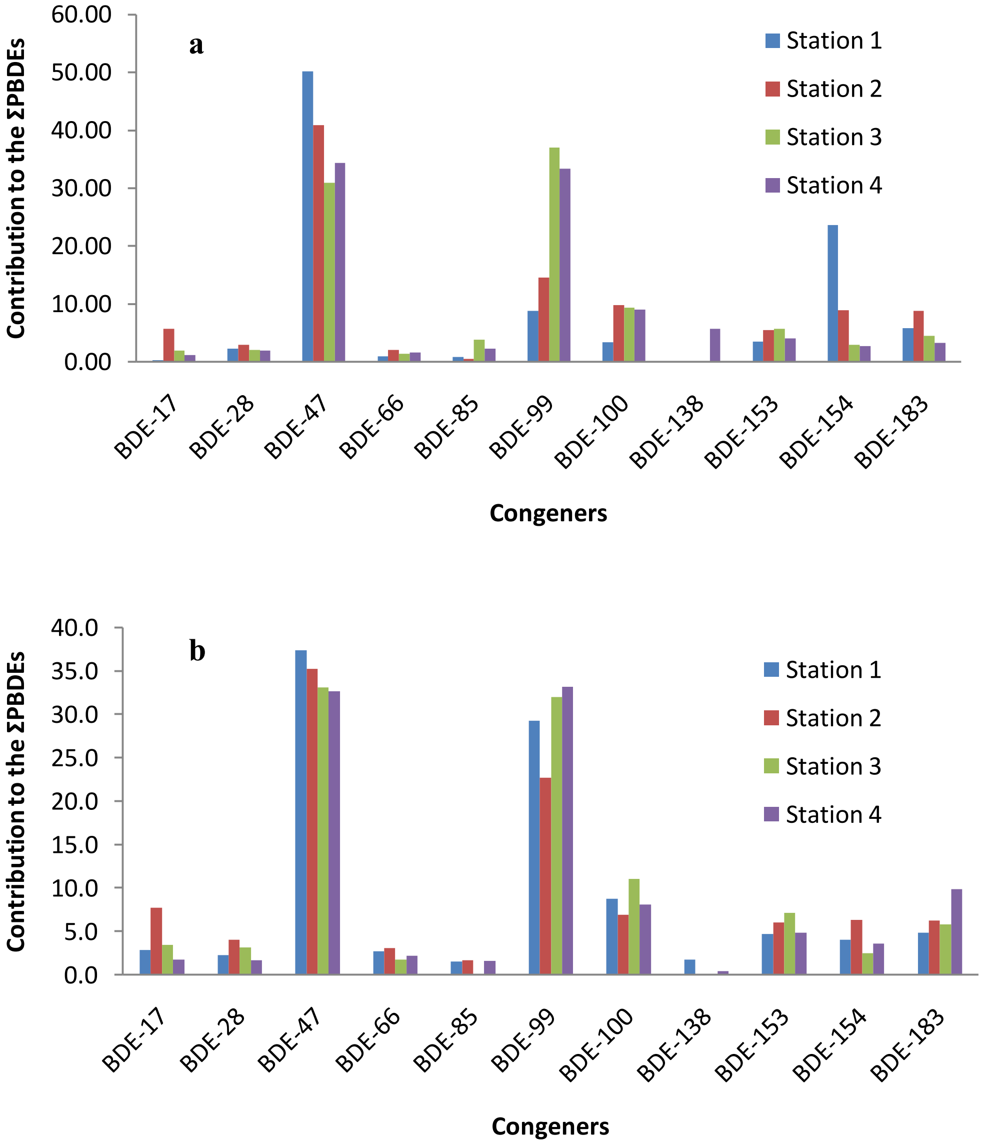


Fig 4.4: Contribution of the congeners to the ΣPBDEs at the different stations.

a) for *L. niloticus*. b) for *O. niloticus*.

4.7 Levels and sources of HCHs in sediments and fish

Three HCH isomers (α, β and γ) were detected in 38% of the fish homogenates. Only the fish from stations A and C of the Napoleon Gulf had detectable HCH levels, while no residues were found in the sediments at other study stations. The concentrations in *L. niloticus* ranged from 5700 to 26,000, 7000 to 34,000 and 5000 to 13,000 pg g^{-1} lw for α-, β- and γ-HCH isomers, respectively (Table 4.7). The maximum concentrations of HCHs in *O.niloticus* were 9000 pg g^{-1} for α-HCH, 8000 pg g^{-1} for β-HCH and 7000 pg g^{-1} for γ-HCH. Regarding the individual contribution to the ΣHCHs, the β isomer accounted for the largest proportion (it contributed >32% in the fish).

Generally, technical HCH contains α-, β-, γ-, δ-HCH isomers of 60-70%, 5-12%, 10-15%, and 6-10%, respectively, while lindane consists of 99% [280]. The physico-chemical properties of these HCH isomers differ from one another. Kim et al.[281] reported that β-HCH is more environmentally persistent than other isomers because of its lower water solubility and vapor pressure, and has 10-30 times higher ability to accumulate in fatty tissues. It should also be noted that α-HCH can be converted to β-HCH in the environment[282]. Therefore, the predominance of β-HCH as the only isomer indicates historical usage of HCHs in the study area[283]. Furthermore, ratios of α-/γ-HCH can be used to establish whether degradation of technical HCH is significant or not, and also whether its input occurred in the past or recently. Yi et al. (2013) established that high ratios (α-/γ-HCH) ranging from 4 to 7 indicate fresh input of technical-HCHs, while low ratios (<3) point at historical use of lindane. The ratios of α-/γ-HCH varied from 0.89 to 1.68 suggesting that the highest HCH residues mainly came from earlier usage and fresh γ-HCH (lindane).

The levels of γ-HCH at all sites were lower compared to other isomers, this could be attributed to better transportability of α-HCH than γ-HCH and/or photochemical transformation and biodegradation of γ-HCH to α-HCH in the environment[284]. Meanwhile, α-HCH has a higher Henry's law constant and vapour pressure than β- or γ-isomers, rendering atmospheric transport more important for this isomer[285].

Table 4.7: Mean concentrations and ranges (pg g^{-1} lw) of HCH isomers in fish from the Napoleon Gulf.

Isomer	Station A		Station C	
	L. niloticus	*O. niloticus*	*L. niloticus*	*O. niloticus*
α-HCH	10,667	4650	15,233	7250
	9000-13,000	n.d.-4800	5700-26,000	n.d.-9000
β-HCH	8667	5100	19,333	7500
	7000-11,000	n.d.-5500	10,000-34,000	n.d.-8000
γ-HCH	6333	5200	11,333	6500
	5000-7000	n.d.-5800	9000-13,000	n.d.-7000
ΣHCHs	25,667	14,950	45,900	21,250
	22,000-28,000	n.d.-15,800	24,700-73,000	n.d.-24,000
α-/γ-HCH	1.68	0.89	1.34	1.12
	1.43-2.60	0.78-1.04	0.63-2.00	0.92-1.29

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

On the whole, the HCH concentrations at station C were higher than those at station A, although statistical analysis showed no significant differences between the two sites. The ΣHCH values in the present study varied widely with mean values ranging from 14,950 to 45,900 pg g^{-1}. The HCH levels were lower than those reported in fish from Tana and Sabaki Rivers in Kenya[185]. In Ghana, Darko et al.[183] reported HCH residues ranging from 700 to 1360 pg g^{-1} lw in fish (*Tilapia zilli*) from Lake Bosomtwi. The results herein were 20 times higher than those by Darko and others. The HCH levels in all the investigated fish were below the extraneous residue limit of 5,000,000 pg g^{-1}, recommended for fish and other fisheries products by FAO/WHO Codex

Alimentarius Commission[286]. In regard to HCHs, the results from the present study give no indication of health risks associated with the consumption of fish from the Napoleon Gulf.

4.8 Safety of fish from Lake Victoria in regard to POPs

The watershed of Lake Victoria has a population of over 1 million people and fish is one of the principal source of protein for these communities. The fish from the lake is intensively consumed and its implications on the health of the population in terms of POPs need to be assessed. The WHO_{2005}-TEQs for PCDD/Fs and dl-PCBs in fish were 0.001-0.16 and 0.001-0.74 pg TEQ g^{-1}ww, respectively. The TEQs were within the permissible level of 3.5 pg g^{-1}ww set for muscle meat of fish and fishery products by the European Commission[263], suggesting minimal health risk from the dietary intake of fish from Lake Victoria. The Agency for Toxic Substances and Disease Registry has determined Minimum Risk Levels (MRL) for oral intake of POPs[287]. With these MRLs, the maximum amount of fish which can be consumed without risk for an average person of 70 kg is calculated with the observed POP concentrations in fish (Table 4.8). For γ-HCH, these amounts were very low, especially in *L. niloticus* (32 g day^{-1}). To exceed the MRL for PCDD/Fs, an average person must eat more than 70 and 140 g of *L. niloticus* and *O. niloticus,* respectively, per day. For PCBs, PBDEs, α-HCH and β-HCH 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 30, 7000, 8000 and 600 ng kg^{-1} body weight day^{-1}, respectively. Therefore, the risk posed by the pollutants is limited, compared to other health and environmental issues, such as the high disease burden due to diarrhea and other infectious diseases, associated with limited availability of sanitation and treated water[288-290].

Table 4.8: Maximum amounts which are recommended to be ate without risk of pollution for an average person of 70 kg based on MRLs[287] and mean concentrations of total PCDD/Fs, PCBs, PBDEs and HCHs found in fish.

	ΣPCDD/Fs	ΣPCBs	ΣPBDEs	α-HCH	β-HCH	γ-HCH
MRL (ng kg^{-1} body weight day^{-1})	0.02	30	7000	8000	600	10
MRL (ng/day) for a person of 70 kg	1.40	2100	490,000	560,000	42,000	700
Mean concentration in *L. niloticus* (ng g^{-1}ww)	0.02	0.36	0.23	32.4	35.0	22.1
Mean concentration in *O. niloticus* (ng g^{-1}ww)	0.01	0.11	0.08	7.74	8.19	7.61
Maximum edible amount of *L. niloticus* (g per day) for a person of 70 kg	70	5833	2,130,434	17,284	1200	32
Maximum edible amount of *O. niloticus* (g per day) for a person of 70 kg	140	19,090	6,125,000	72,351	5128	92

4.9 Relationship between the POP levels in fish and sediments

To establish the relationship between the pollutant concentrations in sediments and fish, the biota-sediment accumulation factors (BSAFs) were calculated. BSAF values give an idea on the bioaccumulation potential of pollutants in living organisms. Usually the values are calculated for organisms that live in contact with the sediment and for which direct partitioning occurs between sediment and organism. This is the case of *O. niloticus*, whose diet is dominated by bottom deposits derived from plankton rain, zoobenthos and zooplankton[291]. The BSAFs for the pollutants in *L. niloticus* were not calculated because it has less contact to the sediments.

The BSAF values for PCDD/Fs, PCBs and PBDEs at the different study locations ranged from 0.003 to 0.029, 0.01 to 0.127 and 0.022 to 0.338, respectively. HCH-BSAFs are not reported because the compounds were not detected in the sediments. The BSAFs for the different pollutants were in the same range, possibly because they have similar log K_{ow} values (6.2-8.3 for PCDD/Fs, 5.0-8.2 for PCBs and 5.7-8.3 for PBDEs). Van der Oost et al.[292] noted that besides the biological or ecological characteristics of the organisms, physical-chemical properties such as K_{ow}, may contribute to POP bioaccumulation in aquatic organisms. PCDD/F-BSAFs in this study were consistent with the data (range 0.0007 to 0.03) found in different studies reviewed by Rifkin and LaKind[293], but were lower than those reported by Matscheko et al.[294] (range 0.1-0.8). The calculated BSAFs for PCBs in this study were lower than those reported by Verhaert et al.[100] in various fish species from the Congo River Basin (range 0.34-126). In Taiwan, Chen et al.[295] reported PBDE-BSAF values ranging from 0.3 to 63.6. The values were an order of magnitude higher compared to the data in this study. Brunson et al.[296] reported an estimated theoretical BSAF value of 1.7, based on partitioning of non-ionic organic compounds between tissue lipids and sediment carbon. The authors noted that a value less than 1.7 indicates less partitioning of an organic compound into lipids than predicted, whereas a value greater than 1.7 indicates more uptake of the pollutant than can be explained by the partitioning theory alone. Therefore, the low BSAFs in this study indicate less partitioning of the pollutants in the tissues of *O. niloticus*, possibly due to its pelagic nature and feeding habits.

In the present study, higher BSAFs were noted at the less polluted sites and with lower total organic carbon (TOC), while lower BSAFs were found at highly contaminated sites with higher TOC. Lake et al.[297] reported PCB-BSAFs to be lower in highly polluted, high organic carbon sediment and sometimes higher in low polluted, low organic carbon sediment. The authors attributed the effect to hydrophobic organic contaminants having greater affinity for polluted, anthropogenic organic carbon; therefore, would be less bioavailable to organisms. To evaluate the applicability of the BSAF concept for risk assessment, correlations between sediment concentrations and BSAFs were analysed using log-transformed data[248]. A significant inverse relationship was found between the sediment POP concentrations and BSAF values in *O. niloticus* (Fig. 4.5). The BSAF values in this study indicate no association between the fish body burden and the contaminants derived from sediments.

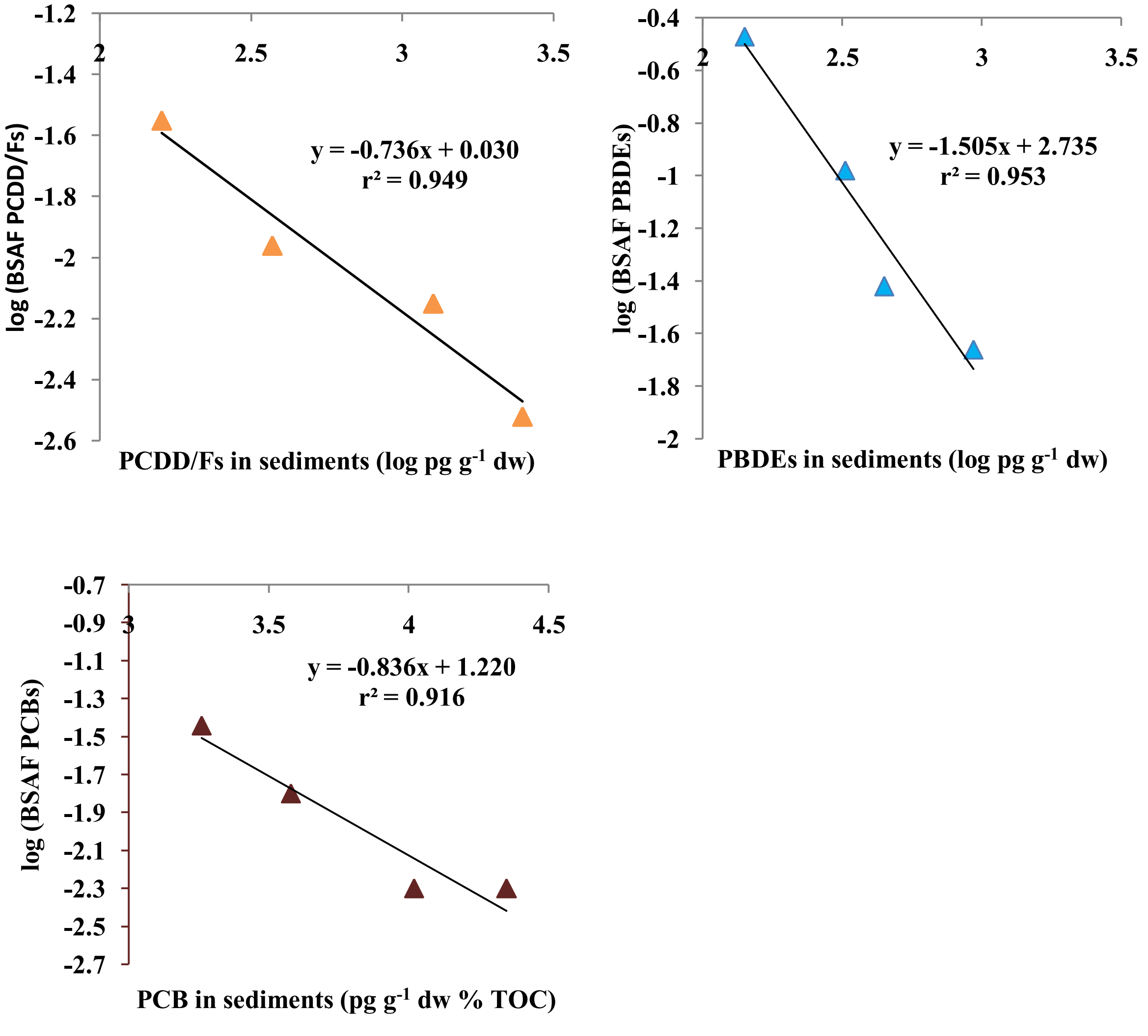


Fig 4.5: Relationship between sediment concentrations and POP-BSAFs

5. CONCLUDING REMARKS AND FUTURE RESEARCH

This study determined the concentrations of PCDD/Fs, PCBs, PBDEs and HCHs in sediments and fish from Lake Victoria (papers 1-V). The stations closest to urban centers, commercial and industrial establishments showed the highest pollutant concentrations compared to those far from point sources. The results from PCA indicated that local combustion processes, including agricultural straw open burning, medical waste incinerators and municipal solid waste combustors were the major sources of PCDD/Fs in the watershed of Lake Victoria (article V).

The POP levels in fish were not significantly related to those in sediments, and the BSAF concept was found to be a poor predictor for the bioavailability and bioaccumulation of environmental pollutants (paper V). WHO-TEQs for PCDD/Fs and dl-PCBs were below the recommended value of 3.5 pg g^{-1} ww set for fish by the European Commission. The HCH levels in the fish were below the extraneous residue limit recommended by FAO/WHO Codex Alimentarius Commission. Based on the Agency for Toxic Substances and Disease Registry criteria, an average person of 70 kg who consumes more than 70 g of fish per day from Lake Victoria was below the set MRLs, indicating minimal health risk associated with dietary exposure in regard to PCDD/Fs, PCBs, PBDEs and HCHs. However, regulatory controls for monitoring and mitigating waste water emissions into Lake Victoria need to be implemented and emphasized by the relevant governmental agencies in Uganda.

Future efforts should be aimed at determining the levels and trends of known POPs as well as novel pollutants in other “hot spots” of Lake Victoria and surrounding water bodies since data in Africa is scarce. In this regard, lakes and rivers should be considered for exposure studies since they are final destinations for most pollutants. Furthermore, the occurrence of persistent chemicals in food of particularly animal origin in Africa needs to be studied. Finally, risk assessments studies of POPs in humans need to be performed given the fact that we are exposed to a mixture of chemicals that could be harmful even at low levels.

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Paper 1

PCDD/Fs and dioxin-like PCBs in surface sediments from Lake Victoria, East Africa

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PCDD/Fs and dioxin-like PCBs in surface sediments from Lake Victoria, East Africa

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HIGHLIGHTS

- Surface sediments from Lake Victoria were analyzed for PCDDs, PCDFs and dl-PCBs.
- The maximum concentration of PCDD/Fs and dl-PCBs was 136pgg—1 dw.
- The WHO2005-TEQs for PCDD/Fs lay in the range of 0.07 to 5.53pg TEQg—1 dw.
- The WHO2005-TEQs for dl-PCBs varied from 0.01 to 0.23pg TEQg—1 dw.
- 23.1% of samples had their PCDD/F results above the set WHOPCDD/Fs-TEQ values.

