



Historical deposition of persistent organic pollutants in Lake Victoria and two alpine equatorial lakes from East Africa: Insights into atmospheric deposition from sedimentation profiles



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HIGHLIGHTS

- Profiles in Lake Bujuku were less altered by local vegetation than in Lake Mahoma.
- General decline of old POPs in recent sediments.
- PCBs and DDTs were the most historically deposited in the mountain lakes.
- DDTs had the highest average deposition flux in the core from Lake Victoria.
- *p,p'*-DDE, Σ CHLs and HCB mainly atmospherically deposited in the Lake Victoria core.

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ABSTRACT

Information on historical deposition of persistent organic pollutants (POPs) for African lakes is very limited. We investigated historical deposition trends and sources of POPs in sediment cores from Lakes Victoria (SC1), Bujuku (Buju2) and Mahoma (Maho2). The latter two lakes are situated in the Rwenzori mountain range in western Uganda. SC1 was taken from a central depositional area within the Ugandan part of the lake. Profiles in Buju2 and Maho2 were used as a reference for historical atmospheric deposition. For the post-1940 sediment deposits in SC1, average focusing factor-adjusted fluxes (FFFs) of Σ DDTs, polychlorinated biphenyls (Σ PCBs), hexachlorocyclohexanes (Σ HCHs) and chlordanes (Σ CHLs) were 390, 230, 210 and 120 ng m⁻² yr⁻¹. Higher fluxes of Σ DDTs, Σ PCBs, and Σ CHLs were observed in Buju2 and Maho2. The average FFF of HCB in Buju2 was the highest while the values for Maho2 and SC1 were similar. The endosulfan FFFs in SC1 were lower than in the alpine lake cores. Generally, Buju2 was a better reference for historical atmospheric deposition of POPs than Maho2 probably due to distortion of the latter's profile by Lake Mahoma's forested catchment. Profiles of *p,p'*-DDE, Σ CHLs and HCB in SC1 were consistent with atmospheric deposition while profiles of PCBs and HCHs were indicative of particle-bound loadings from additional sources. Profiles of endosulfans, DDTs, and chlordanes were consistent with influence of other factors such as anoxia, and dilution. Further studies of spatial resolution of historical deposition, especially in near-shore deposition areas of the lake are recommended.

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1. Introduction

Legacy persistent organic pollutants (POPs) and currently used pesticides (CUPs) have been detected in samples from Lake Victoria, including fish, sediment and water (Henry and Kishimba, 2006; Kasozi et al., 2006; Lipiatou et al., 1996). In the past, persistent

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organochlorine pesticides (OCPs), such as dieldrin and DDT, have been used extensively in the lake's basin (Ejobi et al., 1996; Muyambi, 2007; Wandiga, 2001). The basin is home to diverse agricultural and industrial activity which potentially release chemical pollutants into the environment, including biomass burning (Arinaitwe et al., 2012), dumping waste at poorly maintained landfills, open burning of waste and pesticide application on farms. Lake Victoria also receives significant influx from its tributaries, some of which, like Rivers Kagera, Nzoia, Nyando, Sondumirui and Sio, have very large drainage basins. These water channels drain through agricultural land and urban centers, effectively becoming contaminated by drift, leaching and surface run-off of CUPs and industrial waste chemicals. Other legacy chemicals from historical use in control of mosquitoes and insect pests could also find their way into the lake following atmospheric transfer.

Lake Victoria (surface area of about 68,800 km²) is exposed to significant atmospheric deposition of chemicals from sources within or external to the lake's basin. Atmospheric deposition is known to be the major route of global contamination by semi-volatile lipophilic POPs from point sources (Nizzetto et al., 2010; Stemmler and Lammel, 2009). In developed countries, atmospheric monitoring of POPs has been extensively undertaken through intergovernmental monitoring programs such as the Integrated Atmospheric Deposition Network (IADN), the cooperative program for monitoring and evaluation of the long-range transmission of air pollutants in Europe (EMEP) and the Arctic Monitoring and Assessment Program (AMAP). Similar programs in Africa, such as the Monitoring Network for Africa (MONET AFRICA) provide useful data but lack sufficiently continuous temporal data to enable the analysis of within- and between-year variations over a long period.

