



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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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.

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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 km². 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

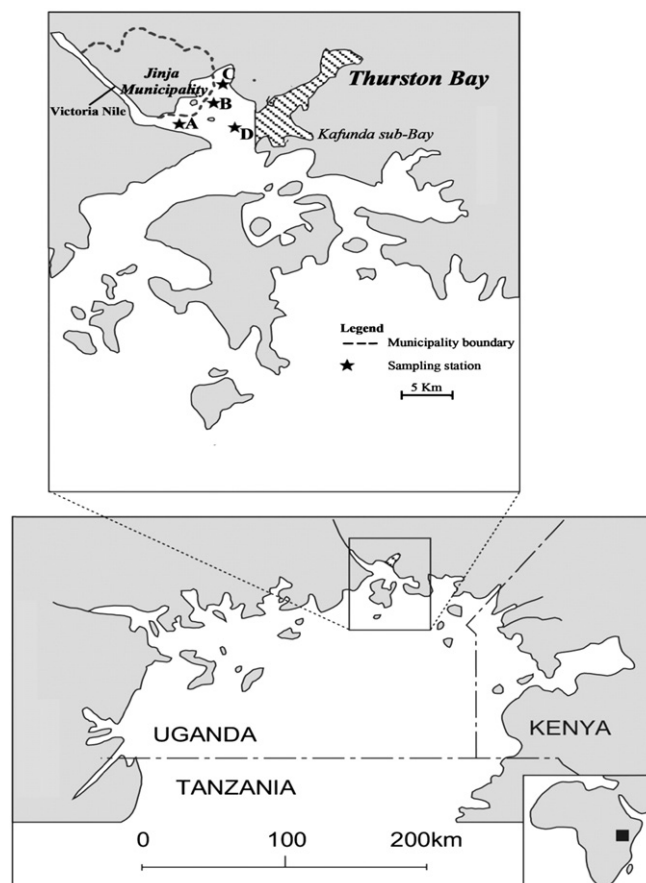


Fig. 1. Map showing the study stations. Adapted from Campbell et al. (2004).

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.

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 ¹³C-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 \times 0.25 mm i.d. \times 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 \times 0.25 mm i.d. \times 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 $^{\circ}\text{C}$ (1 min), 10 $^{\circ}\text{C min}^{-1}$ to 240 $^{\circ}\text{C}$ (1 min), 3 $^{\circ}\text{C min}^{-1}$ to 260 $^{\circ}\text{C}$ (1 min), 20 $^{\circ}\text{C min}^{-1}$ to 300 $^{\circ}\text{C}$ (10 min). The injection port temperature and detector temperature were 250 and 300 $^{\circ}\text{C}$, 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 Kruskal–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

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.

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).

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.

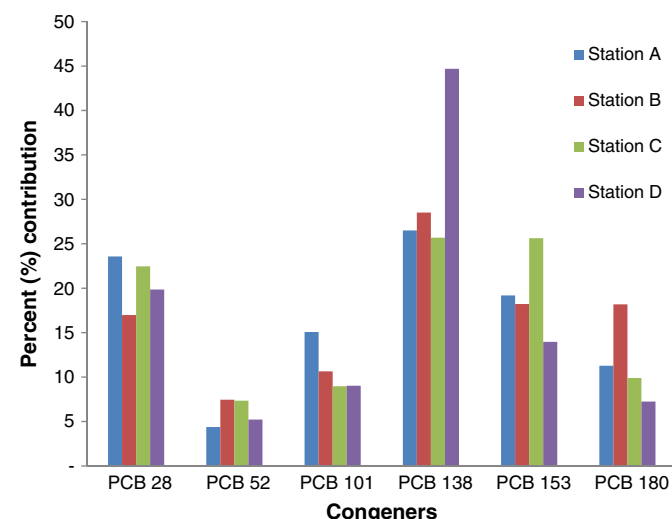


Fig. 2. Percent contribution of the individual congeners to the ΣPCBs in the sediments.