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ABSTRACT

Surface sediments (<60 cm) from the Napoleon Gulf and Thurston Bay on the northern shore of Lake Victoria were analyzed for polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (dl-PCBs). Fifteen PCDD/Fs and eleven dl-PCBs were found in 75.5% of the samples. The maximum concentrations of PCDD/Fs and dl-PCBs were 44.1 and 136 pg g^{-1} dry weight (dw), respectively. Octachlorodibenzo-*p*-dioxin was the predominant PCDD/F congener at the Napoleon Gulf and Thurston Bay area. Regarding the dl-PCBs, a variation in levels was observed between the mono-*ortho* PCBs and non-*ortho* PCBs, with the former having higher levels than the latter. The PCDD/F and dl-PCB levels, in the sediments of Napoleon Gulf, which is near urban centers and industrial areas were markedly higher ($\alpha \le 0.05$) than those from the Thurston Bay, which is offshore, suggesting that human activities could be sources of the pollutants to the surrounding water resources. World Health Organization-toxic equivalency quotients (WHO-TEQs) lay in the range of 0.07–5.53 pg g^{-1} dw for PCDD/Fs and 0.01–0.23 pg g^{-1} dw for dl-PCBs. 23.1% of samples from the Napoleon Gulf had their results above the set $\text{WHO}_{\text{PCDD/Fs}}$-TEQ value.

1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and the dioxin-like polychlorinated biphenyls (dl-PCBs) are carbon-based organic substances. They possess a combination of physical and chemical properties which include being non-biodegradable (Field and Sierra-Alvarez, 2008), undergoing long-range atmospheric transport (Li et al., 2007) and accumulating in the fatty tissue of living organisms (Çok et al., 2007; Bordajandi et al., 2008). In animals, these contaminants elicit a diverse spectrum of toxic and biochemical responses including body weight loss (Fierens et al., 2003), induction of gene expression (Brouwer et al., 1995) and immunotoxicity (Sormo et al., 2009).

The major sources of PCDD/Fs are thermal processes (Lin et al., 2007), forest fires (Kim et al., 2003), fossil fuel combustion, synthesis of chloro-phenols, production of chloro-organic pesticides, pulp bleaching in the process of paper production (Quaß et al., 2004), among others. PCBs can be released into the atmosphere through leakage of old equipments like transformers and capacitors, leaching from landfills, smokestacks and weathering of asphalt, etc. Despite the extensive information on PCB inventory, use and disposal, especially in the developed nations, only the National Environment Management Authority (2007) has documented their status in Uganda, a country which became an official party of the Stockholm Convention in 2004 (Ritter et al., 2007).

In the environment, the atmosphere is a major pathway for the transport and deposition of PCBs and PCDD/Fs. Once released into aquatic systems, the pollutants tend to accumulate in the sediments for a long period of time and are subject to partitioning, degradation and transport processes. The sediments can then act as natural sinks

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for the pollutants whose re-suspension at the water–sediment interface, especially in storm events and during lake mixing, may increase accumulation and bio-availability in the food chain (Evenset et al., 2007). Relative to the water column, surface sediments are a more appropriate environmental compartment that can be related to the concentrations of pollutants in aquatic organisms (Nguyen et al., 2005).

Of recent, L. Victoria is increasingly experiencing pollution from various sources like biomass burning, industry, transport and households (Awange and Obera, 2007; Kwach and Lalah, 2009). A considerable proportion of the pollutants reach the lake, thereby contributing to deterioration in the water quality (Odada et al., 2004). As far as we know, this is the first report on PCDD/Fs and dl-PCBs in the sediments of L. Victoria. The aim of this study was to evaluate the concentration magnitude of the dioxins in the sediments of L. Victoria and to compare the results with those obtained in studies elsewhere.

2. Materials and methods

2.1. Study area and sample collection

Samples of overlying surface sediments were collected in March 2011 from the Napoleon gulf and Thurston bay (Fig. 1). The Napoleon Gulf is on the shoreline of Jinja town with a dense urban development and industrialization. A great deal of bio-mass burning occurs along the banks of the Thurston Bay where sugarcane growing is also significant. The shorelines of the Napoleon Gulf and Thurston Bay which were previously fringed by papyrus (*Cyprus papyrus*), have been extensively degraded by a rapid rising human population, mounting pressure on the lake, in terms of water quality and sedimentation (Wasswa et al., 2010). For the reasons above, the two locations were earmarked as study areas.

Within each location, samples were taken from marked stations. The stations included A (00°24′50″N, 33°14′20″E), B (00°24′60″N, 33°17′10″E) and C (00°25′70″N, 33°16′40″E) at the Napoleon Gulf, and D (00°25′50″N, 33°20′55″E) and E (00°26′50″N, 33°21′50″E) at the Thurston Bay. A sediment tube sampler was used for coring, to minimize the likelihood of cross contamination; all the utensils used were thoroughly washed and rinsed with acetone, followed by hexane before coring. A total of twenty five samples (five from each station) were collected. The number of collected samples was dictated by the high cost of analysis of PCDD/Fs and dl-PCBs. The samples were transferred into clean solvent-rinsed glass bottles. The bottles were immediately transferred to the laboratory and kept at −28 °C until extraction.

2.2. Reagents, chemicals and standards

All the organic solvents (picograde) used, ^{13}C-labeled PCDD/F and dl-PCB standards were purchased from LGC Standards (Wesel, Germany), and their purity was over 99.9%. Anhydrous sodium sulfate, alumina, C_{18}-modified silica gel, sulfuric acid and silica gel were of analytical grade. The silica gel was heated overnight at 200 °C before use, to reduce the background levels of analytes. Reversible carbon columns filled with 100 mg Carboxen 1016, were purchased from Supelco (Taufkirchen, Germany). The screening included a total of fifteen 2,3,7,8 PCDD/F congeners, four non-*ortho* PCBs (77, 81, 126 and 169) and seven mono-*ortho* PCBs (105, 114, 118, 123, 156, 157 and 167), because they are the most toxic and have a common mode of action. Analytical stock solutions were prepared in amber flasks using *n*-nonane and stored in a fridge maintained at 4 °C.

2.3. Sample extraction and purification procedures

The sediment samples were defrosted prior to extraction. Detailed descriptions of the sample extraction and purification procedures have previously been reported by El-Kady et al. (2007). Briefly, 10 g of the sample was spiked with internal standards (^{13}C-labeled PCDD/Fs and dl-PCBs) and converted to a free flowing powder using

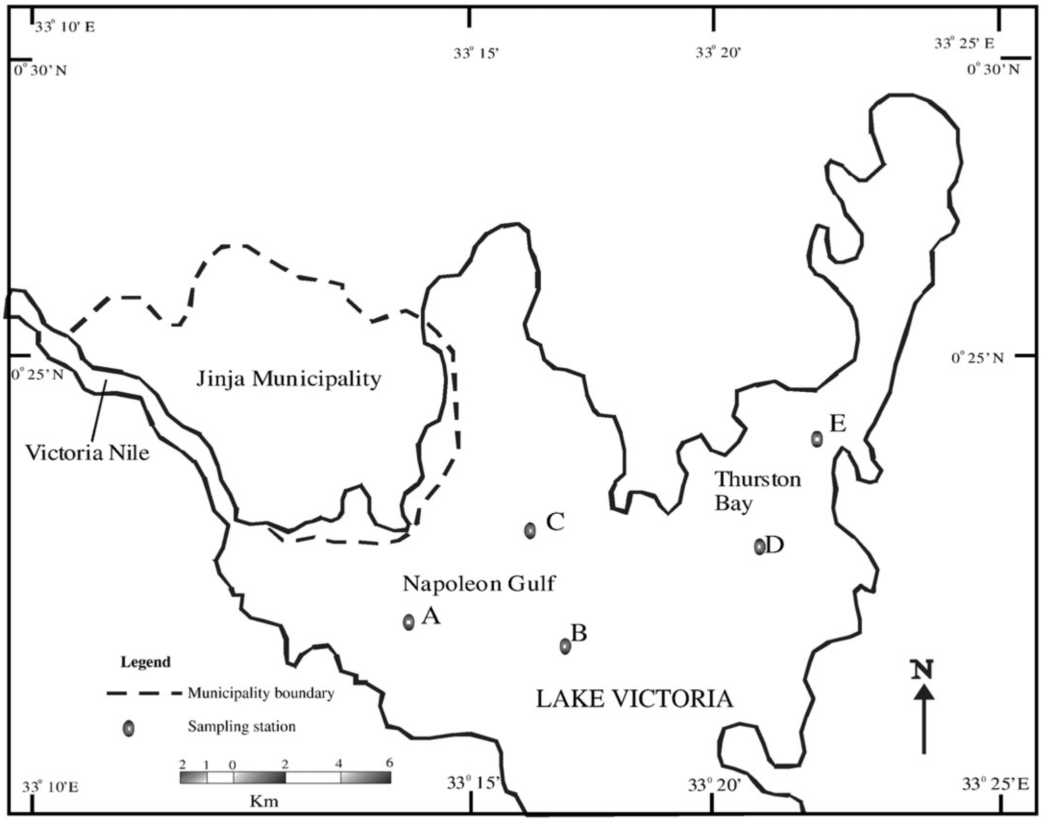

Fig. 1. Map showing the study stations.

hydromatrix™ (Varian). The samples were extracted using an accelerated solvent extractor (ASE 200 Sunnyvatem CA, USA) with two 10 min static cycles of 75/25 *n*-hexane–acetone mixture at 120 °C and a pressure of 120 bar. After extraction, the resultant volume was passed through a drying funnel of anhydrous sodium sulfate and eluted with 100 mL of *n*-hexane. The extracts were reduced to 1 mL on a rotary evaporator and kept for double step purification.

Primary purification was achieved by using a multi-layer silica gel column which was filled with 5 g of anhydrous Na_2SO_4 (bottom), 2 g of silica gel, 3 g of 30% NaOH-silica gel, 1 g of silica gel, 5 g of 44% H_2SO_4-silica gel, 10 g of 22% H_2SO_4-silica gel and 5 g of anhydrous Na_2SO_4 (top). The column was pre-washed with 60 mL of *n*-hexane before it was directly connected to a reversible carbon column (Carboxen 1016, Supelco) that had been rinsed by an equi-volume (25 mL) of toluene and *n*-hexane, respectively. The concentrated extract was added to the multi-layer column and eluted with 100 mL of *n*-hexane. The mono-*ortho* PCBs passed through both columns, while PCDD/Fs and non-*ortho* PCBs were retained in the carbon column. The multi-layer column was removed; the carbon column was washed with 30 mL of dichloromethane/*n*-hexane (1:9, v/v), then reversed and the PCDD/Fs were eluted with 100 mL of toluene. The co-extracted analytes were separately concentrated to 1 mL and kept for secondary purification.

Secondary purification was achieved as described by Çok et al. (2009). Briefly, the first fraction containing PCDD/Fs was eluted through an alumina column filled from top to bottom with 2 g of anhydrous Na_2SO_4, 5 g of alumina and 2 g anhydrous Na_2SO_4. The second fraction containing PCBs was purified using a solid phase extraction cartridge filled with 1 g of C_{18}-modified silica gel. Silica gel was first conditioned with 6 mL of acetonitrile before the extract was added and later eluted with 5 mL of acetonitrile. The resulting purified extracts from both fractions were reduced to <1 mL using a rotary evaporator, transferred to sample vials containing internal recovery standards and then kept for gas chromatographic analysis.

2.4. Instrumental analysis

The analysis of PCDD/Fs and non-*ortho* PCBs was performed with a high resolution mass spectrometer MAT 95S (Thermo Electron GmbH, Bremen, Germany) coupled with an Agilent gas chromatograph 6890 (Agilent Technologies, Palo Alto, CA, USA). Chromatographic separation was achieved by injecting 1 μL extract in the pulsed splitless mode with a period of 120 s on a Restek Rtx-Dioxin2 capillary column (40 m × 0.18 mm i.d. × 0.18 μm). The oven temperature of the gas chromatograph (GC) was held at 130 °C for 1.5 min, rising to 205 °C at 25 °C min^{-1}, then to 310 °C at 4 °C min^{-1} and finally holding it at 310 °C for 15 min. The injection temperature was 300 °C, while the transfer-line temperature was 300 °C. The analysis of mono-*ortho* PCBs was carried out using an Agilent GC 5890 Series 11 equipped with a high resolution mass spectrometer MAT 95S. The separation was achieved by injecting 1 μL on a Phenomenex MultiResidue-2 column (30 m × 0.25 mm i.d × 0.20 μm). The oven temperature was initially programmed at 90 °C. It was then held for 1.5 min, increased to 170 °C at 20 °C min^{-1}, held for 7.5 min, increased to 265 °C at 3.5 °C min^{-1}, then to 310 °C at 20 °C min^{-1} and held for 10 min. Helium was used as a carrier gas at a constant pressure of 16 psi. The mass spectrometer was operated at a resolution of over 10,000 under positive electron ionization conditions (45 eV), and data was obtained in SIM mode using the two most intense ions of the molecular ion cluster for labeled and unlabeled isomers.

2.5. Quality assurance and data analysis

To ensure quality of the data, replicate analyses for precision control, recovery standards for control of losses and internal reference materials for accuracy control were used. The results presented and discussed in this study were blank corrected. One procedural blank was performed for every 5 samples. In the absence of detected blank values the limit of detection (LOD) was defined by a signal to noise ratio (S/N) > 3 times the average baseline variation. If blank values were detected, the LOD was defined as 3 times the standard deviation of all blanks analyzed. The LOD was a function of the extracted mass and varied between analytes (0.01–0.33 pg^{-1} for PCDD/Fs and 0.02–1.00 pg^{-1} for dl-PCBs). The recoveries for surrogate standards ranged between 45% and 108%. All concentrations in the present study were normalized to dry sample weight (dw). Arithmetic means were calculated from only positive quantifiable samples and statistical significance was defined at $p < 0.05$.

3. Results and discussion

3.1. Concentrations of PCDD/Fs in the sediments

The present study is the first investigation on the concentrations of PCDD/Fs in sediments from the Napoleon Gulf and Thurston Bay of L. Victoria. The concentrations of sigma PCDDs varied widely from one location to another (Table 1). The maximum concentration at the study stations was 44.1 pg g^{-1} dw. Octachlorodibenzodioxin (OCDD) was the major contributor in the majority of the samples (>29.7% to sum of seven PCDD congeners) from the Gulf and Bay area. The reason for the domination of OCDD could be that it is emitted at higher rates in the atmosphere than other PCDD congeners. Octachlorodibenzodioxin is usually produced in high amounts as a by-product during combustion of domestic and fuel oil mixtures (Anderson and Fisher, 2002), in sewerage sludge and open fire of garden waste (Wevers et al., 2004). Therefore, the Napoleon Gulf being near urban centers and industrial areas could be receiving OCDD related compounds from the above sources. It was observed that 1,2,3,4,7,8-HxCDD was detected at low levels in most sediments from the stations. One of the reasons for the low 1,2,3,4,7,8-HxCDD levels found could be biotransformation processes. Muir et al. (1985) have reported 1,2,3,4,7,8-HxCDD to undergo more rapid biotransformation compared to other PCDDs.

The concentrations of PCDDs in the sediments from station B were remarkably higher compared to A and C. It was noted that the sediments from station B were clay in nature while those from A and C were sandy. This suggests that grain size of sediments could be an important factor governing the levels of PCDDs. At the Thurston Bay, station E had higher concentrations of PCDDs than D (Table 1). In the vicinity of station E is Kakira sugar plantation, where open burning of sugarcane is normally done every harvesting season. The concentrations of the $\sum$PCDDs recorded in the present study varied from 0.42 to 52.8 pg g^{-1} dw. Our results are nearly the same as those in sediments from Liaohe River in China (Zhang et al., 2010) and the Oder River in Poland (Niemirycz and Jankowska, 2011).

Polychlorinated dibenzofurans in the Napoleon Gulf sediments ranged from 0.07 to 5.61 pg g^{-1} dw. Congener 1,2,3,4,7,8,9-HpCDF was the most abundant homologue in the sediments from station B (based on mean concentrations it represented 21.3% of the $\sum$PCDFs) while 2,3,7,8-TeCDF, the reference compound of the furans group, was the least detected (it contributed less than 8.56% to the $\sum$PCDFs). The concentration ranges in pg g^{-1} dw and contribution to the $\sum$PCDFs by other furans in the sediments from station B were 2,3,4,7,8-PeCDF (1.42–2.72, 12.7%); 1,2,3,4,7,8-HxCDF (1.51–4.13, 15.0%) and 1,2,3,4,7,8,9-HpCDF (2.07–4.89, 21.3%). The PCDF concentrations at station B were 2 fold higher than those from A. The levels of PCDFs from station A were one order of magnitude higher than those at C. It was noted that for most sediments from the Napoleon Gulf, congener 2,3,4,7,8 PeCDF was predominant. Baker and Hites (2000) have reported that 2,3,4,7,8 PeCDF principally comes from technical PCBs and combustion processes. Therefore, their prevalence in the Gulf sediments can be attributed to urban waste incineration

Table 1
Mean concentrations and ranges (pg g^{-1} dw) of PCDD/Fs in sediments from the study stations.

Congeners	Napoleon Gulf			Thurston Bay	
	Station A	Station B	Station C	Station D	Station E
PCDDs					
2, 3, 7, 8-TeCDD	1.15	1.33	n.d	n.d	n.d
	1.10–1.18	1.28–1.38			
1, 2, 3, 7, 8-PeCDD	0.52	2.47	2.43	n.d	n.d
	0.49–0.55	2.33–2.61	2.18–2.70		
1, 2, 3, 4, 7, 8-HxCDD	0.74	1.50	n.d	n.d	0.30
	0.21–1.15	1.48–1.52			0.28–0.32
1, 2, 3, 6, 7, 8-HxCDD	0.70	15.7	0.42	0.33	0.56
	0.44–1.08	4.74–26.7	0.4–0.44	0.31–0.35	0.44–0.67
1, 2, 3, 7, 8, 9-HxCDD	0.66	1.17	0.39	n.d	0.54
	0.57–0.75	1.12–1.22	0.25–0.52		0.25–0.83
1, 2, 3, 4, 6, 7, 8-HpCDD	5.20	2.43	2.23	0.41	3.63
	4.92–5.47	1.83–3.02	0.67–4.16	0.16–0.65	0.37–6.88
OCDD	35.4	10.4	28.8	2.45	36.3
	35–35.5	10.4–10.5	28.7–28.9	2.41–2.49	28.9–44.1
$\sum$PCDDs	44.4	35.1	34.2	3.19	41.4
PCDFs					
2, 3, 7, 8-TeCDF	0.59	1.40	0.35	n.d	0.40
	0.55–0.63	1.38–1.42	0.18–0.51		0.28–0.51
2, 3, 4, 7, 8-PeCDF	0.89	2.07	1.21	0.08	0.47
	0.27–1.74	1.42–2.72	0.32–2.87	0.07–0.09	0.38–0.55
1, 2, 3, 4, 7, 8-HxCDF	0.61	2.45	1.24	0.09	0.38
	0.26–1.26	1.51–4.13	0.31–3.27	0.07–0.11	0.31–0.50
1, 2, 3, 6, 7, 8-HxCDF	0.57	1.81	0.71	n.d	0.59
	0.24–1.15	1.45–2.18	0.40–1.22		0.50–0.68
1, 2, 3, 7, 8, 9-HxCDF	0.23	3.71	0.39	n.d	0.53
	0.19–0.29	1.81–5.61	0.37–0.41		0.41–0.64
1, 2, 3, 4, 6, 7, 8-HpCDF	0.92	1.43	0.85	n.d	1.13
	0.82–1.02	1.41–1.45	0.78–0.92		0.92–1.34
1, 2, 3, 4, 7, 8, 9-HpCDF	0.16	3.48	0.21	n.d	0.21
	0.15–0.17	2.07–4.89	0.18–0.25		0.20–0.23
OCDF	1.14	n.d	1.17	n.d	n.d
	1.12–1.15		1.11–1.27		
$\sum$PCDFs	5.11	16.4	6.13	0.17	3.71
$\sum$PCDD/Fs	49.5	51.4	40.4	3.36	45.1
WHO$_{2005}$-TEQ (PCDD/Fs)	0.98	4.24	1.32	0.08	0.42
	0.21–2.05	1.67–5.53	0.40–1.38	0.07–0.09	0.08–0.69
Ratios ($\sum$PCDFs/$\sum$PCDDs)	0.12	0.47	0.18	0.05	0.13
	0.06–1.86	0.51–0.62	0.10–1.67	0.02–0.05	0.08–1.37

n.d—nondetectable.

and chronic PCB runoff as sources of contamination. A different PCDF congener profile was observed in the Thurston Bay sediments, with 1,2,3,4,7,8 HxCDF being the prevalent congener at station D (52.9% to the grand $\sum$PCDFs) and 1,2,3,4,6,7,8 HpCDF at E (30.5% to the $\sum$PCDFs). Octachlorodibenzofuran (OCDF) was not detected in any sample from the Thurston Bay. Bakoglu et al. (2005) have reported that the main source of OCDF into the environment is mainly chemical manufacturing. Therefore, since the Thurston Bay is located far from industrial and urban activities such a result would be expected.