The tropical climate of Africa leads to higher volatilization rates of organic pollutants (Tanabe et al., 1994; Wania and Mackay, 1995; Barrie et al., 1992), underscoring the importance of sustained monitoring for atmospheric prevalence and estimation of spatial and deposition profiles of these chemicals. In Africa, only a few isolated studies, such as those in Malawi (Karlsson et al., 2000), Ghana (Poza et al., 2006; Adu-Kumi et al., 2012), South Africa (Poza et al., 2006; Batterman et al., 2008, 2009), several African sites (Klanova et al., 2009) and more recently, in Uganda (Arinaitwe et al., 2014), have demonstrated the atmospheric prevalence of POPs. Other studies on POPs in East Africa, have reported on point sources, such as from contaminated sites (Marco and Kishimba, 2005a), previous use in agriculture (Wandiga, 2001), availability in terrestrial (Ssebugere et al., 2010; Marco and Kishimba, 2005b, 2006) and aquatic systems (Henry and Kishimba, 2006; Kasozi et al., 2006; Ssebugere et al., 2009). None of these studies have investigated long term historical trends of atmospheric deposition of POPs in East African lakes. In water bodies, the ultimate sink of the POPs and related chemicals is the sediment where they are deposited and buried, although recycling by benthic organisms and other processes at the water–sediment interface (Jönsson et al., 2003; Froeschels et al., 2000; Looser et al., 2000) can be important. Analysis of POPs in dated lake sediment cores has been undertaken in many regions (Eisenreich et al., 1989; Rawn et al., 2001a, 2000; Smol, 2009) to establish historical deposition profiles. Some of these studies have shown that sedimentation profiles of POPs in cores of remote mountain lakes are a useful reference in temporal resolution of the historic atmospheric deposition of POPs in the vicinity of the lake (Rawn et al., 2001b; Muir et al., 2007). However, to our knowledge, only one study reported POPs in a dated sediment core from an African lake (Lipiatou et al., 1996) and it did not investigate the contribution from atmospheric deposition.

We investigated the historical deposition of POPs, in a sediment core taken from a central depositional zone of the Ugandan part of

Lake Victoria (Fig. 1). We also provide an insight into the historical state of the tropical atmospheric environment in East and Central Africa, with respect to atmospheric prevalence of selected POPs, by reporting their profiles in sediment cores taken from two tropical lakes, Bujuku and Mahoma. The two lakes are located in the Rwenzori Mountains (Panizzo et al., 2008; Russell et al., 2009; Eggermont et al., 2007) which are remote from urban and industrial activity. Hence, the measurements of POPs in these alpine lakes would represent background atmospheric loading by dry and wet deposition.

2. Materials and methods

2.1. Sampling

A brief description of the studied lakes is given in the supplementary material. Two sediment cores from Lake Victoria (SC1 and SC2) were taken at 74 m depth (0° 33' 13" S, 33° 15' 30" E) (Point SC in Fig. 1), in March 2009, using a Uwitec (Mondsee Austria) gravity corer, aboard RV IBIS, a research boat from the Uganda National Fisheries Resources Research Institute, NaFIRRI. The cores (9 cm diameter) were sub-sampled from the field using a core extruding apparatus to obtain 0.5 cm slices (top 10 cm) and 1 cm slices (the next 20 cm) up to a 30 cm core depth. Lake water collected at the sampling point was used for rinsing the slicing tool during sub sampling. The slices were stored in Nalgene jars (0.5 cm slices) and Whirl-Pak bags (1 cm slices) and stored at sub-zero temperatures (−18 to −20° C).

Four sediment cores (Buju 1–4 and Maho 1–4) were taken from the deep flat parts of each of lakes Bujuku and Mahoma, respectively, in June 2003, using a Glew Gravity corer (Glew, 1991). The sampling of these cores has been described in a study of mercury deposition profiles in one of the cores from each lake (Yang et al., 2010). The cores were extruded in the field and sub-samples for POPs analysis were taken every 0.5 cm and 1 cm for Buju and Maho, respectively. The sub-samples were stored in polyethylene bags and kept as cold as possible until analysis.