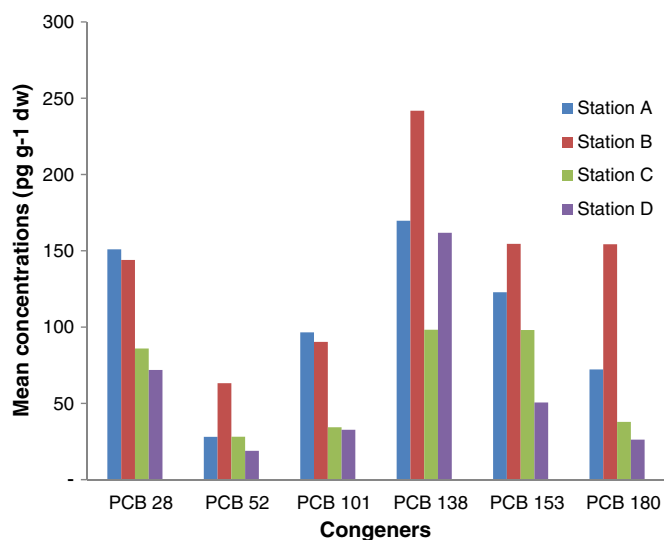


Fig. 3. Mean PCB concentrations in sediments from the different sampling sites.

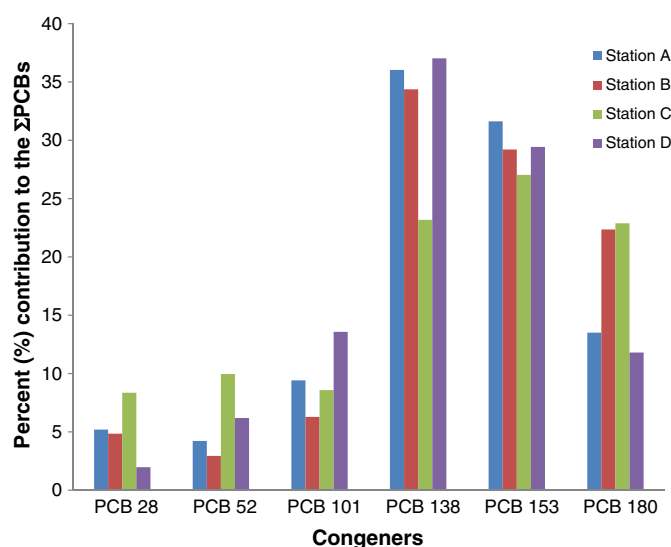


Fig. 4. Percent contribution of the individual congeners to the ΣPCBs in *L. niloticus*.

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

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

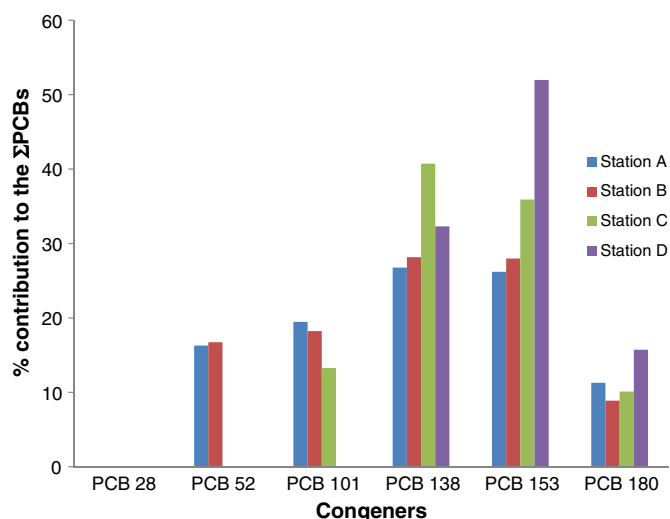


Fig. 5. Contribution of the individual congeners to the ΣPCBs in *O. niloticus*.

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