The levels of $\sum$PCDD/Fs from this study (range 0.20–56.7 pg g^{-1} dw), were an order of magnitude higher than were reported by Kim et al. (2009) in the sediment of Han River, Korea (range 23.1–368 pg g^{-1} dw). The calculated ratios ($\sum$PCDFs/$\sum$PCDDs) ranged from 0.06 to 1.86 and were >1 in 26.3% of the sediments, confirming combustion as one of the possible sources of PCDD/Fs and related compounds into the Napoleon Gulf and Thurston Bay. The ratios of PCDDs/PCDFs can be used to identify the main source(s) of dioxins to the environment. Normally, if the ratio (PCDFs/PCDDs) is >1, the major source is especially combustion (Huang and Buekens, 1995).

3.2. Concentrations of the dl-PCBs in sediments

The dl-PCBs (non-*ortho* and mono-*ortho* PCBs) were detected in 76.9% of the samples. PCB 77 was the major contributor to the $\sum$non-*ortho* PCBs in most samples from the Gulf (stations A 89.3% and C 83.8%) and Bay area (stations D 88.5% and E 85.2%). The high levels of PCB 77 could be attributed to the fact that it does not undergo degradation easily (Miao et al., 1999). Although this isomer was dominant in our study, its concentrations are comparable to those in sediments from Lake Maggiore in Northern Italy (Castro-Jiménez et al., 2008) and, residential and industrial areas in central South Africa (Nieuwoudt et al., 2009). The contribution to the $\sum$non-*ortho* PCBs by PCB 77 was followed by 126 (4.57% for sediments from station A, 11.5% for D and 8.38% for E). On the contrary, the contribution by PCB 77 at station C was followed by 169 (it contributed 8.27% to the $\sum$non-*ortho* PCBs). Low levels of PCB 81 were detected from station A (mean 1.07 ± 0.04 pg g^{-1} dw). The low levels could be due to the high limits of detection observed in our laboratory. Similarly, PCB 169 exhibited low concentrations at all sampling stations (Table 2). PCB 169 tends to show limited mobility to molecular diffusion processes (Chiarenzelli et al., 2001) and is more susceptible to the dissolution loss process (Li et al., 2009). Therefore, the low concentrations in the present study could be attributed to the fact that the congener is more soluble in water. The levels of the $\sum$non-*ortho* PCBs in our study ranged from 1.24 to 39.8 pg g^{-1} dw. The levels are in the same range as those in sediments from Marmara Sea in Turkey (Telli-Karakoç et al., 2002).

With regard to the levels of mono-*ortho* PCBs, 105 and 118 were the most dominant at stations A and C, followed by 114 and 156. This kind of distribution of congeners is also reported by Zhou et al. (2012) in surface sediments from Lake Taihu in China. A variation in concentrations was observed between the mono-*ortho* PCBs and

Table 2
Mean concentrations and ranges (pg g^{-1} dw) of the dl-PCBs in sediment samples from each study station.

Congeners	Napoleon Gulf		Thurston Bay	
	Station A	Station C	Station D	Station E
Non-ortho PCBs				
PCB 77	28.2	14.0	28.2	18.2
	26.0–30.3	13.9–14.1	22.1–34.2	13.7–22.7
PCB 81	1.07	n.d	n.d	n.d
	1.04–1.10			
PCB 126	1.44	1.33	2.78	1.79
	1.09–1.63	1.30–1.35	1.74–3.82	1.32–2.25
PCB 169	0.86	1.40	n.d	1.38
	0.48–1.23	1.19–1.71		1.19–1.57
$\sum$Non-*ortho* PCBs	31.5	16.7	31.0	21.4
Mono-ortho PCBs				
PCB 105	60.1	40.8	25.2	n.d
	47.0–73.3	39.9–41.6	23.1–27.3	
PCB 114	5.99	3.28	3.77	n.d
	5.69–6.28	3.09–3.47	3.21–4.33	
PCB 118	121	81.9	66.1	n.d
	105–136	79.7–84.1	49.7–82.5	
PCB 123	6.73	4.56	n.d	n.d
	6.06–7.40	4.14–4.98		
PCB 156	16.1	23.3	31.7	n.d
	16.0–16.2	13.2–41.1	28.2–35.3	
PCB 157	5.88	6.24	9.08	n.d
	5.68–6.07	3.19–11.8	8.91–9.25	
PCB 167	n.d	12.5	13.4	n.d
		12.0–13.3	12.7–14.1	
$\sum$Mono-*ortho* PCBs	216	173	149	–
$\sum$dl-PCBs	247	189	180	21.4
WHO$_{2005}$-TEQ (dl-PCBs)	0.17	0.09	0.02	0.21
	0.13–0.20	0.01–0.18	0.01–0.03	0.18–0.23

n.d—nondetectable.

non-*ortho* PCBs, with the former having slightly higher concentrations than the latter. Mono-*ortho* PCB congeners are usually present at high concentrations in most commercial PCB mixtures and in environmental samples, and are environmentally persistent than the non-*ortho* PCBs (Hornbuckle et al., 2006). Therefore, the high levels of the mono-*ortho* PCBs are likely to be due to the fact that they undergo slow desorption kinetics. The levels of $\sum$mono-*ortho* PCBs in the present study (range 65.1–244 pg g^{-1}) are in the same range as those reported in sediments from River Nile in Egypt (El-Kady et al., 2007), and are lower compared to the ones in the Great Lakes of Canada (Li et al., 2009). In Uganda, there is no information about the use or any possible source of PCBs. Neither is there any regulation that addresses the control of PCB use nor their disposal. However, the possible sources of the dl-PCBs could be releases from damaged old electric transformers and commercial PCB products.

3.3. World Health Organization-toxic equivalency quotients for PCDD/Fs and dl-PCBs

Toxic equivalency quotients (TEQs) for each sample were calculated using the 2,3,7,8-TCDD toxic equivalent factors (TEFs) according to suggestions by World Health Organization (WHO) (van den Berg et al., 2006). The WHO$_{2005}$-TEQs lay in the range of 0.07–5.53 pg TEQ g^{-1} dw for PCDD/Fs and 0.01–0.23 pg TEQ g^{-1} dw for dl-PCBs. Concerning the WHO-TEQs for PCDD/Fs, OCDD was the most dominant and it accounted for between 36.4 and 83.7% of the fifteen congeners. PCBs 105 and 118 were the main dl-PCBs and together they contributed >61.2% to the TEQs. The TEQs for dl-PCBs in all the samples from the Napoleon Gulf and Thurston Bay were below the interim sediment quality guideline of 0.85 pg WHO-TEQ g^{-1} dw recommended by the Canadian Council of Ministers of the Environment (2002). While, the TEQs for PCDD/Fs in sediments from Thurston Bay were below 0.85 pg g^{-1}, 23.1% of samples from the Napoleon Gulf had their TEQs above the set value.

The WHO$_{PCDD/Fs}$-TEQ values at station B (range 1.67–5.53 pg g^{-1} dw) were higher compared to those at D (0.07–0.09 pg g^{-1} dw) and E (0.08–0.69 pg g^{-1} dw), but comparable to the ones of stations A (range 0.21–2.05 pg g^{-1} dw) and C (0.09–3.21 pg g^{-1} dw). The WHO$_{PCDD/Fs}$-TEQs in the present study were twice those of sediments from the Pearl River Delta in China (Zhang et al., 2009), in the same range as those of the Mondego estuary in Portugal (Nunes et al., 2011), but lower than the ones of Liaohe River in China (Zhang et al., 2010) and the trans-boundary rivers in Belgium–France (Sanctorum et al., 2011). In light of the above, it could be concluded that the contamination of the lake is within permissible limits for dl-PCBs except for PCDD/Fs in 23.1% of the samples.

4. Conclusion

In this study, detectable levels of PCDD/Fs and dl-PCBs were found in surface sediments from L. Victoria. The measured concentrations and WHO-TEQ values for PCDD/Fs and dl-PCBs were lower compared to those reported in most studies dealing with fresh water sediments. However, since sediments play a role of first supplier to benthic feeders and consequently the biomagnifications of pollutants to higher trophic animals, the presence of even trace amounts of PCDD/Fs and dl-PCBs in the sediments of L. Victoria could be of harm since they may end up being ingested by aquatic organisms like fish.

Conflict of interest

There is no conflict of interest.

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Paper 11

PCDD/Fs and dioxin-like PCBs in fish species from Lake Victoria, East Africa

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PCDD/Fs and dioxin-like PCBs in fish species from Lake Victoria, East Africa

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HIGHLIGHTS

- Fish species collected from Lake Victoria were extracted for PCDD/Fs and dl-PCBs.
- Concentrations of the $\sum$PCDD/Fs in the muscles ranged from 0.06 to 0.59 pg g^{-1} fw.
- The $\sum$dl-PCBs in the fish muscles and livers ranged from 0.2 to 19.0 pg g^{-1} fw.
- The WHO$_{2005}$-TEQs varied from 0.001 to 0.74 pg TEQ g^{-1} for PCDD/Fs and dl-PCBs.
- Based on the calculated WHO-TEQs, the fish was fit for human consumption.

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ABSTRACT

Two commercially important fish species, Nile perch (*Lates niloticus*) and Nile tilapia (*Oreochromis niloticus*) belonging to different trophic levels were collected from the Napoleon Gulf and Thurston Bay in Lake Victoria. Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (dl-PCBs) were extracted from the fish muscles and livers using the ^{13}C isotope dilution method, followed by multiple column chromatography clean-up. Analysis was achieved by a high resolution gas chromatography coupled with a high resolution mass spectrometer. The concentrations of analytes ranged from 0.07 to 0.59 pg g^{-1} fresh weight (fw) and 0.3–19.0 pg g^{-1} in *L. niloticus* and 0.06–0.18 and 0.2–15.7 pg g^{-1} in *O. niloticus*, for $\sum$PCDD/Fs and $\sum$dl-PCBs, respectively. Differences in congener concentrations were observed between the two fish species and study sites, and this was attributed to differences in feeding habits and trophic levels. World Health Organization-toxic equivalents (WHO-TEQs) were in the range 0.01–0.16 pg TEQ g^{-1} for the PCDD/Fs and 0.001–0.74 pg TEQ g^{-1} for the dl-PCBs. The TEQ values in the present study were lower compared to those of most fish samples reported in literature and were within permissible levels recommended by the European Union, implying that the fish was fit for human consumption.

1. Introduction

Lake Victoria has the largest fresh water fishery in East Africa with annual yields exceeding 800 000 metric tons (Odongkara et al., 2009). By 2008, increasing industrialization had been noted in the watershed together with a large human population of more than 30 million people that depend on the fish as a primary source of fats and proteins (Johnson, 2009). In spite of the importance of fish to the diet of the local population, as well as it being an important export commodity, a survey of literature shows no available data concerning the profiles of PCDD/Fs and dl-PCBs in the aquatic environment of Lake Victoria. Globally, these organic pollutants are widely spread in the environment and of concern especially because they are highly hydrophobic and, resistant to biotic and abiotic degradation (Laisi et al., 2008). The congeners of PCDD/Fs with chlorine substitution in the 2, 3, 7, 8 positions are thought to pose a risk to human health due to their toxicity, carcinogenic potency and potential effects on animal immunological and reproductive systems (Kumar et al., 2001). They tend to accumulate in top predators (Braune and Simon, 2003), and are thought to damage natural hormones (Minh et al., 2004).

Elsewhere in the World, concern about the health implications of PCDD/Fs and dl-PCBs has led to numerous surveys to determine their levels in fish products among other foods in order to assess human exposure (Zhang et al., 2008; Moon and Choi, 2009). The

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fish tend to accumulate amounts of such non-ionic organic chemicals at thousands of times their concentration in the surrounding medium through consumption of phytoplankton, insects, suspended particles and contaminated sediments (Bush and Kadlec, 1995). Consequently, human dietary intake of these chemicals is considered one of the major pathways, especially through consumption of fish (Bayarri et al., 2001). This study was aimed at obtaining preliminary information on the occurrence of PCDD/Fs and dl-PCBs in fish species from Lake Victoria. The generated data will be used as a benchmark to compare the levels of the pollutants over time.

2. Materials and methods

2.1. Study area and sampling

Two sampling areas namely, the Napoleon Gulf and Thurston Bay in Jinja Municipality were selected for this study. Jinja Municipality has dense urban development and industrialization, and these land use activities could be sources of pollutants into Lake Victoria via runoff. The Napoleon Gulf is adjacent to Jinja Municipality on the northern shoreline of Lake Victoria, where River Nile (the only outlet of the lake) begins its journey to the Mediterranean Sea, while the Thurston Bay which is also located on the same shoreline is rather far from Jinja town (Fig. 1). A total of 64 fish of different species (*Lates niloticus* and *Oreochromis niloticus*) and ages 1–3 years were collected in March, 2011. The weights of *L. niloticus* varied from 1522.6 to 2876.9 g (mean 2262.9 g) while those of *O. niloticus* ranged from 274.8 to 552.4 g (mean 406.3 g). The mean length (±standard deviation) of the latter species was 27.6 ± 2.6 cm, whereas that for the former was 65.2 ± 4.7 cm. Specimens were individually rinsed with distilled water to remove any impurities. Subsequently, the muscles and livers of 4–5 fish of the same species and location were separately excised, pooled and macerated to obtain homogenized samples. The samples were transferred into clean glass bottles and sealed using aluminum caps. The bottles were kept in cooling boxes containing ice packs and transferred to the German Research Center for Environmental Health for laboratory chemical analysis, and frozen at −28 °C until analysis.

2.2. Chemical analysis

The tissues were allowed to thaw and a known mass was spiked with ^{13}C-labelled PCDD/F and dl-PCB surrogate standards. The spiked tissues were then mixed with hydromatrix™ (Varian) to remove any moisture. Extraction was carried out using an accelerated solvent extractor (Dionex 200, Sunnyvatem CA, USA) at a temperature of 120 °C and pressure of 120 bar, using a 3:1 v/v of *n*-hexane/acetone as extraction solvent. The resulting organic extract was passed through a drying funnel of anhydrous sodium sulfate and eluted with 100 mL of *n*-hexane. The extraction volume was concentrated to 2 mL on a rotary evaporator and kept for purification to remove interferences by two sequential liquid chromatography steps.

The first purification step was performed by adsorption chromatography using a multilayered sandwich glass column (packed successively from bottom to top with 2 g silica gel, 5 g 33% silica gel-sodium hydroxide, 2 g silica gel, 5 g 44% silica gel-sulfuric acid, 10 g 22% silica gel-sulfuric acid and 5 g sodium sulfate). The column was washed with 60 mL *n*-hexane before being connected to a reversible carbon column (Carboxen 1016, Supelco) that had been rinsed by an equivalent volume (25 mL) of toluene and *n*-hexane, respectively. The concentrated extract was then applied to the column and eluted with 100 mL of *n*-hexane. PCDD/Fs and non-*ortho* PCBs were retained in the carbon column, while the mono-*ortho* PCBs passed through both columns. The sandwich column was removed and the carbon column was further eluted with

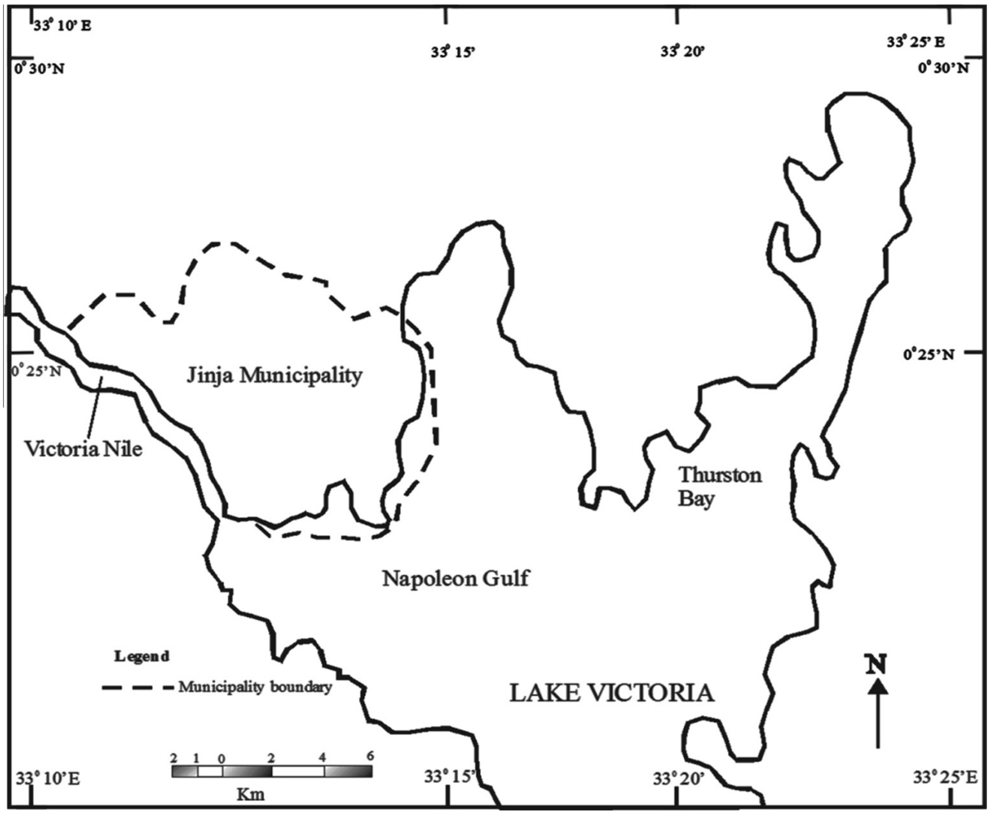

Fig. 1. Map showing the study areas (adopted from Ssebugere et al. (in press).

30 mL of 10% dichloromethane in *n*-hexane, and the resultant solution concentrated. The carbon column was reversed, eluted with 100 mL toluene and the resultant solution was concentrated to <2 mL. The second purification step was achieved by using a chromatographic column filled with 5 g alumina B super 1 as described in Çok et al. (2009). The purified fractions were transferred to sample vials followed by addition of internal standards containing ^{13}C-labelled PCDD/Fs and dl-PCBs prior to gas chromatographic determinations.

Sample extracts were analyzed for PCDD/Fs and non-*ortho* PCBs using an Agilent model 6890 gas chromatograph (GC) equipped with a high resolution mass spectrometer (HRMS) MAT 95S. Chromatographic separation was achieved by injecting 1 µL of extract in the pulsed splitless mode with a period of 120 s on a Restex Rtx-Dioxin 2 capillary column (40 m × 0.18 mm i.d. × 0.18 µm). The instrumental analysis of the mono-*ortho* PCBs was performed on an Agilent model 5890 GC series 11 coupled with a HRMS MAT 95S. The separation was achieved by injecting 1 µL of the extract on a Phenomenex MultiResidue-2 column (30 m × 0.25 mm i.d × 0.20 µm). The MS was operated at a dynamic mass resolution >10000 and quantification was achieved by isotope dilution method.

2.3. Quality assurance

Matrix spiking tests were run for every five samples. The limit of detection (LOD) for PCDD/Fs and dl-PCBs, defined by a signal to noise ratio (S/N) > 3 times the average variation of the baseline, ranged from 0.01 to 0.07 and 0.01 to 7.9 pg g^{-1} fw, respectively. The recoveries from spiked samples were between 42% and 109%. Concentrations of PCDD/Fs and dl-PCBs were calculated using HRMS software for only samples above the LOD. Samples below the LODs were not considered so as to eliminate artificial bias. All statistically significant correlations were expressed as $p < 0.05$.