2.2. Analysis for ²¹⁰Pb and ¹³⁷Cs

For Lake Victoria, the core SC2 was used for dating and sedimentation rates were calculated using both the Constant Rate of Supply (CRS) and the Constant Initial Concentration (CIC) models (Appleby and Oldfield, 1978) using a gamma spectrometer with a lithium drifted germanium detector (for ¹³⁷Cs activity) and an alpha spectrometer (for lead 210 (²¹⁰Pb) activity). The determination of ²¹⁰Pb activity by alpha spectroscopy is destructive of the sample (Rawn et al., 2000; Appleby and Oldfield, 1978; Eakins and Morrison, 1978) (see supplementary material for a brief description). Maho3 and Buju3 were ²¹⁰Pb and ¹³⁷Cs dated using non-destructive gamma spectroscopy at the Centre for Environmental Research, University of Sussex, and sediment accumulation rates determined using the Constant Rate of Supply (CRS) model (Appleby and Oldfield, 1978; Eakins and Morrison, 1978). These dates were cross-correlated to other cores using unambiguous tie-points in the stratigraphic profiles of loss-on-ignition and dry weight (Fig. S2) and showed good temporal agreement.

2.3. Sample preparation, analysis and quality assurance

Sediment samples were extracted in a clean room (positively pressured HEPA™ and carbon filtered air) at the Canada Centre for Inland Waters, Burlington, Ontario. The extraction procedure was similar to the one in Rawn et al. (2000), with a few modifications. The details of sample preparation and GC analysis are given in the supplementary material.