3. Results and discussion

3.1. Concentrations of PCDD/Fs in the fish muscle homogenates

The results from analysis of PCDD/Fs from fish homogenates on a fresh-weight (fw) basis are presented in Table 1. The highest concentration of PCDD/Fs in the muscles of *L. niloticus* was 0.21 pg g^{-1}. Congener 1,2,3,7,8-PeCDD was the only detected PCDD (range n.d to 0.1 pg g^{-1}) in the muscle homogenates of *L. niloticus* from the Napoleon Gulf. The concentration ranges of PCDFs in the muscle of the same fish species from the Gulf were 0.09–0.21, 0.08–0.09, n.d to 0.13, 0.12–0.17, 0.08–0.09, n.d to 0.08, n.d to 0.06, n.d to 0.15 pg g^{-1} for 1,2,3,4,6,7,8,9-OCDF, 1,2,3,6,7,8-HxCD, 1,2,3,4,7,8,9-HpCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,6,7,8-HxCDF, 1,2,3,4,7,8-HxCDF, 2,3,4,7,8-PeCDF and 1,2,3,7,8-PeCDF, respectively. The PCDF ranges in *L. niloticus* from the Thurston Bay were n.d to 0.09 pg g^{-1} for 1,2,3,4,7,8-HxCDF, 0.07–0.14 pg g^{-1} for 1,2,3,4,6,7,8-HpCDF, n.d to 0.17 pg g^{-1} for 1,2,3,4,6,7,8,9-OCDF. The levels of PCDFs in most cases were higher than PCDDs suggesting combustion as a possible source of these pollutants. Baker and Hites (2000) pointed out that for given samples if PCDFs are greater than PCDDs, then combustion is the major source of dioxins and/or related compounds into the environment.

It was observed that OCDF was the major contributor to the $\sum$PCDD/Fs in most *L. niloticus* muscle homogenates (18.6–45.6%). The pattern of PCDD/Fs characterized by high OCDF in the present study is similar to that reported by Wan et al. (2005) in six fish species (*Lateolabras japonicus*, *Chaeturichthys sitgmatias*, *Platycephalus indicus*, *Nibea albiflora*, and *Liza so-iuy*) from Bohai Bay in northern China. The $\sum$PCDD/Fs in the muscles of *L. niloticus* in our study varied from 0.07 to 0.59 pg g^{-1} fw. The levels are lower than those reported by Bordajandi et al. (2003) in fresh water fish species (*Salmo trutta*, *Anguilla Anguilla* and *Barbus barbus*) from River Turia basin in Spain and Fang et al. (2007) in fish (*Ictalurus punctatus*, *Pampus agrenteus* and *Cyprinus carpio*) from Lake Dongting in the people's republic of China.

For *O. niloticus* from the Napoleon Gulf, the only detected PCDD/F in the muscle homogenates was 1,2,3,4,6,7,8-HpCDF, at a maximum concentration of 0.1 pg g^{-1}. Three congeners were detected in *O. niloticus* from the Thurston Bay, these included 1,2,3,4,7,8-HxCDF (range n.d to 0.06 pg g^{-1}), 1,2,3,6,7,8-HxCDF (range n.d to 0.05 pg g^{-1}) and 1,2,3,4,6,7,8-HpCDF (range 0.06–0.07 pg g^{-1}). On the whole, the concentrations of PCDD/Fs in *L. niloticus* were higher than those in *O. niloticus* from the Gulf and Bay area. The high levels of PCDD/Fs in *L. niloticus* could be attributed to the fact the species is a benthic feeder. Traunspurger and Drews (1996) revealed that fish living in benthic regions of aquatic habitats display higher whole body levels during their cycle due to their close contact with sediments. The levels of PCDD/Fs in *O. niloticus* in the study herein were 5–10-fold lower compared to those reported by El-Kady et al. (2007) in the same fish species from River Nile in Egypt.

The concentrations of low chlorinated 2,3,7,8-substituted-PCDD/Fs did not exhibit statistically significant trends ($p > 0.05$) with trophic levels while those of the high chlorinated 2,3,7,8-substituted-PCDD/Fs declined significantly with increasing trophic levels, implying that the congeners undergo trophic dilution. This observation is similar to that reported by Naito et al. (2003) in fish species (*Lateolabrax japonicus*, *Dasyatis akajel*, *Conger nyriaster*, *Konosirus punctatus*, *Kareius bicoloratus* and *Limanda yokohamae*)

Table 1
Mean concentrations (pg g^{-1} fw) of PCDD/Fs in fish muscle homogenates.

Congener	Napoleon Gulf		Thurston Bay	
	L. niloticus (*n* = 4)	*O. niloticus* (*n* = 3)	*L. niloticus* (*n* = 3)	*O. niloticus* (*n* = 4)
PCDDs				
1,2,3,7,8-PeCDD	0.10	<0.09	<0.08	<0.07
1,2,3,4,7,8-HxCDD	<0.06	<0.05	0.15	<0.04
PCDFs				
1,2,3,7,8-PeCDF	0.15	<0.01	<0.01	<0.01
2,3,4,7,8-PeCDF	0.06	<0.01	<0.01	<0.01
1,2,3,4,7,8-HxCDF	0.08	<0.01	0.09	0.06
1,2,3,6,7,8-HxCDF	0.09	<0.01	<0.01	0.05
1,2,3,4,6,7,8-HpCDF	0.17	0.11	0.21	0.07
1,2,3,4,7,8,9-HpCDF	<0.02	0.13	<0.02	<0.01
OCDF	0.16	0.09	0.17	<0.02
$\sum$PCDD/Fs	0.81	0.33	0.62	0.18
WHO$_{2005}$-TEQ	0.07	0.01	0.03	0.02

n – number of fish muscle homogenates.

Table 2
Mean concentrations (pg g^{-1} fw) of dl-PCBs in fish tissues.

Congener	Napoleon Gulf				Thurston Bay			
	L. niloticus		*O. niloticus*		*L. niloticus*		*O. niloticus*	
	Muscle (*n* = 4)	Liver (*n* = 4)	Muscle (*n* = 3)	Liver (*n* = 3)	Muscle (*n* = 3)	Liver (*n* = 3)	Muscle (*n* = 4)	Liver (*n* = 4)
Non-ortho PCBs								
PCB #77	2.2	<2.0	0.4	<0.2	<0.2	<4.4	<3.7	7.8
PCB #126	<0.1	<1.6	<0.4	<0.1	0.3	<0.1	0.2	7.2
Mono-ortho PCBs								
PCB #123	<0.3	<2.6	<0.2	14.1	0.8	<7.9	<0.2	<3.3
PCB #157	<0.2	5.1	<0.2	6.9	0.5	<2.1	<0.1	6.6
PCB #167	<0.3	11.8	<0.2	7.6	0.9	19.0	<0.1	15.7
∑PCBs	2.2	16.9	0.4	28.6	2.5	19.0	0.2	37.3
WHO_{2005}-TEQ	0.001	0.02	0.001	0.01	0.01	0.02	0.02	0.74

n – number of fish tissue homogenates.

from the aquatic food web of Tokyo Bay. None of the fish liver homogenate in the current study had detectable levels of PCDD/Fs. This could be attributable to poor limits of quantification observed in our laboratory for the livers. Toxic equivalents quotients (TEQs) were calculated for each homogeneous sample using 2,3,7,8-TCDD toxic equivalent factors (TEFs) reported by WHO in 2005 for fish (van den Berg et al., 2006). The WHO_{2005}-TEQs for PCDD/Fs in the present study ranged from 0.01 to 0.17 pg TEQ g^{-1} fw. Our TEQs were lower than those reported in different fish species from water bodies elsewhere in the world (Matthews et al., 2008; Storelli et al., 2011; Pacini et al., 2013).

3.2. Levels of dl-PCBs in the fish tissues

The analyzed categories were the non-*ortho* and mono-*ortho* PCBs. Total PCB concentrations (as the sum of all the PCB congeners) varied largely between fish species with values ranging from 0.2 to 37.3 pg g^{-1} fw. For *L. niloticus* from the Napoleon Gulf, PCB 77 was the only detected non-*ortho* congener in the muscle homogenates (mean 2.1, range 1.8–2.4 pg g^{-1}). The high levels of PCB 77 could be attributed to the fact that it is subject to greater specific contaminant exposure in comparison to other congeners. Adu-Kumi et al. (2010) have also reported high levels of PCB 77 (up to 56 pg g^{-1}) compared to 81, 126 and 169 in fish from Lakes; Volta, Bosumtwi and Weija in Ghana. Only two mono-*ortho* PCB congeners (157 and 167) were detected in the livers of *L. niloticus* at concentrations of 5.1 ± 0.1 and 11.8 ± 0.1 pg g^{-1}, respectively. The muscles of *L. niloticus* from the Thurston Bay presented four dl-PCB congeners, these included 167 (mean 0.9 pg g^{-1}), 123 (0.8 pg g^{-1}), 157 (0.5 pg g^{-1}) and 126 (0.3 pg g^{-1}). The low levels of PCB 126 could be due to the ability of the two fish species to eliminate this pollutant via urinary excretion and/or during respiration. McFarland and Clarke (1989) reported that PCB congeners with low chlorination grade are more readily metabolized and eliminated from tissues than the highly chlorinated congeners. For *O. niloticus*, only the non-*ortho* congeners were detected in the muscles of fish from the Napoleon Gulf and Thurston Bay. The detected congeners were PCB 77 (mean concentration 0.4 ± 0.1 pg g^{-1}) and 126 (0.2 ± 0.1 pg g^{-1}). The dl-PCB concentrations in the livers of *O. niloticus* from the Gulf were higher than in the muscles (Table 2). The mean concentrations of PCB 77, 126, 157 and 167 in the liver homogenates of *O. niloticus* from the Bay were 7.8, 7.2, 6.6, 15.7 pg g^{-1}, respectively.

In most cases, the dl-PCBs reflected higher contaminant levels in *L. niloticus* compared to *O. niloticus*, although the opposite was true for PCDD/Fs, possibly due to differences in relative accumulation rates. The accumulation of pollutants by a particular species could be attributed to differences in exposure route via trophic transfer. This may have played part in the observed differential response of the fish species. The dl-PCBs in the fish livers were 2–3 folds higher than those in the muscle and this could be attributed to differences in uptake between the two tissues. The tissue homogenates of *O. niloticus* from the Thurston Bay had higher dl-PCB levels compared to *L. niloticus* which is at a high trophic level in the food-web, and this could be due to differences in desorption kinetics of the compounds and/or the diet of the fish. Barron (1990) revealed that the diet and feeding habits coupled with physiological and biochemical processes within the organism play an important role in the accumulation of contaminants.

The PCDD/F and dl-PCB levels in the fish tissues from the Thurston Bay were less consistent and no statistical differences ($\alpha = 0.05$) were observed between species. The small catchment area of the Thurston Bay, its vegetation cover and processes in the catchment could probably affect contaminant transfer and deposition. The WHO_{2005}-TEQ values for PCBs in fish from the Gulf and Bay lay in the range of 0.001–0.74 pg g^{-1} fw. The mono-*ortho* PCBs contributed the highest percentage to the total TEQ content than the non-*ortho* PCBs in both *L. niloticus* and *O. niloticus*. Similar studies by Bordajandi et al. (2003) and Tanabe et al. (2004) have also reported the domination of the mono-*ortho* PCBs in comparison to the non-*ortho* PCBs. The TEQ values in our study were lower compared to those reported literature (Baeyens et al., 2007; Bocio et al., 2007; Moon and Choi, 2009) and were also within the permissible level of 4 pg g^{-1} fw set by the European Union (EU, 2001).

4. Conclusion

PCDD/F and dl-PCB congeners were detected in fish homogenates from Lake Victoria. The concentrations for the pollutants were in the descending order of mono-*ortho* PCBs, non-*ortho* PCBs, PCDFs and PCDDs. The congener specific contribution to WHO-TEQs showed mono-*ortho* PCBs as the most abundant congeners compared to the non-*ortho* PCBs. The concentrations and WHO_{2005}-TEQs in our study were lower compared those reported in literature and were below the maximum residues limit recommended by the European Union. Therefore, based on our results the consumption of fresh fish from Lake Victoria does not represent a risk to human health.

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Paper III

Polychlorinated biphenyls and hexachlorocyclohexanes in sediments and fish species from the Napoleon Gulf of Lake Victoria, Uganda

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Polychlorinated biphenyls and hexachlorocyclohexanes in sediments and fish species from the Napoleon Gulf of Lake Victoria, Uganda

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HIGHLIGHTS

- High concentrations of PCBs were found at locations near Jinja Municipality.
- The PCB and HCH levels were low to moderate compared to other locations worldwide.
- The fish from the Napoleon Gulf was fit for consumption in regard to PCBs and HCHs.
- The ratios of α-/γ-HCH were low suggesting past input of lindane.

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Lake Victoria

ABSTRACT

Polychlorinated biphenyls (PCBs) and hexachlorocyclohexanes (HCHs) were analyzed in surface sediments (<30 cm depth) and two fish species: Nile perch (*Lates niloticus*) and Nile tilapia (*Oreochromis niloticus*). The samples were collected from the Napoleon Gulf on the northern shore of Lake Victoria. The analysis was done using a gas chromatograph (GC) coupled to a high resolution mass spectrometer for PCBs and a GC equipped with an electron capture detector for HCHs. Total (Σ) PCBs in the muscles of fish varied widely with mean values ranging from 41 to 670 pg g^{-1} lipid weight (lw). The PCB levels in *L. niloticus* were significantly greater than those in *O. niloticus*. The large variability observed in the data was attributed to differences in feeding habits and trophic levels. While *O. niloticus* is a filter-eating fish species feeding mainly on phytoplankton and zooplankton, *L. niloticus* have predatory feeding behaviors and prefer a diet of live fish and, therefore, are more prone to bioaccumulate contaminants. The mean PCB concentrations in the sediments varied from 362 to 848 pg g^{-1} dry weight. Variations in PCB levels were observed from one study site to another, this was attributed to the nature and particle size of the sediments. HCH isomers were detected in fish at mean concentrations of up to 45,900 pg g^{-1} lw. The PCB and HCH concentrations were lower than those from previous studies elsewhere in literature and were below the maximum residue limits set by the European Commission and FAO/WHO Codex Alimentarius Commission, implying that the fish was fit for human consumption.

1. Introduction

Lake Victoria is the world's second largest freshwater lake after Lake Superior in North America. Economically millions of people depend on the lake as a source of drinking water and fishing as a principal source of proteins to the local diet. The lake's fisheries contribute an annual foreign income of USD $124 million to Uganda's economy constituting the third foreign earner, after coffee and flowers (MAAIF, 2010). In recent years the lake is increasingly experiencing pollution from human and industrial wastes. The possible pollutants reaching the lake could be polychlorinated biphenyls (PCBs) and hexachlorocyclohexanes (HCHs). PCBs are synthetic organic compounds that were widely used in electric transformers, capacitors, printers and ink paints in the 1960s (Breivik et al., 2002, 2004). PCBs can still be formed as unintentional byproducts of waste incineration and industrial processes

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(Liu et al., 2009). However, concerns about their persistent and toxic nature led to their ban throughout the world in the late 1970s (Breivik et al., 2007). Technical HCH was widely used in Uganda and other parts of the world as an insecticide on fruits and vegetables in the 1980s (Ejobi et al., 1996; Lopez et al., 2012). The chemical was also banned worldwide in the 1990s because it is highly toxic (Li, 1999). Although, the use of PCBs and HCHs was stopped, their impacts on the environment cannot be neglected.

These pollutants reach water bodies via run-off and/or atmospheric transport. In aquatic ecosystems, small amounts of PCBs and HCHs may be re-dissolved at the water–sediment interface, but mostly tend to partition into sediments and suspended particles (Horzempa and Ditoro, 1983; Eisenreich et al., 1989). The chemicals have the potential to bioaccumulate across the food chain, building up in top predators through consumption of contaminated biota (Bjermo et al., 2013; Frouin et al., 2013). The fish at the top of the aquatic food chain are most likely to be affected by exposure to such pollutants (Bervoets and Blust, 2003; Wang et al., 2011). Different species of fish occupy different habitats in the same ecosystem and have different feeding behaviors. As a result, they are used as a good proxy to assess the influence of the environment and biological factors concerning the bioaccumulation of pollutants (Hu et al., 2009; Sullam et al., 2012). Understanding the primary factors influencing bioaccumulation of those compounds in fish from aquatic ecosystems is critical in predicting and assessing risks to upper-trophic level consumers including humans. Although, the levels of dioxin-like PCBs (mono-*ortho* and non-*ortho* PCBs) have recently been reported in the fish (Ssebugere et al., 2013a) and sediments (Ssebugere et al., 2013b) from the Napoleon Gulf, our survey of literature shows no data concerning HCHs and indicator PCBs. The study was aimed at obtaining preliminary information on the occurrence of HCHs and indicator PCBs in sediments and fish from the Napoleon Gulf. The resulting data will be used as a benchmark to compare the concentrations of the contaminants over time.

2. Materials and methods

2.1. Study area

The study area was the Napoleon Gulf which is located on the northern shoreline of Lake Victoria (0° 24′ 34″ N, 33° 14′ 50″ E). It is located south east of Jinja Municipality and covers an area of about 200 km^2. The Gulf supports approximately half a million inhabitants; however in recent years it has been subjected to strong anthropogenic pressures. The major pollution sources are untreated wastewater from sewage systems, industrial plants, waste oil from parking lots and car repair garages. The industrial plants in the vicinity of the Gulf include a water treatment plant and a copper smelting plant. In addition, catchment wetlands which previously played the vital role of tertiary purification of effluent before discharging it into the Gulf have long been encroached on for settlement and degraded.

2.2. Sampling

A total of 24 surface sediments (<30 cm depth) were collected in March, 2011 from the Napoleon Gulf using a sediment corer at four stations: A, B, C and D (Fig. 1). Within each station, 6 sediment samples were randomly taken at distances of approximately 200 m from one another. The number of collected sediment samples was dictated by the high cost of analysis of PCBs. A total of 96 fish (12 of the same species, summing up to 24 fish per station) were also collected from the four stations using gill nets. The fish weights and lengths were measured, and they varied from 520 to 2045 g and 34 to 60 cm, respectively. 10 g of muscle tissues from every four fish of the same species, with approximately the same length and weight were separately pooled. The pooled tissues were homogenized into composite samples using a blender. The fish were pooled to reduce the cost of analysis that would be incurred on the bulky individual samples. The sediments and homogenized fish samples were transferred into acetone rinsed glass bottles and kept at −20 °C prior to extraction.

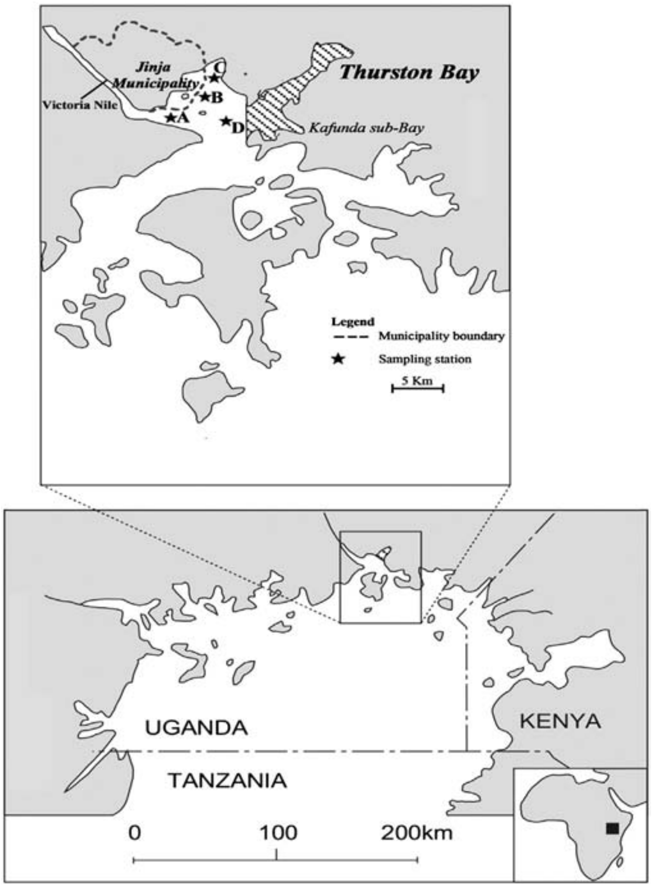

Fig. 1. Map showing the study stations.
Adapted from Campbell et al. (2004).

2.3. Chemical analysis of polychlorinated biphenyls

The analytical procedures used in this study are described in detail by Ssebugere et al. (2013a). Briefly, samples were allowed to thaw and 10 g of each was separately spiked with ^{13}C-labeled PCB surrogate standards (US EPA defined 68A-LCS). The spiked tissues were then mixed with hydromatrix™ (Varian) to remove any moisture. Extraction was performed on an accelerated solvent extraction device (Dionex ASE 200), using a 3:1 *v*/*v* mixture of *n*-hexane/acetone, and the resulting extracts were concentrated to 2 mL using a rotary evaporator. The concentrated extract was divided into two subsamples for the fish (0.3 and 1.7 mL). The 0.3 mL was used for lipid determination by gravimetric methods while the 1.7 mL was passed through a drying funnel of anhydrous sodium sulfate and eluted with 100 mL of *n*-hexane. The resultant mixture was purified using two successive chromatographic steps.

The first clean-up step involved use of a multilayer column containing from bottom to top: 2 g silica gel, 5 g of 33% silica gel-sodium hydroxide, 2 g silica gel, 5 g of 44% silica gel-sulfuric acid, 10 g of 22% silica gel-sulfuric acid and 5 g anhydrous sodium sulfate. The column was pre-washed with 60 mL of *n*-hexane before the extract was added and this was followed by elution with 60 mL of *n*-hexane. The eluate was concentrated using a rotary evaporator and transferred to a carbon column. It was then eluted with 100 mL of *n*-hexane (this was the second clean-up step). The resulting eluate was reduced to <1 mL using a stream of nitrogen. An aliquot of 20 μL nonane and the eluate were quantitatively transferred into vials. The mixture was then evaporated to 25 μL using nitrogen. Internal standards (US EPA defined 68A-IS) were then added to the vials and kept for instrumental analysis. Analysis was conducted using an Agilent model 5890 gas chromatograph series 11 coupled with the high resolution mass spectrometer (GC–HRMS) MAT 95S. All the instrumental conditions were reported in Ssebugere et al. (2013b). Chromatographic separation was achieved on a Phenomenex MultiResidue-2 column (30 m × 0.25 mm i.d. × 0.20 μm).

2.4. Chemical analysis of hexachlorocyclohexanes

The samples were extracted and analyzed as described by Javedankherad et al. (2013). Briefly, the samples were freeze-dried and 5 g was spiked with HCH internal standards. The spiked sample was transferred into an extraction thimble and then extracted in a Soxhlet apparatus for 4 h with 100 mL of 3:1 v/v *n*-hexane/acetone. The resultant extract was concentrated to <2 mL using a rotary evaporator and divided into subsamples for fish (0.3 and 1.7 mL). The 0.3 mL was used for lipid determination, while the 1.7 mL was cleaned up using a column filled from bottom to top with 8 g of acidified silica gel and 2 g of anhydrous sodium sulfate. The column was eluted with 15 mL hexane followed by 10 mL of dichloromethane. The resulting eluate was concentrated under a gentle stream of nitrogen to < 100 μL and kept for analysis. Analysis was performed using a GC equipped with an electron capture detector (ECD) and a DB-5 capillary column (60 m × 0.25 mm i.d. × 0.25 μm). Helium was used as the carrier gas at a flow rate of 2 mL min^{-1}. The operating conditions were split injection mode. The temperature program was as follows: 100 °C (1 min), 10 °C min^{-1} to 240 °C (1 min), 3 °C min^{-1} to 260 °C (1 min), 20 °C min^{-1} to 300 °C (10 min). The injection port temperature and detector temperature were 250 and 300 °C, respectively.

2.5. Quality assurance and data analysis

To ensure the quality of the data, a procedural blank was run after every five samples. The recoveries for six indicator PCB congeners (28, 52, 101, 138, 153 and 180) and three HCH isomers (α, β and γ) in fish varied from 70.3 to 90.4% and 60.3 to 80.1%, respectively. The recoveries for PCBs in sediments were in the range 60.5–97.2%. Limit of detection (LOD) was defined as three times the standard deviation of all analyzed blanks. The LOD was a function of the extracted mass and varied between analytes (0.01–0.32 pg g^{-1} for PCBs and 4–20 pg g^{-1} for HCHs). Treatment of obtained data was done using STATISTICA 6.0 software. During statistical analysis, non-detectable (n.d.) data was assigned a value of zero. 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 Kruskall–Wallis test when more than two groups were considered. In these two non-parametric tests, statistical significance was set at $p < 0.05$.

3. Results and discussion

3.1. Concentrations of PCBs in sediments

The concentrations of the different PCB congeners in sediments from the Napoleon Gulf are presented in Table 1. CB 138 was the predominant congener at all stations (based on mean values it contributed >25 to the ΣPCBs). Barakat et al. (2012) also reported the predominance of CB 138 in sediments from Lake Maryut, Egypt. In the present study, variations in contributions to the ΣPCBs were observed between other congeners (CB 28, 52, 101, 153 and 180) and sampling stations (Fig. 2). Total (Σ) PCBs varied widely between stations, with mean values ranging from 362 to 848 pg g^{-1} dry weight (dw). The levels in this study were 5-fold higher than the ΣPCBs of similar congeners reported by de Souza et al. (2008) in sediments from the Guanabara Bay in Brazil (range, 18–184 pg g^{-1} dw), in the same range as those reported from Congo River Basin in the Democratic Republic of Congo (<50–1400 pg g^{-1} dw; Verhaert et al., 2013) and Istanbul strait in Turkey (13–699 pg g^{-1} dw; Okay et al., 2009), but lower than the ones from Dagu Drainage River in China (9687–22,148 pg g^{-1} dw; Liu et al., 2007) and River Nile (518–4975 pg g^{-1} dw; El-Kady et al., 2007) and Lake Qarun in Egypt (1480–137,200 pg g^{-1} dw; Barakat et al., 2013).

Table 1
Mean concentrations and ranges (pg g^{-1} dry weight) of PCBs in sediments from the Napoleon Gulf.

Congener	Station A	Station B	Station C	Station D
CB 28	151 (135–160)	144 (n.d.–177)	86 (43–121)	72 (n.d.–80)
CB 52	28 (19–40)	63 (n.d.–69)	28 (n.d.–39)	19 (n.d.–21)
CB 101	97 (76–121)	90 (n.d.–100)	34 (n.d.–43)	33 (n.d.–44)
CB 138	170 (124–224)	242 (108–391)	98 (n.d.–117)	162 (135–209)
CB 153	123 (99–140)	155 (78–224)	98 (n.d.–120)	51 (n.d.–60)
CB 180	72 (58–90)	154 (48–434)	38 (27–50)	26 (n.d.–31)
ΣPCBs	640 (538–730)	848 (177–1015)	383 (43–422)	362 (144–356)

The number of samples per station (n) = 6.

Station B had the highest ΣPCB (mean: 848 pg g^{-1}) followed by A (640 pg g^{-1}), while station D showed the least ΣPCB (362 pg g^{-1}). The levels of the individual PCB congeners in sediments from station B were significantly higher ($p < 0.05$) than those from stations C and D. The PCB concentrations for station A were also significantly greater than those at stations C and D. However, statistical analysis between stations A and B showed no significant difference ($p > 0.05$) between the 2 sites. The high PCB levels at stations A and B compared to other locations could be attributed to wastewater runoff from Jinja Municipality which is highly urbanized and industrialized. The use of electrical appliances like transformers and capacitors containing PCB oils was common in Uganda before their ban in the 1970s (National Environment Management Authority, 2007). Some of these appliances are still in use in Jinja Municipality; it is possible that they constitute sources of PCBs through leakage of transformer oils and/or disposal of old electrical equipments. The PCB concentrations at stations C and D were quite uniform, with no significant difference between the 2 sites. Station C is located near Kakira where sugarcane growing is significant. The PCB levels at station C could be due to bio-mass burning of sugarcane which is done almost every harvesting season. Eckhardt et al. (2007) have previously reported bio-mass burning as one of the sources of PCBs in the environment. The low PCB levels at station D compared to other locations (Fig. 3) could be due to its far offshore distance from the point sources.

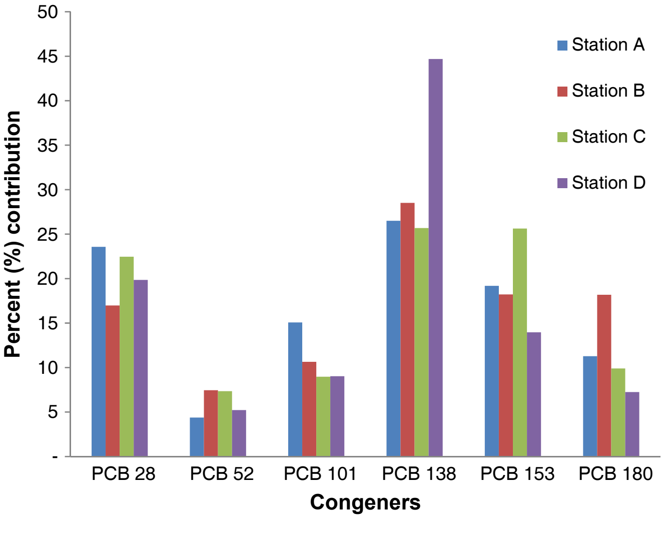

Fig. 2. Percent contribution of the individual congeners to the ΣPCBs in the sediments.

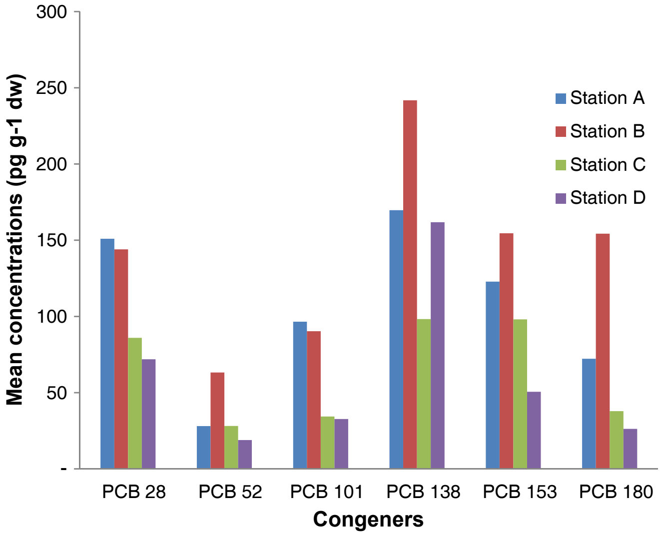

Fig. 3. Mean PCB concentrations in sediments from the different sampling sites.

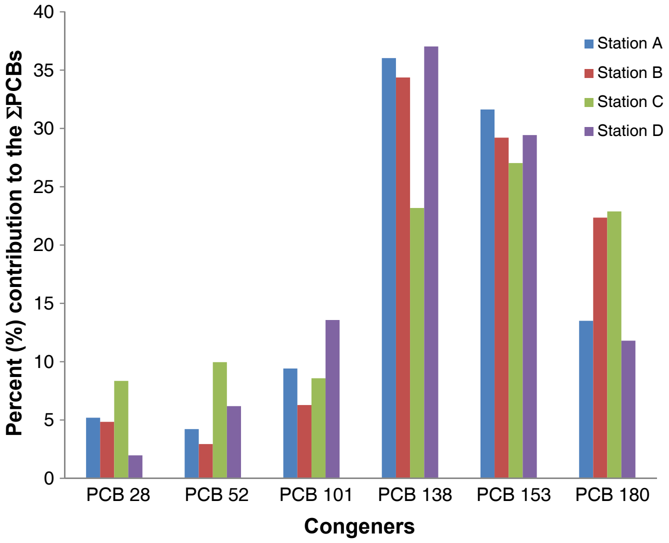

Fig. 4. Percent contribution of the individual congeners to the ΣPCBs in *L. niloticus*.

3.2. Concentrations of PCBs in fish muscle tissues

Muscle tissues were analyzed for PCBs and 63% of them contained detectable levels of the contaminant. The muscle tissues were taken because they are known to bioaccumulate more PCBs than other fish tissues and they evidently serve as an important depot of chemicals (Brázová et al., 2012). Table 2 shows the levels of PCBs on a lipid weight (lw) basis in muscle homogenates of two commercially important fish species (*Lates niloticus* and *Oreochromis niloticus*). CB 138 was the major contributor to the ΣPCBs in *L. niloticus* compared to the other congeners (based on mean values it contributed >33% at majority of the sites (Fig. 4). CB 138 was followed by CB 153 (27–32%) and then CB 180 (≥12–23%). CB 138 and CB 153 were also predominant in *O. niloticus* at all study stations (Fig. 5). Niimi (1996) established that CB 138 and CB 153 are not readily metabolized by certain organisms compared to the rest of the congeners.

The ΣPCBs in *L. niloticus* at the different locations were higher than those in *O. niloticus* (Table 2). The large variability observed in the data could be attributed to differences in feeding habits and trophic levels. While *O. niloticus* is a filter-eating fish species feeding mainly on phytoplankton and zooplankton, *L. niloticus* have predatory feeding behaviors and prefer a diet of live fish (Bwanika et al., 2006; Semyalo et al., 2011). The concentrations of the individual PCB congeners in *L. niloticus* at station A were statistically greater ($p < 0.05$) compared to other sites. However, statistical analysis for the different PCB congeners between stations B and C, stations B and D, and stations C and D showed no significant difference. For *O. niloticus*, statistical analysis showed no significant difference among the study sites.

The mean PCB concentrations in the present study ranged from 41 to 670 pg g^{-1} lw. The concentrations were lower than the data reported in fish from freshwater bodies in Africa such as Lake Tanganyika in Burundi (range, 24,300–77,700 pg g^{-1} lw; Manirakiza et al., 2002) and Lake Burullus in Egypt (3320–72,060 pg g^{-1} lw; Said et al., 2008). The results of the present study were also lower than the data reported in fish from water bodies outside Africa like the Pearl River Delta in China (5150 to 226,000 pg g^{-1} lw; Nie et al., 2006) and Manoa stream in Hawai'i (51,900–89,420 pg g^{-1} lw; Yang et al., 2008). Recently, European Community authorities have set a maximum level of 75,000 pg g^{-1} wet weight for the 6 indicator PCBs in muscle meat of fish and fishery products (EC, 2011). The set value is far over the values found in this study, suggesting that the fish was fit for human consumption with respect to PCBs.

3.3. Levels of hexachlorocyclohexanes in sediments and fish

Three HCH isomers (α, β and γ) were detected in 38% of the fish homogenates. Only the fish species from stations A and C had detectable HCHs, while no residues were found in the sediments at all the study

Table 2
Mean concentrations and ranges (pg g^{-1} lipid weight) of PCBs in fish from the Napoleon Gulf.

Congener	Station A		Station B		Station C		Station D	
	L. niloticus	*O. niloticus*	*L. niloticus*	*O. niloticus*	*L. niloticus*	*O. niloticus*	*L. niloticus*	*O. niloticus*
CB 28	35	n.d.	18	n.d.	18	n.d.	3	n.d.
	n.d.–40		n.d.–20		n.d.–23		n.d.–3	
CB 52	28	16	11	16	21	n.d.	10	n.d.
	n.d.–32	n.d.–18	n.d.–13	n.d.–16	n.d.–28		n.d.–11	
CB 101	63	20	24	18	18	7	21	n.d.
	n.d.–75	15–24	n.d.–30	n.d.–19	n.d.–25	n.d.–10	18–26	
CB 138	241	27	131	27	49	22	58	13
	218–284	21–30	88–195	20–31	22–99	n.d.–34	49–70	n.d.–13
CB 153	212	26	112	27	57	20	46	21
	190–250	24–28	97–136	25–29	18–97	n.d.–22	34–57	20–24
CB 180	90	11	85	9	49	5	19	6
	80–107	6–19	61–99	6–10	12–96	n.d.–5	18–21	4–9
ΣPCBs	670	101	382	97	212	54	157	41
	488–778	85–104	328–430	60–103	52–330	n.d.–65	133–165	25–40

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

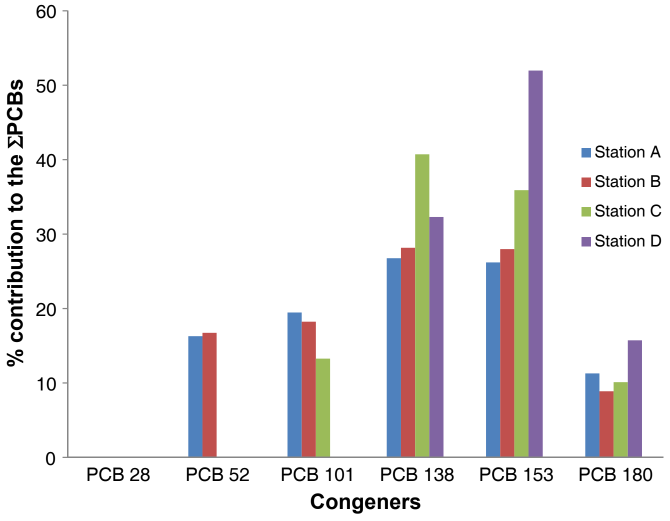

Fig. 5. Contribution of the individual congeners to the ΣPCBs in *O. niloticus*.

stations. The concentrations in *L. niloticus* ranged from 5700 to 26,000, 7000 to 34,000 and 5000 to 13,000 pg g^{-1} lw for α-, β- and γ-HCH isomers, respectively. The maximum concentrations of HCHs in *O. niloticus* were 9000 pg g^{-1} for α-HCH, 8000 pg g^{-1} for β-HCH and 7000 pg g^{-1} for γ-HCH. Regarding the individual contribution to the ΣHCHs, the β isomer accounted for the largest proportion (it contributed >32% in the fish). Kim et al. (2002) reported that β-HCH is more environmentally persistent and has 10–30 times higher ability to accumulate in fatty tissues than other isomers. On the whole, the HCH concentrations at station C were higher than those at station A, although statistical analysis showed no significant differences between the 2 sites.

The ΣHCH values in the present study varied widely with mean values ranging from 14,950 to 45,900 pg g^{-1} (Table 3). The HCH levels were lower than those reported in fish samples from Tana and Sabaki Rivers in Kenya (Lalah et al., 2003) and the Lagos Lagoon in Nigeria (Adeyemi et al., 2008). In Ghana, Darko et al. (2008) reported HCH residues ranging from 700 to 1360 pg g^{-1} lw in fish (*Tilapia zilli*) from Lake Bosomtwi. The results herein were 20 times higher than those by Darko and others. In a related study, Ejobi et al. (2007) reported levels of up to 400 pg g^{-1} lw in *L. niloticus* from Ggaba landing site on the northern shore of Lake Victoria. The levels were lower compared to the ones in the present study. The HCH levels in all the investigated fish were below the extraneous residue limit of 5,000,000 pg g^{-1}, recommended for fish and other fishery products by the Codex Alimentarius Commission of FAO–WHO, 1997. In regard to HCHs, the results from the present study give no indication of health risks associated with the consumption of fish from the Napoleon Gulf.

Table 3
Mean concentrations and ranges (pg g^{-1} lw) of HCH isomers in fish from the Napoleon Gulf.