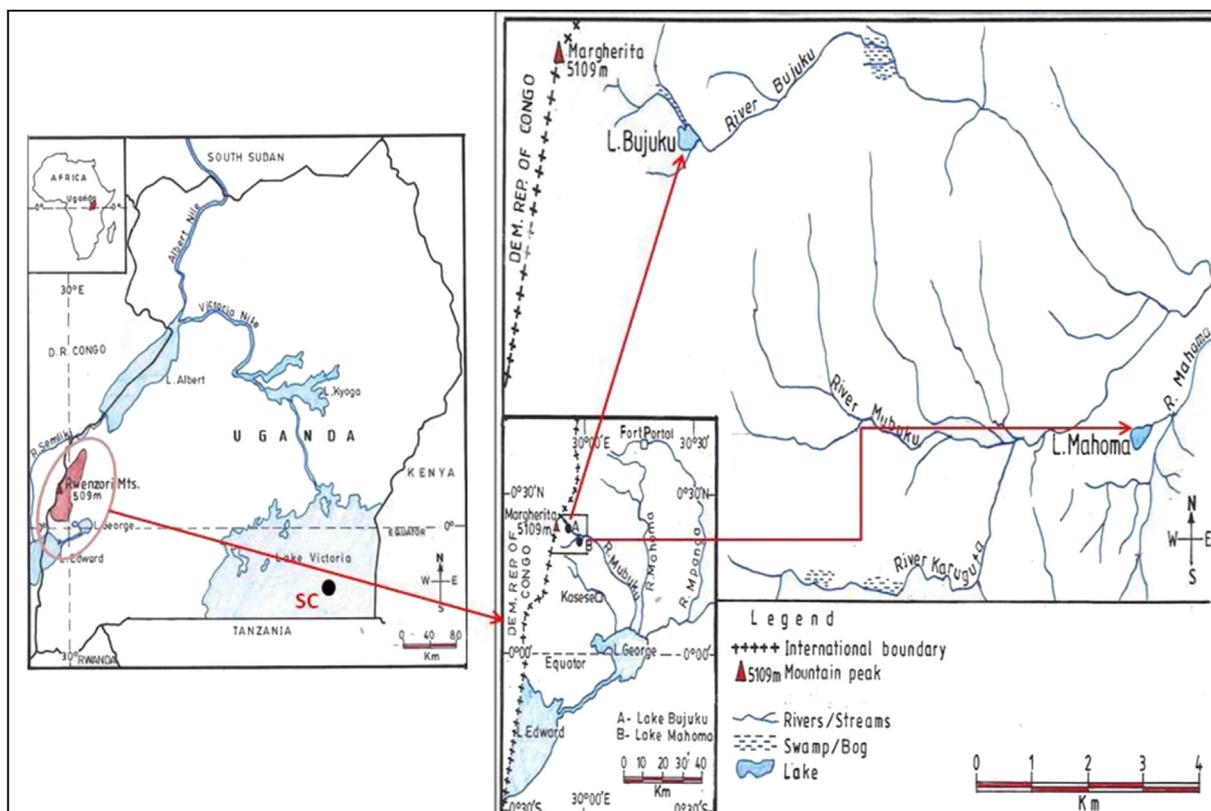


Fig. 1. Map of Uganda showing Lakes Bujuku, Mahoma and Victoria; Point SC is the location of the sampling site in Lake Victoria.

Prior to extraction, samples and procedural blanks were spiked with internal standards for recovery of PCBs (PCB 30, 204) and OCPs (135-TBB (1,3,5-tribromobenzene) and δ -HCH. The percent recoveries (\pm standard deviation) of these standards were $93 \pm 13\%$ for PCB 30, $103 \pm 11\%$ for PCB 204, $109 \pm 28\%$ for 135-TBB and $93 \pm 17\%$ for δ -HCH. Non-spiked procedural blanks were also analyzed. All results were blank corrected and concentrations expressed as ng/g dry weight. One half of the instrument detection limit (IDL) (Table S1) was used where the analyte's blank value was zero. Multilevel calibration ($R^2 > 0.99$) was used for GC analysis. Reference standards were purchased from Accustandard Inc., U.S.A. and Wellington Laboratories Inc., ON, Canada. All solvents were GC Grade (EMD Chemicals Inc., Darmstadt, Germany and Caledon Laboratories Ltd., Georgetown ON, Canada).

2.4. Data analysis

Reprocessing of chromatograms and spectra used Agilent Chemstation software. During statistical analysis, all correlation probabilities were Bonferroni adjusted (SYSTAT 13, Systat Software Inc., Chicago, IL) for significance at the 95% confidence level.

3. Results and discussion

3.1. Sediment chronology and lithostratigraphy

The sediment dating results for Buju3 and Maho3 have previously been reported (Panizzo et al., 2008; Yang et al., 2010). The ^{210}Pb activity and sediment accumulation profiles determined for Buju3, Maho3 and SC1 are given in Figs. S3 and S4, respectively. The ^{137}Cs activity profile of SC1 did not show the expected sub-surface peak in the sediment dated to the early 1960s. However, it did show subsurface step-variation in the sediment dated to the early 1990's

and around 1982. Given the depositional location of the sampling site, these observations could be due to one or a combination of any of the following: (a) extended residence times of ^{137}Cs in the water column and (b) diffusive redistribution of ^{137}Cs in aqueous phase within the sediment column. However ^{210}Pb showed an exponential decline with depth consistent with the CRS model enabling year of deposition and sedimentation rates to be estimated for SC1.

Average sedimentation rates computed for the cores were 0.05 (0.031 – 0.079) $\text{g cm}^{-2} \text{yr}^{-1}$ for Buju3, 0.021 (0.005 – 0.027) $\text{g cm}^{-2} \text{yr}^{-1}$ for SC1 and 7.9×10^{-3} (5.1×10^{-3} to 1.4×10^{-2}) $\text{g cm}^{-2} \text{yr}^{-1}$ for Maho3. The higher sedimentation rate of Buju3, compared to that of Maho3, is possibly due to high soil erosion in the catchment area of Lake Bujuku. This is reflected in the sediment accumulation profiles of the two lakes (Fig. S4). The sedimentation rate computed for SC1 is comparable to other rates previous determined in sediment cores from Lake Victoria (Lipiatou et al., 1996; Verschuren et al., 1998; Campbell et al., 2003).

The total organic matter (%LOI) in Maho3 was much higher than in Buju3 (Fig. S5) probably due to rain-driven loading of particulate matter from the forested catchment. In general, the average fraction of organic carbon (%OC) was about half the %LOI in Buju3. The high fraction of inorganic carbon in this core could be due to high loads brought in by the glacial melt waters and rain-driven soil erosion on the surrounding steep slopes. There is less exposure of Lake Mahoma to such erosion and other sources of inorganic carbon and, therefore, although %OC was not determined for Maho cores, it may be assumed that these cores had much higher %OC values than the cores from Lake Bujuku. The %OC profile of SC1 shows a gradual decline from the 1950s up to the late 1970s from where it increases rapidly. This increase in %OC coincides with a decrease in the carbon to nitrogen (C/N) ratio from approximately 12 to 11, indicating an autochthonous source of OC (Devesa-Rey and Barral, 2012). Indeed, the increase of algal populations in Lake

Victoria, resulting from increased eutrophication of the lake during this period is thought to have significantly contributed to anoxia in the off-shore sediment (Verschuren et al., 2002). This anoxia was found to be well correlated with anoxia-tolerant microbial populations and has implications for the fate of organic chemicals in the sediment. Hydrocarbon analysis of surface sediments collected in 1990 from a nearshore depositional site on the eastern side of the lake also indicated a significantly high algal detritus input (Lipiatou et al., 1996). The C/N ratio and algal populations profiles in Lake Bujuku have been previously reported elsewhere (Panizzo et al., 2008).

3.2. Sediment mixing and focus factors (FFs)

The mixed depths of SC1 and Maho3 were estimated to be 2 and 3.5 cm, respectively, while mixing was not observed in Buju3. The shallow mixed depth in SC1 is most likely due to anoxia and, therefore, limited bioturbation of the sediment in this depositional part of the lake. Previous measurements at the sediment–water interface in this part of Lake Victoria indicated that concentrations of oxygen were below 0.1 mg/L (Verschuren et al., 1998, 2002). The larger mixed depth in Maho3 could be due to a combination of higher organic matter from its forest/bamboo catchment (Eggermont et al., 2007; Yang et al., 2010) and higher oxygen concentration at the sediment–water interface (Eggermont et al., 2007), possibly supporting benthic feeders and burrowing organisms in the mixed layer.

Since no direct measurements of soil-derived estimates of atmospheric fluxes for ^{210}Pb have been reported within Equatorial Africa, a global long term estimate for geographical areas within $0\text{--}10^\circ\text{N}$ ($15.8\text{ mBq cm}^{-2}\text{ yr}^{-1}$) (Baskaran, 2011) was applied. Consequently the focus factor-adjusted fluxes (FFFs) have to be interpreted with caution since the global long term ^{210}Pb flux does not account for variations in the micro-climates of the three lakes or the morphology of the lakes' watersheds. The estimated FFs for the three cores were 2.4 for Buju3, 2.6 for SC1, and 5.6 for Maho3. The ratio of focusing between the two mountain lake cores (FF of Maho3/FF of Buju3) is 2.3. This ratio is similar to the one estimated from mercury measurements (2.1) for the Buju1 and Maho1 sediment cores (Yang et al., 2010). If these FFs represent the true situation in the cores, the values suggest relatively higher sedimentation in Maho3 than in Buju3, which could be largely due to a combination of secondary release of organic and particulate matter from the lakes forest/bamboo catchment, especially during rain events. Given that Lake Victoria receives suspended sediments from inflowing tributaries, runoff from the near-shore land surface and atmospheric deposition, the relatively low FF estimated here for SC1 is most likely due to the long distance over which sediment has to travel, both horizontally and vertically, to eventually be deposited at the sampling site. Both FF adjusted and unadjusted sedimentation fluxes of POPs in the three lakes are discussed in this paper. The POP profiles discussed here cover the sediment core depths dated to the period 1940–2008 (for SC1) and 1940–2003 (for Maho2 and Buju2) while the endosulfan profiles are for the sediment estimated to have been deposited after 1960 in all three sediment cores.