Isomer	Station A		Station C	
	L. niloticus	*O. niloticus*	*L. niloticus*	*O. niloticus*
α-HCH	10,667	4650	15,233	7250
	9000–13,000	n.d.–4800	5700–26,000	n.d.–9000
β-HCH	8667	5100	19,333	7500
	7000–11,000	n.d.–5500	10,000–34,000	n.d.–8000
γ-HCH	6333	5200	11,333	6500
	5000–7000	n.d.–5800	9000–13,000	n.d.–7000
ΣHCHs	25,667	14,950	45,900	21,250
	22,000–28,000	n.d.–15,800	24,700–73,000	n.d.–24,000
α-/γ-HCH	1.68	0.89	1.34	1.12
	1.43–2.60	0.78–1.04	0.63–2.00	0.92–1.29

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

Ratios of α-/γ-HCH can be used to establish whether degradation of technical HCH is significant or not, and also whether its input occurred in the past or recently. Yi et al. (2013) established that high ratios (α-/γ-HCH) ranging from 4 to 7 indicate fresh input of technical-HCHs, while low ratios (<3) point at historical use of lindane. The ratios of α-/γ-HCH were low (0.63–2.60) suggesting past input of lindane into the Napoleon Gulf.

4. Conclusion

The present study analyzed PCBs and HCHs in sediments and fish species from the Napoleon Gulf of Lake Victoria. The concentration values of the pollutants were lower and in some cases comparable to those reported in literature. However, the presence of PCBs and HCHs in sediments and fish could be harmful since they may find their way into large organisms like humans. Therefore, regulatory controls for monitoring and mitigating wastewater emissions into Lake Victoria need to be implemented and emphasized by the relevant government organs.

Conflict of interest

There is no conflict of interest.

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Paper 1V

Polychlorinated biphenyls in sediments and fish species from the Murchison Bay of Lake Victoria, Uganda

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Polychlorinated biphenyls in sediments and fish species from the Murchison Bay of Lake Victoria, Uganda

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HIGHLIGHTS

- High concentrations of PCBs were found at sites near wastewater discharges.
- The PCB concentrations were low to moderate compared to other locations worldwide.
- Based on the European Commission set TEQ, the fish were fit for human consumption.

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ABSTRACT

Polychlorinated biphenyls (PCBs) were determined in sediments and two fish species collected from the Murchison Bay in Lake Victoria, using high resolution gas chromatography coupled to a high resolution mass spectrometer. Total PCB concentrations (Σ_{18}PCBs) varied widely with mean values ranging from 777 to 4325 pg g^{-1} dry weight (dw) for sediments and 80 to 779 pg g^{-1} wet weight (ww) for fish. The PCB levels in the sediments were significantly higher at the station closest to Nakivubo channel, presumably due to effluents discharged by the channel, which may contain domestically produced commercial PCB mixtures. For fish, the concentrations in Nile perch (*Lates niloticus*) were significantly greater than those in Nile tilapia (*Oreochromis niloticus*) at all study stations, possibly due to dietary differences among species. World Health Organization-toxic equivalents (WHO$_{2005}$-TEQs) for the dioxin-like PCBs were 0.04–0.64 pg g^{-1} dw and 0.01–0.39 pg g^{-1} ww for sediments and fish, respectively. The non-*ortho* PCBs exhibited the highest contribution to the Σ_{12}TEQs (>75%) compared to the mono-*ortho* PCBs in both fish species. The TEQs in the present study were lower than many reported worldwide in literature for fish and were within the permissible level recommended by the European Commission, implying that the fish did not pose health hazards related to PCBs to the consumers.

1. Introduction

Lake Victoria is a vital natural aquatic resource in Uganda, but its ecological health is being threatened, mainly by rapid urbanization and industrialization. These developments coupled with a population of over one million people in its catchment area have resulted in increased pollution (Banadda et al., 2009, 2011a, 2011b; Wasswa et al., 2010). Polychlorinated biphenyls (PCBs) have previously been documented in the lake (Ssebugere et al. 2013a, b; Ssebugere et al., 2014). The PCBs are released into the environment as a result of industrial applications such as capacitors, hydraulic fluids in electronic equipment, and coolants and lubricants in transformers (Gidarakos et al., 2009; Moon et al., 2012; Lundin et al., 2013; Miller et al., 2013).

Exposure to PCBs has been associated with diseases like diabetes mellitus, cancer and hypertension (Lee et al., 2007a). Other adverse effects caused by the pollutants include endocrine disruption, neurobehavioral deficits and fetal toxicity (Lee et al., 2007b; Ha et al., 2007; 2009; Uemura et al., 2009). The PCBs undergo long-range transport causing global pollution (Wiberg et al., 2009; Sundqvist et al., 2009a, b). Furthermore, the contaminants can accumulate in sediments close to point sources and in benthic organisms and fish due to their hydrophobic nature, low metabolic transformation and long half-lives (Suedel et al., 1994; Jönsson et al., 2003; Åberg et al., 2008; 2010; Lavandier et al., 2013). Fish can be an important monitoring tool, because they concentrate PCBs directly from water through their diet, and indicate hazards to piscivorous predators and humans (Van

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der Oost et al., 2003; Huang et al., 2006; Gustavson et al., 2008; Parnell et al., 2008).

To our knowledge whereas PCBs have been reported in fish and sediments from the Napoleon Gulf and Thurston Bay of Lake Victoria (Ssebugere et al., 2013a, b; Ssebugere et al., 2014) and from other water bodies in the world (Manirakiza et al., 2002; Adu-Kumi et al., 2010; Darko et al., 2008; Helm et al., 2008; Eqani et al., 2013; Verhaert et al., 2013), literature shows no work about these pollutants in the Murchison Bay. The Bay is a major hotspot for discharge of industrial outflow, urban runoff and municipal effluents from Kampala, Uganda's capital city (Banadda et al., 2009). The objectives of this study were to investigate the occurrence and contamination levels of PCBs in sediments and fish from the Murchison Bay.

2. Materials and methods

2.1. Study area

The area of study was the Murchison Bay located in the northwestern part of Lake Victoria and southeast of Kampala city (Fig. 1). The Bay covers an area of 60 km^2 and includes semi-enclosed inner and outer parts. The Murchison Bay is the main recipient of sewage effluents, industrial- and municipal-waste from the central district of Kampala City, via the Nakivubo channel. The channel passes through dense residential settlements and commercial areas. In the past, the channel ended in wetland areas, allowing wastewater to be drained into papyrus swamps before entering the Bay. However, in recent years the wetlands which played the role of secondary treatment have been degraded due to settlement and commercial activities (Wasswa, 2009). The water turbidity in the Bay has increased while the water transparency has declined due to excessive waste input.

2.2. Sample collection

A total of 24 surface sediments (<25 cm depth) were taken using a sediment corer from four stations namely 1 and 2 in the inner Bay, and 3 and 4 in the outer Bay (Fig. 1) in March, 2013. Six sediments were randomly taken from each station at distances of approximately 200 m from one another. The geographical coordinates of the inner and outer parts of the Murchison Bay are 00°15′72″N, 32°38′74″E and 00°08′71″N, 32°37′58″E, respectively. Ninety six fish of different species namely: Nile Perch (*Lates niloticus*) a carnivorous top-predator and Nile Tilapia (*Oreochromis niloticus*) a detritivorous species were collected using gill nets within the same locality as the sediments. These two species exhibit different feeding habitats and thus may be exposed differently to contamination through trophic transfer. 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 cm, and 68 ± 5 cm, respectively. The muscle tissues were dissected between the pectoral fin and vent of the fish, minced into pieces and subsamples taken. Subsamples of 4 fish from the same location and, of similar length and species were pooled and homogenized. The sediments and homogenized fish were transferred into acetone rinsed glass bottles and shipped to the Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences where they were freeze-dried.

2.3. Analytical method

Sample extraction and cleanup procedures followed the method reported by Wang et al. (2013). Briefly, 2 g of the freeze-dried sample (fish or sediment) was ground with 10 g of anhydrous sodium sulfate to a free floating powder using a mortar and pestle. The mixture was spiked with ^{13}C-labeled surrogate standards (US EPA defined 68A-LCS) and extracted on an accelerated solvent extractor device (ASE 300, Dionex, USA) using 1:1 v/v mixture of *n*-hexane/dichloromethane (DCM). The resultant extract was concentrated to 2 mL on a rotary evaporator. The concentrated extract was divided into two subsamples for fish (0.3 and 1.7 mL). The 0.3 mL was used for gravimetric determination of the lipid content while the 1.7 mL was kept for clean-up. For the sediments, activated copper granules were added to the extract to remove elemental sulfur before purification with column chromatography. The extracts were then cleaned up with a multilayered silica column (packed from bottom to top with 1 g silica gel, 4 g basic silica gel, 1 g silica gel, 8 g acid silica gel, 2 g silica gel and 2 g anhydrous sodium sulfate), an alumina column (6 g basic alumina and 3 g anhydrous sodium sulfate

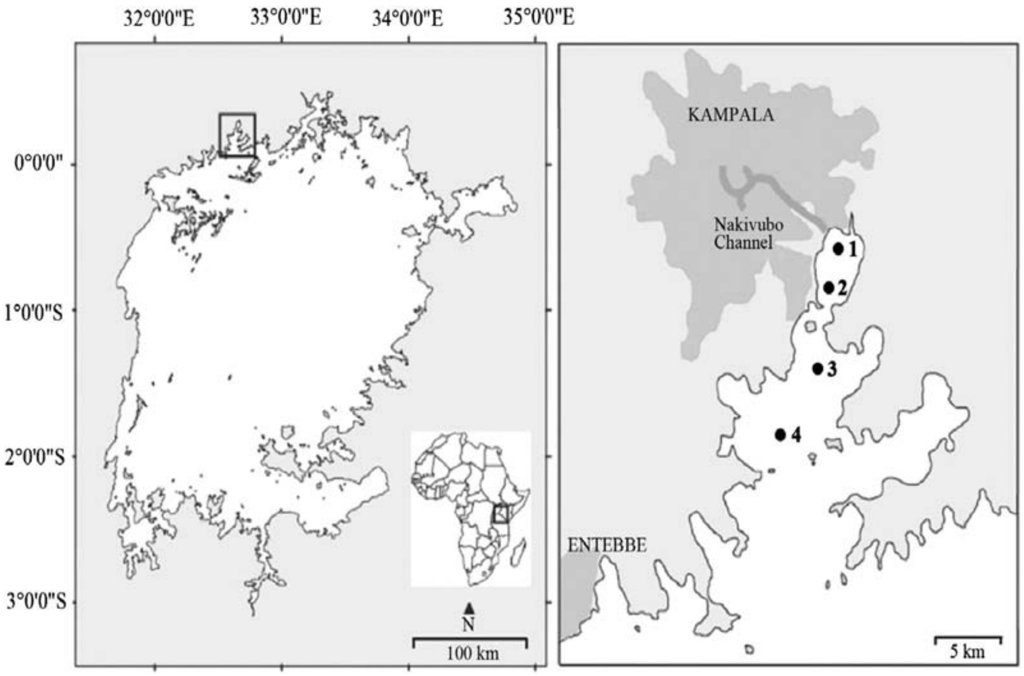

Fig. 1. Map of the Murchison Bay showing the study stations.
Adapted from Haande et al. (2011).

Table 1
Mean concentrations and ranges (pg g^{-1} dry weight) of PCBs in sediments from the Murchison Bay.

Congener	Station 1	Station 2	Station 3	Station 4
CB 28	375 ± 25 353–402	318 ± 30 284–340	103 ± 54 51–158	63 ± 21 45–85
CB 52	507 ± 99 397–589	293 ± 31 259–319	169 ± 14 153–179	108 ± 44 58–137
CB 101	585 ± 234 420–853	343 ± 176 229–546	158 ± 68 105–235	122 ± 41 75–150
CB 138	933 ± 446 631–1445	553 ± 396 230–1009	240 ± 131 151–390	153 ± 87 79–249
CB 153	898 ± 240 721–1171	585 ± 465 251–1115	227 ± 90 173–331	177 ± 85 110–272
CB 180	218 ± 63 152–276	117 ± 54 61–169	49 ± 23 26–72	29 ± 6.1 23–35
Σ indicator-PCBs	3516 2749–4652	2209 1464–3488	946 719–1282	652 409–921
CB 77	82 ± 20 60–99	56 ± 6.3 52–64	12 ± 4.5 6.3–14	8.8 ± 1.2 8.1–10
CB 81	1.9 ± 1.1 0.7–2.8	2.5 ± 0.9 1.6–3.3	1.0 ± 0.6 0.6–1.4	0.8 ± 0.3 0.6–1.0
CB 126	4.9 ± 1.4 3.3–5.8	2.2 ± 0.7 1.7–3.0	1.2 n.d.–1.2	0.9 ± 0.5 n.d.–1.2
CB 169	n.d.	0.3 n.d.–0.3	n.d.	0.1 n.d.–0.1
Σ non-*ortho*-PCBs	89 66–105	61 55–70	14 6.8–17	11 ± 2.0 8.9–13
CB 105	140 ± 70 99–221	59 ± 18 47–79	26 ± 10 19–38	22 ± 13 11–36
CB 114	6.7 ± 3.3 4.4–11	2.9 ± 0.6 2.3–3.4	1.9 ± 0.6 1.5–2.6	1.2 ± 0.6 0.7–1.9
CB 118	434 ± 214 302–682	174 ± 74 139–228	110 ± 59 71–177	72 ± 45 32–120
CB 123	53 ± 19 42–76	24 ± 7.4 18–32	13 ± 5.3 8.7–19	7.4 ± 6.2 1.0–13
CB 156	54 ± 28 36–86	28 ± 14 16–43	14 ± 5.1 9.2–19	7.5 ± 3.3 4.9–11
CB 157	7.7 ± 5.2 4.2–14	3.4 ± 1.3 2.2–4.8	2.0 ± 1.0 1.3–3.1	1.3 ± 0.7 0.8–2.1
CB 167	16 ± 7.9 11–26	8.9 ± 5.0 4.6–14	4.4 ± 1.1 3.1–5.2	1.9 ± 0.4 1.5–2.2
CB 189	8.0 ± 1.9 6.6–10	5.5 ± 2.7 3.0–8.4	2.7 ± 1.3 1.3–3.9	1.1 ± 0.4 0.8–1.6
Σ mono-*ortho*-PCBs	720 508–1124	306 231–413	174 124–266	114 53–189
Σ_{12}dl-PCBs	809 613–1190	367 289–482	188 131–283	125 62–202
Σ_{18}PCBs	4325 3762–5842	2576 1753–3970	1134 850–1365	777 471–1123
WHO$_{2005}$-TEQs	0.55 0.37–0.64	0.25 0.19–0.33	0.11 0.04–0.12	0.10 0.07–0.14

The number of sediment samples per station (N) = 6; n.d. — non-detectable.

at the top) and carbon column (1.5 g of 18% carbon dispersed in cilite, 3 g anhydrous sodium sulfate at the top), respectively. The clean eluate was concentrated to 1 mL using a rotary evaporator and transferred into vials containing 20 μL nonane. The contents in the vials were further dried to 25 μL using a gentle stream of nitrogen and later spiked with injection standards (US EPA defined 68A-IS) prior to analysis.

2.4. Instrumental analysis

The analysis of 18 PCB congeners including 6 indicator PCBs (IUPAC numbers 28, 52, 101, 138, 153 and 180) and twelve dioxin-like PCBs (IUPAC numbers 77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 167 and 189) was done using high-resolution gas chromatography coupled with high-resolution mass spectrometry (HRGC/HRMS) (AutoSpec Ultima, Waters, USA). Chromatographic separation was achieved by injecting 1 μL of sample on a fused silica capillary column (DB5MS, 60 m × 0.25 mm i.d. × 0.25 μm film thicknesses). The oven temperature of the GC was held at 120 °C for 1 min, then increased to 150 °C at a rate of 30 °C min^{-1} and finally to 300 °C at 2.5 °C min^{-1}. Helium was used as a carrier gas at a flow rate of 1.0 mL min^{-1}. The HRMS was operated in VSIR mode at a resolution of ≥ 10000 under positive electron ionization conditions (35 eV) and a source temperature of 270 °C.

2.5. Quality assurance and quality control

To ensure the quality of data, surrogate PCB standards, blanks, replicates and a certified reference material (CRM) were included in the analysis. Recoveries for majority of the surrogate standards met the requirements of US EPA methods 1668A (were in the range of 75–127%) except for CBs 28, 118, and 138 whose recoveries were $<75\%$ (range: 56–74%). The recoveries resulting from triplicate determinations (N = 3) of the certified reference material were between 82% and 106%. The limit of detection (LOD) which was calculated as three times the signal-to-noise ratio, varied from 0.04 to 0.96 pg g^{-1} dry weight (dw) for sediments and 0.02 to 0.80 pg g^{-1} wet weight (ww) for fish. PCBs in the field and procedural blanks were detected at $<15\%$ of the concentration in each batch of 12 samples, so the reported data were not blank corrected.

2.6. Statistical data analysis

Statistical analysis was performed using Statistica® 7 for Windows. Kolmogorov–Smirnov test was used for verifying the normality of data. Since concentration values were not normally distributed, non-parametric statistics was applied. The Mann–Whitney *U*-test was used for pair wise comparison and the Kruskal–Wallis test when more than two groups were considered. In these two non-parametric tests, statistical significance was set at $p < 0.05$. In the present study, toxic

Table 2
Comparison of sediment PCB concentrations (pg g^{-1} dry weight) in this study with those from other locations.

Area	PCBs (pg g^{-1} dry weight)	References
Napoleon Gulf and Thurston Bay of Lake Victoria, Uganda	64–1262 (18 congeners)	Ssebugere et al. (2013b) and Ssebugere et al. (2014)
Nile River, Egypt	1461–2244 (18 congeners)	El-Kady et al. (2007)
Pangani River basin, Tanzania	357–11000 (28 congeners)	Hellar-Kihampa et al. (2013)
Congo River basin, Democratic Republic of Congo	n.d.–1400 (33 congeners)	Verhaert et al. (2013)
Ghar El Melh lagoon, Tunisia	n.d.–3987 (20 congeners)	Ameur et al. (2011)
Mekong River delta, Vietnam	110–2000 (13 congeners)	Carvalho et al. (2008)
Riverine and coastal waters of Surabaya, Indonesia	n.d.–420000 (62 congeners)	Ilyas et al. (2011)
Scheldt River, Belgium	n.d.–200000 (27 congeners)	Covaci et al. (2005)
Remote lakes and coastal areas, Norwegian Arctic	180–13000 (15 congeners)	Jiao et al. (2009)
Mersey Estuary, United Kingdom	36000–1409000 (7 congeners)	Vane et al. (2007)
Lake Maggiore, Italy and Switzerland	n.d.–3000 (12 congeners)	Vives et al. (2007)
Besòs River, Barcelona	6–14 (30 congeners)	Castells et al. (2008)
Indiana Harbor and Ship Canal, Lake Michigan, United States of America	53000–35000000 (163 congeners)	Martinez et al. (2010)
Sea Lots, Port of Spain, Trinidad and Tobago	62000–601000 (136 congeners)	Mohammed et al. (2011)

n.d. — non-detectable.

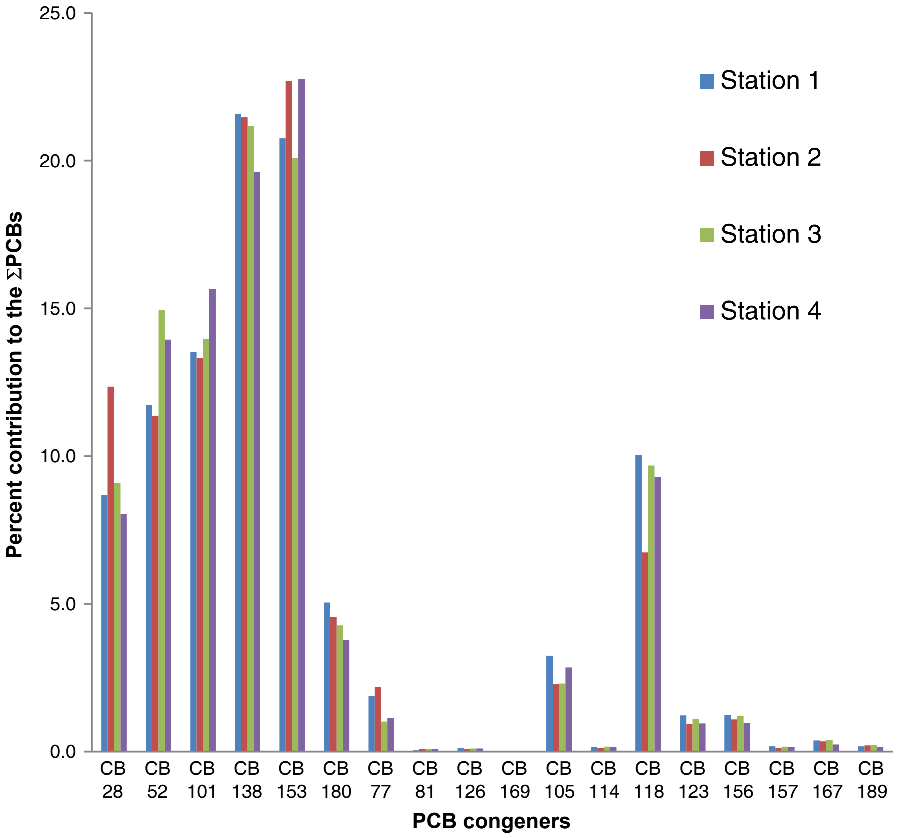

Fig. 2. Percent contribution of the congeners to the Σ_{18}PCBs per station.

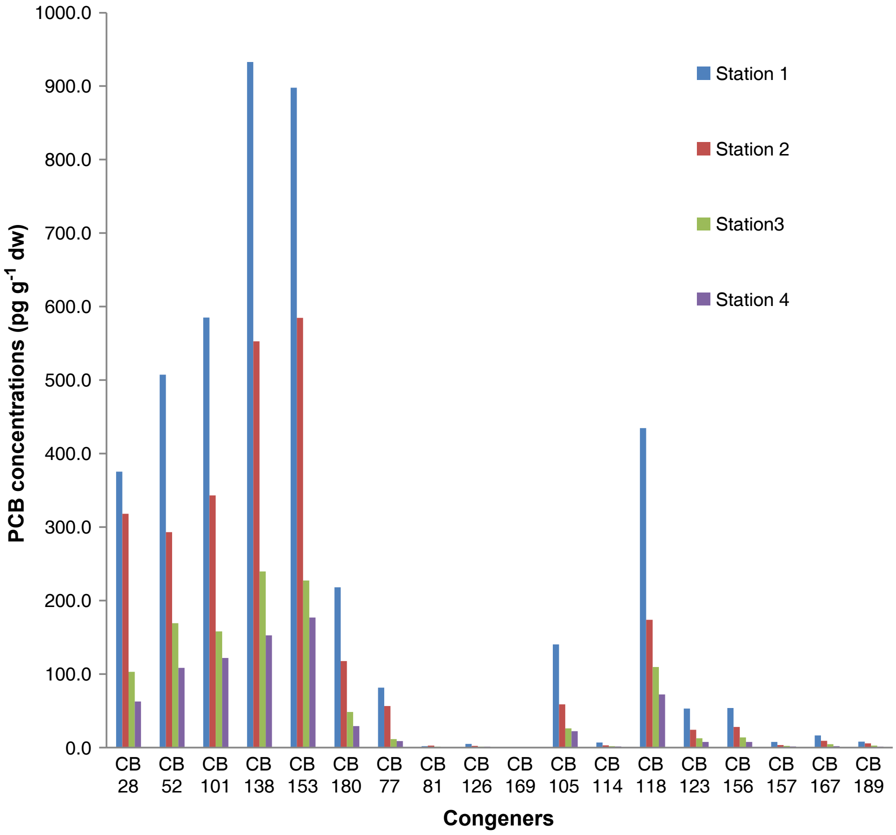

Fig. 3. Mean concentrations of the PCB congeners in the sediments per station.

equivalents (TEQs) for the 12 dioxin-like (dl) PCBs were calculated using individual congener concentrations and the 2005 toxic equivalency factors (TEFs) published by the World Health Organization (WHO) (van den Berg et al., 2006).

3. Results and discussion

3.1. Concentrations of PCBs in sediments from the Murchison Bay

Total PCB concentration (Σ_{18}PCBs) in Murchison Bay sediments varied widely with mean values ranging from 777 to 4325 pg g^{-1} dw (Table 1). They were 3-fold higher than those reported in sediments from the Napoleon Gulf and Thurston Bay of Lake Victoria (Ssebugere et al., 2013b; Ssebugere et al., 2014). The PCB concentrations in the present study were comparable to data from other fresh water bodies in Africa: the Nile River in Egypt (El-Kady et al., 2007), Ghar El Melh lagoon in Tunisia (Ameur et al., 2011), Pangani River basin in Tanzania (Hellar-Kihampa et al., 2013) and Congo River basin in the Democratic Republic of Congo (Verhaert et al., 2013). The results were also in the range of data found in sediments from tropical areas in Asia: the Mekong River delta in Vietnam (Carvalho et al., 2008) and riverine and coastal waters of Surabaya in Indonesia (Ilyas et al., 2011). The levels were lower compared to concentrations reported for industrialized areas in Europe and United States of America (Covaci et al., 2005; Vane et al., 2007; Vives et al., 2007; Castells et al., 2008; Jiao et al., 2009; Martinez et al., 2010; Mohammed et al., 2011) (Table 2).

The proportion of the 6 indicator PCBs (Σ_6PCBs) to Σ_{18}PCBs in this study were between 80 and 88% (with a mean value of 84%). CB 138 and 153 were the predominant congeners, collectively accounting for 21% of the Σ_{18}PCBs on average (Fig. 2). Barakat et al. (2012) also reported the predominance of CB 138 and CB 153 in surface sediments from Lake Maryut in Egypt. The accumulation of PCBs in the environment is directly linked to their degree of chlorination, stereochemistry and lipophilicity (Mazet et al., 2005). PCBs with a high degree of chlorination

Table 3
Mean concentrations and ranges (pg g^{-1} wet weight) of PCBs in fish species from the Murchison Bay.

	Station 1		Station 2		Station 3		Station 4	
	L. niloticus	*O. niloticus*	*L. niloticus*	*O. niloticus*	*L. niloticus*	*O. niloticus*	*L. niloticus*	*O. niloticus*
Lipid content (%)	2.8	1.5	2.3	1.4	2.2	1.1	1.6	1.2
Congener								
CB 28	38 ± 22	14 ± 7.0	62 ± 21	13 ± 3.6	23 ± 7.5	32 ± 14	26 ± 15	9.3 ± 0.7
	13–53	6.0–19	40–83	8.4–15	18–31	12–68	10–41	8.6–10
CB 52	39 ± 23	30 ± 24	60 ± 52	22 ± 18	19 ± 8.2	13 ± 4.6	12 ± 3.2	8.1 ± 2.1
	23–65	7.8–55	10–128	9.0–43	10–27	10–18	9.6–16	5.6–9.5
CB 101	58 ± 11	23 ± 18	86 ± 39	17 ± 4.9	50 ± 25	13 ± 2.6	21 ± 6.5	9.8
	45–66	2.1–37	50–128	12–22	22–65	10–15	16–28	4.9–14
CB 138	183 ± 98	43 ± 9.9	84 ± 38	17 ± 13	61 ± 31	10 ± 1.3	62 ± 32	18 ± 13
	72–259	32–51	52–127	7.7–32	25–81	9.8–12	33–96	5.9–31
CB 153	220 ± 58	37 ± 13	98 ± 55	22 ± 14	76 ± 43	12 ± 2.1	72 ± 15	18 ± 12
	75–386	22–49	51–158	13–39	30–112	10–14	37–133	4.9–30
CB 180	98 ± 57	12 ± 10	8.3 ± 2.2	3.8 ± 2.5	13 ± 6.7	3.2 ± 1.4	11 ± 6.7	4.9 ± 3.0
	37–151	5.1–24	n.d.–9.8	1.1–7.7	8–21	1.7–4.5	6.0–18	1.5–7.3
Σ indicator-PCBs	636	159	398	95	242	83	204	68
	363–904	75–228	288–573	57–124	144–307	60–122	112–322	32–100
CB 77	5.5 ± 1.7	3.1 ± 2.5	n.d.	1.0 ± 0.4	2.2 ± 0.9	1.3 ± 0.2	1.3 ± 0.1	0.7
	3.8–7.1	1.2–5.9		0.6–1.4	1.3–3.1	1.0–1.4	n.d.–1.4	n.d.–0.7
CB 81	0.9	1.1 ± 0.6	n.d.	n.d.	0.9 ± 0.2	n.d.	0.5 ± 0.4	1.1 ± 0.1
	n.d.–0.9	n.d.–1.5			n.d.–1.5		n.d.–0.8	n.d.–1.1
CB 126	1.0 ± 0.7	n.d.	0.2	n.d.	n.d.	n.d.	0.8	n.d.
	0.5–1.5		n.d.–0.2				n.d.–0.8	
CB 169	n.d.	0.2	n.d.	n.d	n.d.	n.d.	0.2	0.1
		n.d.–0.2					n.d.–0.2	n.d.–0.1
Σ non-*ortho* PCBs	7.4	4.4	0.2	1.0	3.1	1.3	2.8	1.9
	5.7–7.6	2.4–6.6	n.d.–0.2	0.6–1.4	2.4–3.1	1.0–1.4	n.d.–3.2	0.7–1.1
CB 105	23 ± 11	7.8 ± 1.8	1.8 ± 0.4	1.9 ± 0.3	3.6 ± 0.1	1.8 ± 0.8	6.7 ± 5.3	1.8 ± 0.3
	13–34	5.8–8.9	1.4–2.2	1.7–2.2	3.5–3.7	1.0–2.6	1.5–12	1.4–2.1
CB 114	1.5 ± 0.8	1.6 ± 0.7	4.2	n.d.	0.2 ± 0.1	0.2	0.7 ± 0.2	0.3
	0.8–2.4	0.5–3.6	n.d.–4.2		n.d.–0.2	n.d.–0.2	n.d.–0.9	n.d.–0.3
CB 118	80 ± 47	47 ± 13	23 ± 16	6.5 ± 1.2	17 ± 3.1	6.0 ± 1.6	25 ± 19	5.5 ± 1.9
	35–128	19–97	14–41	5.5–7.8	13–19	4.2–7.2	7.5–46	3.3–6.8
CB 123	14 ± 8.7	5.9 ± 2.2	3.3 ± 2.3	0.8 ± 0.3	2.3 ± 0.5	0.7 ± 0.3	4.3 ± 3.9	0.7
	4.4–22	2.7–12	1.4–5.8	n.d.–1.0	1.8–2.7	0.3–0.9	0.8–8.5	0.4–1.0
CB 156	8.5 ± 4.7	2.3 ± 0.2	2.8 ± 2.1	0.8 ± 0.5	2.3 ± 0.8	0.6 ± 0.1	3.3 ± 2.4	0.7 ± 0.3
	3.3–12	2.1–2.5	1.0–5.4	0.5–1.4	1.5–3.0	n.d.–0.6	1.9–6.1	n.d.–1.0
CB 157	1.7 ± 1.3	0.4 ± 0.1	n.d	0.1	1.8 ± 0.4	0.1	1.0 ± 0.6	0.6
	0.5–3.0	0.3–0.4		n.d.–0.1	0.3–2.9	n.d.–0.1	n.d.–1.4	n.d.–1.1
CB 167	5.1 ± 3.7	0.7 ± 0.2	2.8 ± 1.7	0.3 ± 0.2	1.2 ± 0.4	0.3 ± 0.1	1.6 ± 1.2	0.6
	1.3–8.6	0.6–0.9	1.2–5.9	n.d.–0.5	0.5–2.0	n.d.–0.4	0.4–2.8	n.d.–0.6
CB 189	2.5 ± 0.3	1.0 ± 0.7	3.1	n.d.	0.5	0.1	1.4	n.d.
	n.d.–2.8	n.d.–1.5	n.d.–3.1		n.d.–0.5	n.d.–0.1	n.d.–1.4	
Σ mono-*ortho* PCBs	136	66	41	10	29	9.7	44	10
	58–213	34–123	20–67	7.8–12	21–34	6.1–12	12–80	5.9–12
Σ_{12}dl-PCBs	143	71	41	11	32	11	47	12
	66–219	37–126	20–67	9.0–13	24–37	7.1–11	12–83	7.0–13
Σ_{18}PCBs	779	229	439	106	274	93	251	80
	429–1123	201–275	308–641	66–137	168–343	72–135	124–404	39–111
WHO_{2005}-TEQs	0.16	0.03	0.15	0.06	0.10	0.04	0.07	0.06
	0.07–0.22	0.02–0.03	0.01–0.39	0.01–0.15	0.03–0.20	0.02–0.07	0.04–0.09	0.02–0.11

The number of fish muscle homogenates per species per station (N) = 3; n.d. — non-detectable.

(such as CB 138 and CB 153) have lower chemical degradation rates than the less chlorinated PCBs and thus are retained in aquatic systems to a greater degree, where they may bioaccumulate in organisms like fish (Lavandier et al., 2013).

The PCB levels at station 1 were significantly higher ($p < 0.05$, Mann–Whitney *U*-test) than those from stations 3 and 4. However, statistical analysis between stations 1 and 2 showed no significant difference between the two locations. Station 1 is located near the mouth of the Nakivubo channel which brings in effluents from a number of chemical manufacturing factories, industrial waste treatment plants and municipal solid waste incinerators. Furthermore, in close vicinity of the station 1 are industrial facilities and Port bell where ships anchor on their way to and from Ports like Mwanza in Tanzania, Kisumu in Kenya and Jinja Port in Uganda. Although, the shipping industry promotes economic development in the area, it inevitably brings pollution related problems.

High concentrations of PCBs were also observed at station 2 (mean value of the Σ_{18}PCBs in sediments was 2576, range 1753–3970 pg g^{-1} dw). Station 2 is situated close to the Ggaba landing site and a local market. Besides the market a lot of commercial, domestic and industrial activities occur near the station and could be releasing domestically-produced commercial PCB mixtures into the lake. The PCB levels in sediments from the outer part of the Murchison Bay (stations 3 and 4), both far from industrial complexes, were an order of magnitude lower compared to those in the inner Bay (Fig. 3). The low PCB levels in the outer Murchison Bay are likely due to the far offshore distance from the pollution sources.

The WHO_{2005}-TEQs for the 12 dl-PCBs in sediments of the present study varied from 0.04 to 0.64 pg TEQ g^{-1}. Station 1 had the highest TEQ values ranging from 0.37 to 0.64 pg TEQ g^{-1} (mean 0.55 pg TEQ g^{-1}) while station 4 had the lowest (range 0.07 to 0.14, mean 0.10 pg TEQ g^{-1}). CB 126 was the highest contributor to the Σ_{12}TEQs (70–89%) compared to other congeners at all locations. The WHO_{2005}-TEQs in most samples from the Murchison Bay were below the interim sediment quality guideline of 0.85 pg TEQ g^{-1} dw recommended by the Canadian Council of Ministers of the Environment (2002). Thus the dl-PCB concentrations in the present work are unlikely to pose a threat to benthic organisms.

3.2. Concentrations of PCBs in fish from the Murchison Bay

Table 3 presents the PCB concentrations in fish muscle homogenates of *L. niloticus* and *O. niloticus*. Average concentrations of the Σ_{18}PCBs ranged from 80 to 779 pg g^{-1} wet weight (ww). The concentrations were in the same range as those reported in similar fish species from the Napoleon Gulf of Lake Victoria (94–716 pg g^{-1} ww: Ssebugere et al., 2013a; Ssebugere et al., 2014) and River Nile in Egypt (695–853 pg^{-1} ww; El-Kady et al., 2007). In neighboring Burundi, Manirakiza et al. (2002) reported PCB concentrations between 44 900 and 166 700 pg g^{-1} lw in *O. niloticus* collected from Lake Tanganyika.

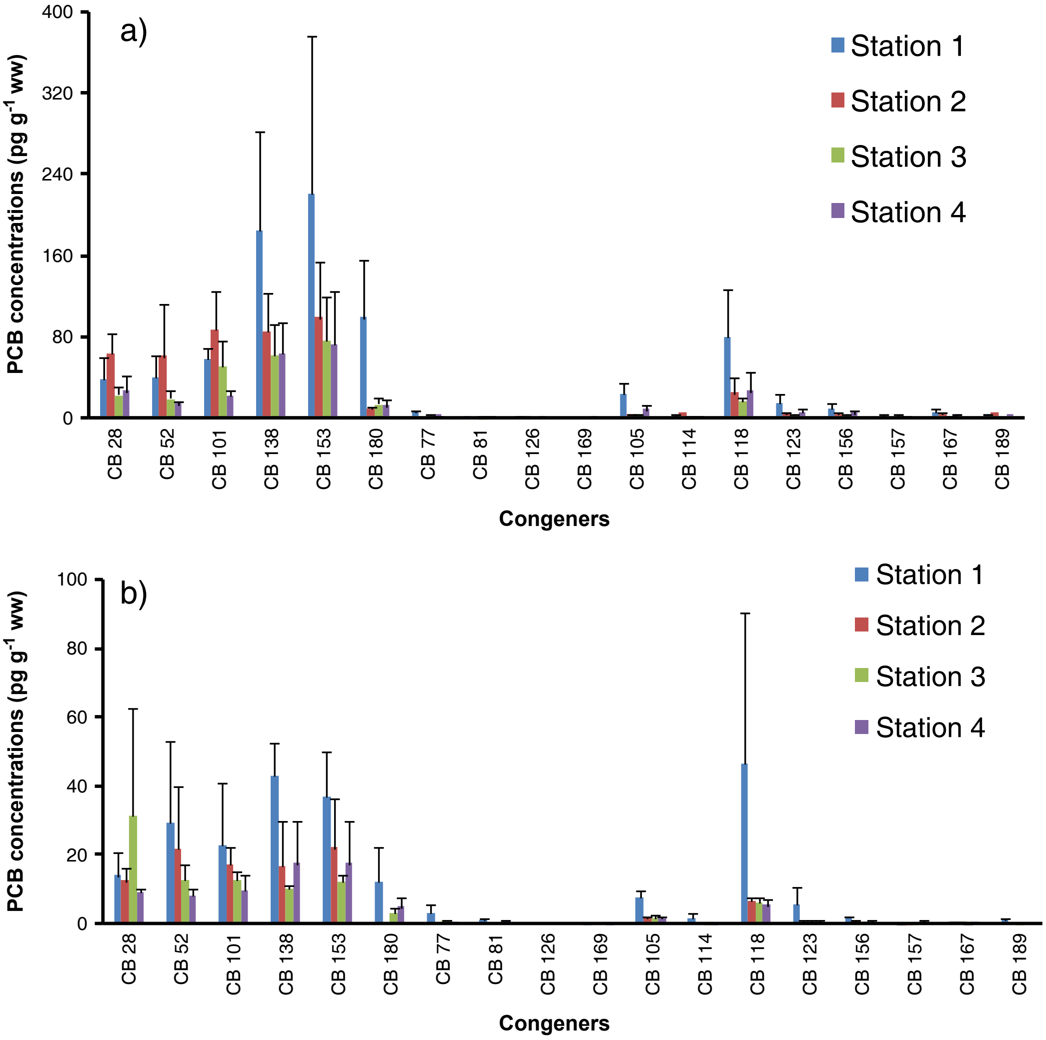

Fig. 4. Mean PCB concentrations in fish per station. a) Mean PCB concentrations in *L. niloticus* per station. b) Mean PCB concentrations in *O. niloticus* per station.

These levels were at least an order of magnitude higher compared to the present results. In Egypt, Said et al. (2008) reported PCB concentrations varying from 3320 to 72060 pg g^{-1} lw in fish (*O. niloticus* and *Clarias gariepinus*) from Lake Burullus. The concentrations were again higher than the data in the study herein. Studies outside Africa have also reported PCBs in fish. In Hawai'i, PCB levels ranging from 51900 to 89420 pg g^{-1} lw were reported in tilapia (*Oreochromis mossambicus*) collected from the Manoa stream and Ala Wai Canal of O'ahu (Yang et al., 2008). The PCB levels were also higher than those reported in this study. In South China, Nie et al. (2006) reported PCB concentrations ranging from 5150 to 226000 pg g^{-1} lw in tilapia (*Tilapia mossambica*) from Pearl River Delta. The concentrations are still higher compared to the data in the current study.

The Σ_{18}PCBs in *L. niloticus* at station 1 was one-fold higher than that at station 2 and about three times higher than that at station 4. The Σ_{18}PCBs at stations 3 and 4 were quite similar. For *O. niloticus*, still station 1 showed the highest levels of the Σ_{18}PCBs (mean value 229 pg g^{-1}) compared to other stations. Station 1 was followed by station 2 and then station 3, while station 4 had the lowest concentration. The concentrations of the different PCB congeners at the different stations for the 2 fish species are shown in Fig. 4.

The PCB concentrations in *L. niloticus* were markedly higher ($p < 0.05$, Mann–Whitney *U*-test) than those in *O. niloticus* at all study sites. The nature of the diet could be a possible explanation for such an inter-specific differences in concentrations. *L. niloticus* are exclusively piscivorous, consuming all available fish species including its own siblings whereas *O. niloticus* relies on zooplankton and macro-invertebrates (Ogari, 1984; Njiru et al., 2004). Earlier studies established that PCBs can have great variations among different fish, as well as within species depending on their feeding habits and trophic levels (Suedel et al., 1994; Vander Zanden and Rasmussen, 1996; Davis et al., 2002; van der Oost et al., 2003).

On the whole, the Σ_6 indicator PCBs in fish of the present study were an order of magnitude higher than the Σ_7 mono-*ortho*-PCBs and Σ_4 non-*ortho*-PCBs. However, the Σ_6 indicator PCB levels (83–636 pg g^{-1}) in this study were within the limit of 75000 pg g^{-1} ww set for fish by the European Commission (EC, 2011), implying that the study fish were fit for human consumption in regard to indicator PCBs. Indicator PCB congeners such as CB 138 and CB 153 which were predominant in the sediments were also major contributors in *L. niloticus* (Fig. 5). However, variations in contribution of the indicator PCBs to the Σ_{18}PCBs were observed for *L. niloticus* at the different stations. For the coplanar PCBs, the mono-*ortho* PCBs were the predominant congeners in most fish (their contribution to the Σ_{12}PCBs ranged from 84 to 100%). CB 118 was the most abundant congener, accounting for 46–69% of the Σ_{12}PCBs, in comparison to other coplanar compounds. The high levels of CB 118 in the two fish species could be due to the inability of the species to metabolize the congener after accumulation. The presence of CB 118 in our samples suggests paint additives, municipal waste plants, iron ore sintering plants or medical waste incinerators as potential sources of PCBs (Abad et al., 2006; Aries et al., 2006; Shin et al., 2006; Chen et al., 2009; Jartun et al., 2009). Furthermore, it is likely that the

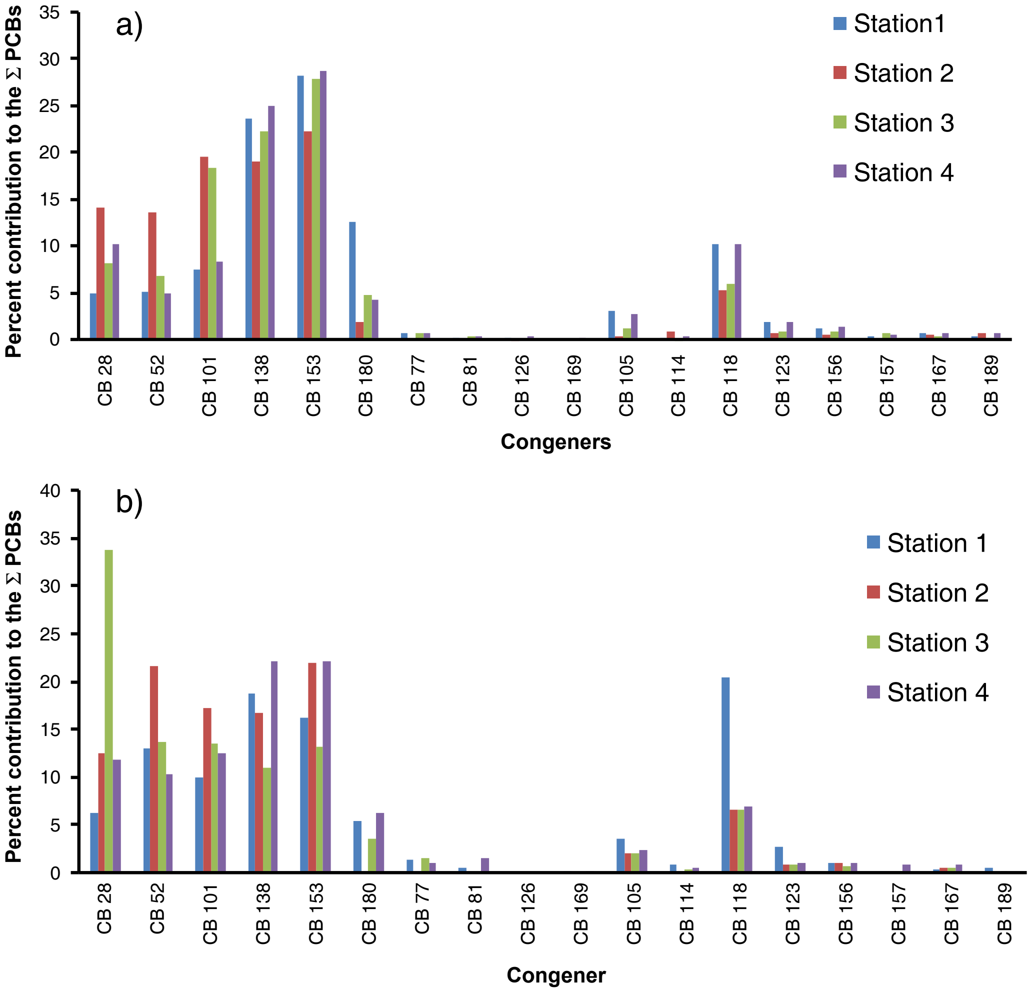

Fig. 5. Percent contribution of the congeners to the ΣPCBs in the fish. a) Contribution of the different congeners to the Σ_{18}PCBs in *L. niloticus*. b) Contribution of the different congeners to the Σ_{18}PCBs in *O. niloticus*.

PCBs came from the residues of former applications like transformers, electrical equipment and other industrial uses. The use of these applications containing PCBs was common in Uganda before their commercial production was banned worldwide in 1979 (National Environment Management Authority, 2007).

The WHO_{2005}-TEQs for dl-PCBs in the study herein were 0.01–0.39 and 0.01–0.11 pg g^{-1} ww for *L. niloticus* and *O. niloticus*, respectively. The non-*ortho* PCBs exhibited the highest contribution to the Σ_{12}PCBs (>75%) compared to the mono-*ortho* PCBs in both fish species. A similar observation was also reported by Ssebugere et al. (2013a) in fish from the Napoleon Gulf and Thurston Bay of Lake Victoria. The WHO_{2005}-TEQs in the present study were within the permissible level of 3.5 pg g^{-1} ww set by the European Commission (EC, 2011), suggesting that the fish from the Murchison Bay were safe for consumption with respect to dl-PCBs.

4. Conclusions

To our knowledge this is the first study to record PCB concentrations in sediments and fish from the Murchison Bay. The station closest to Nakivubo channel had the highest PCB concentrations compared to other locations. Land based activities such as urban centers, commercial and industrial establishments are likely the major sources of PCB contamination. The PCB concentrations in the present study were generally comparable to those of most studies in Africa but lower than the data examined elsewhere in the world. The WHO-TEQs were below the recommended values set for sediments and fish, indicating that the risk posed by PCBs is limited, compared to other health and environmental issues, such as the high disease burden due to diarrhea and other infectious diseases, associated with a limited availability of sanitation and treated water (Yeka et al., 2012; Tumwebaze et al., 2013; McElligott et al., 2013; Bwire et al. 2013; Katukiza et al., 2014). However, regulatory controls for monitoring and mitigating wastewater emissions into Lake Victoria need to be implemented and emphasized by the relevant governmental agencies in Uganda.

Conflict of interest

There is no conflict of interest.

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Paper V

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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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

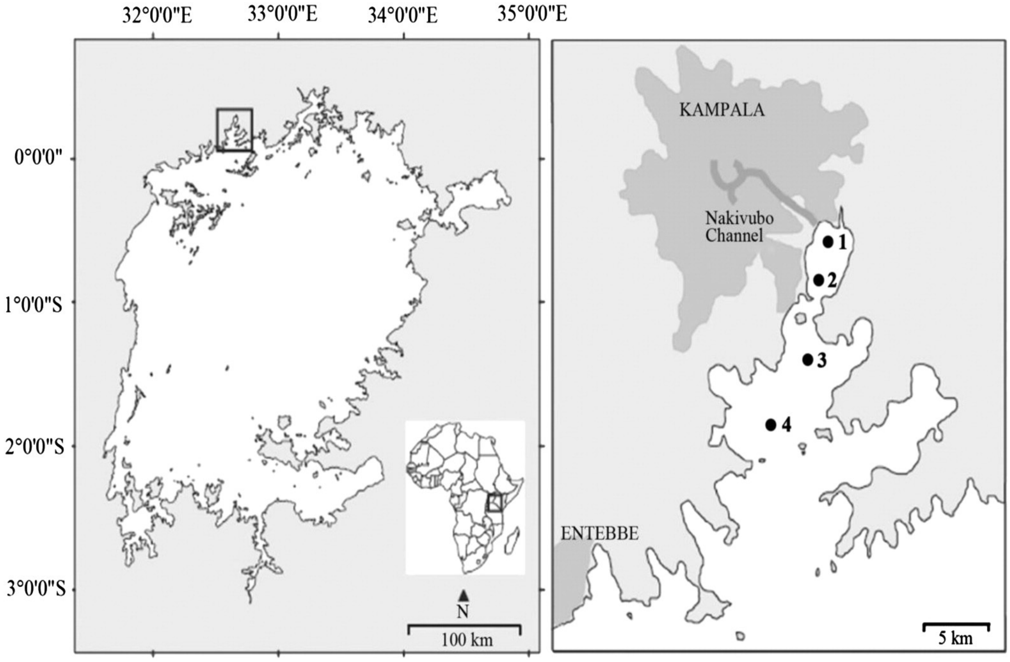

Fig. 1. Geographical location of the Murchison Bay.
Adapted from Haande et al. (2011).

(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

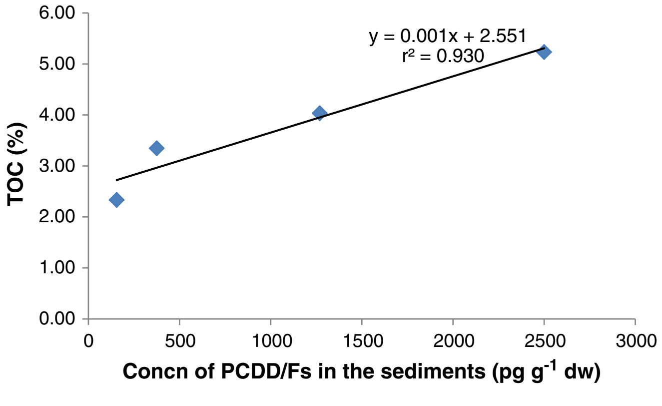

Fig. 2. Correlation between the sediment PCDD/F concentrations and percent TOC.

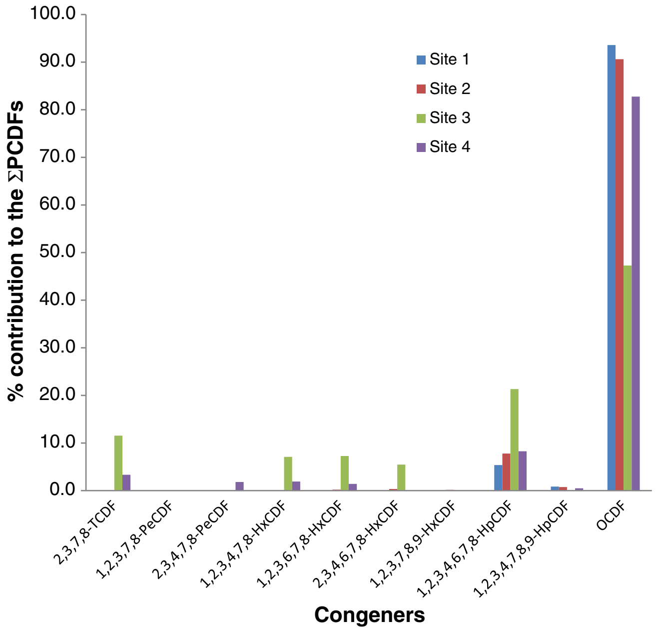

Fig. 3. Percent contribution of the congeners to the ΣPCDFs.

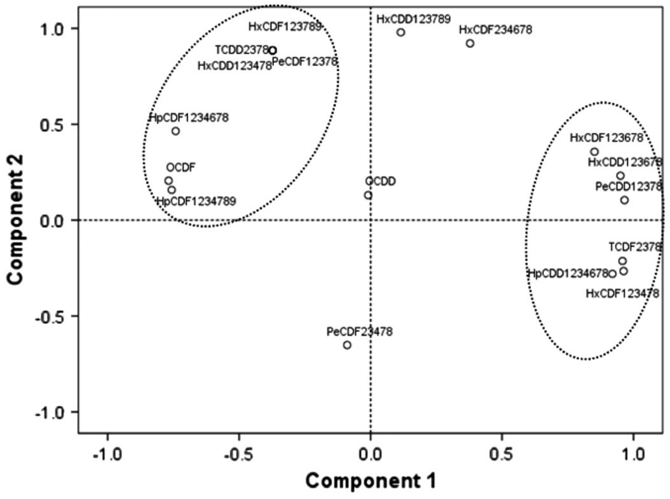

Fig. 4. PCA plot corresponding the PCDD/F concentrations to the known sources in literature.

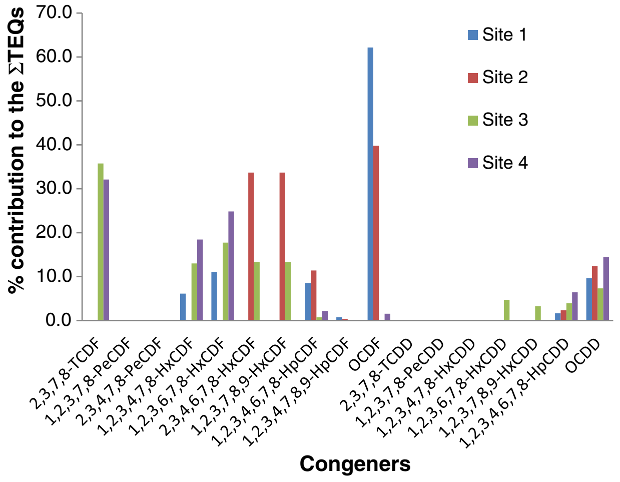

Fig. 5. Percent contribution of PCDD/F congeners to the ΣTEQs.

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

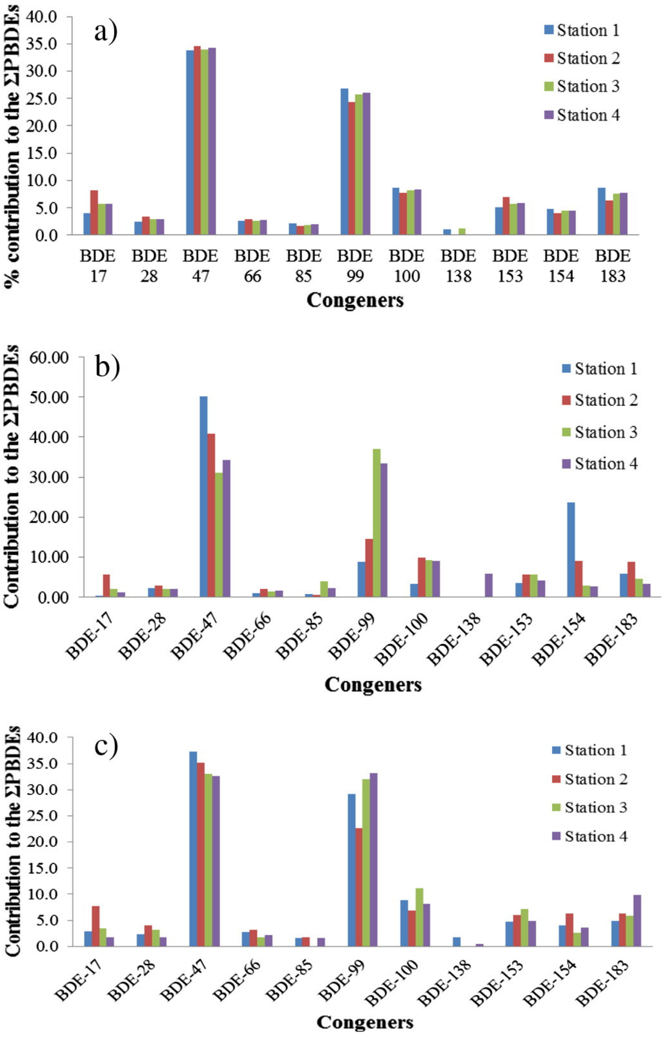

Fig. 6. Contribution of the different congeners to the ΣPBDEs. a) In sediments. b) In *L. niloticus*. c) In *O. niloticus*.

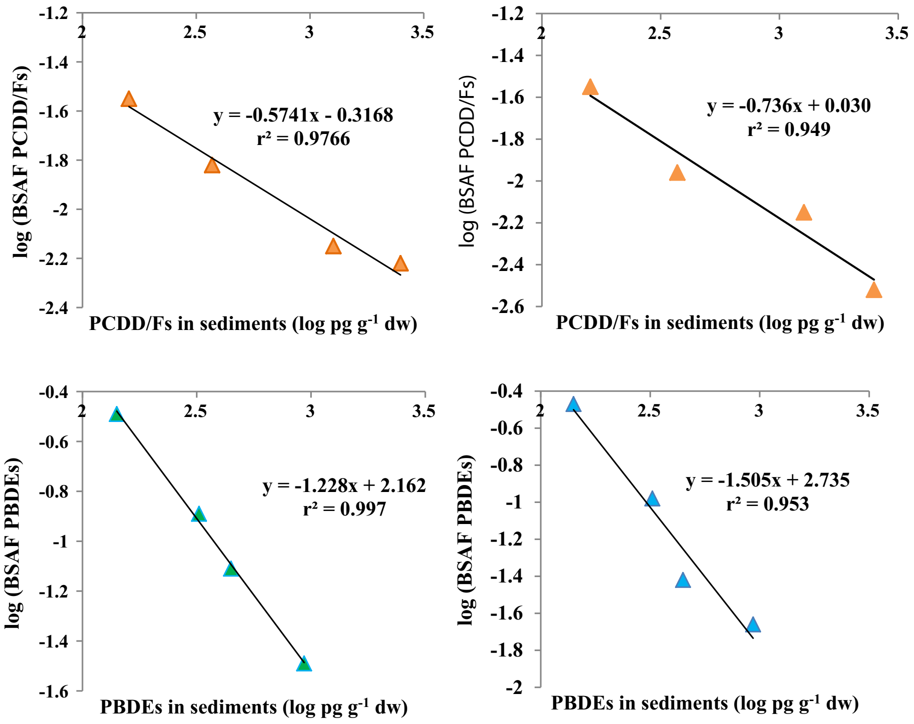

Fig. 7. Relationship between sediment concentrations and BSAF for PCDD/Fs and PBDEs. a) Left hand side — *L. niloticus*. b) Right hand side — *O. niloticus*.

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 (ATSDR, 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 (ATSDR, 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.

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