3.3. Profiles of POPs in the sediment cores

3.3.1. DDT residues

Of all the POPs targeted for analysis in this study, total DDT related compounds (ΣDDTs) were the most abundant in SC1 (Table S2) having the highest sub-surface maximum and surface concentrations (13 and 4.2 ng/g dw, respectively). In SC1, ΣDDTs also had the highest average FFFs of $390\text{ ng m}^{-2}\text{ yr}^{-1}$. Generally, the FFFs of ΣDDTs in the three cores were in the order Buju2 > Maho2 > SC1 (Fig. S6). The average FFFs for ΣDDT in Buju2

and Maho2 were 730 and 610 $\text{ng m}^{-2}\text{ yr}^{-1}$, respectively, although Maho2 had generally higher concentrations of ΣDDT than Buju2. The sub-surface peak concentrations of ΣDDT in Maho2, Buju2 and SC1 were dated to 1997 ± 3 (88 ng/g dw), 1986 ± 2 (5.7 ng/g dw) and 1993 ± 2 (13 ng/g dw), respectively.

From the 1950s to the 1960s, ΣDDTs in the alpine lakes decreased before increasing again from the 1970s to the early 1990s (Figs. S6 – S8). ΣDDT flux and concentration profiles in SC1 also show an increase from the early 1970s, peaking in the early 1990s with a second peak in the early-2000s before falling slightly in the late-2000s. There was an apparent similarity in the profiles of p,p' -DDE in the three cores (Fig. S9). The profiles show the same period of maximum p,p' -DDE flux (1980 – mid 1990s), peaking a few years later in SC1. This similarity suggests atmospheric deposition as a major contributor of p,p' -DDE in SC1 from Lake Victoria.

The proportion of p,p' -DDT in Buju2 and Maho2 was much higher than in SC1. The sediment sub-samples, from the former two cores, with the high proportions of p,p' -DDT also had the highest concentrations of ΣDDT . This may be due to slow degradation of DDT. The presence of DDT metabolites in amounts exceeding p,p' -DDT suggested old sources for DDT profile in SC1. This possibly follows atmospheric deposition of “weathered” DDT, metabolism of particle-adsorbed p,p' -DDT during the sediment focusing, especially from non-depositional near-shore areas and from constant deposition of autochthonous particles, such as dead algae. In SC1, p,p' -DDT was present only in sediment dated to early-2000s and the 1980s, and corresponding to periods of high deposition of ΣDDT residues in Buju2 and Maho2. The profile of DDT degradation products in SC1 was dominated by p,p' -DDE and o,p' -DDD while the predominant metabolites in Buju2 and Maho2 were p,p' -DDE and p,p' -DDD. The latter two are aerobic and anaerobic metabolites of DDT, respectively, while o,p' -DDD is the anaerobic metabolite of o,p' -DDT (Huang et al., 2001; Ricking and Schwarzbauer, 2012). Therefore atmospheric transfer of p,p' -DDE and anaerobic degradation of p,p' -DDT to p,p' -DDD in the sediment column were likely the major sources of the two metabolites in the alpine lake sediments. In SC1, the predominance of p,p' -DDE and o,p' -DDD is consistent with atmospheric deposition, aerobic degradation of p,p' -DDT on suspended particulates in the water column, elevated metabolism and abundance anoxia-tolerant biota in the sediment where SC1 was collected (Verschuren et al., 1998). Studies elsewhere have reported slower degradation rates of o,p' -DDT than for p,p' -DDT in tropical soil and in sediment (Huang et al., 2001; Ricking and Schwarzbauer, 2012), as well as higher re-volatilization potential of the former isomer whose vapor pressure is about 7.5 times that of the latter isomer. Thus over the long term, atmospheric deposition could increase the proportion of o,p' -DDT relative to p,p' -DDT in the lake, resulting in increased o,p' -DDD following anaerobic dechlorination of o,p' -DDT. Photodegradation of DDT could also lead to depletion of p,p' -DDT in atmosphere, yielding p,p' -DDE and other products (Göthe et al., 1976).

Profiles (based on dry weight) between p,p' -DDD and p,p' -DDE showed significant correlation in Buju 2 ($r = 0.906$) and Maho 2 ($r = 0.820$) (Table S3). The significant correlations are consistent with atmospheric deposition of DDTs followed by anaerobic degradation of p,p' -DDT to p,p' -DDD in the sediment column. The organic carbon-normalized concentrations in SC1 were higher than in Buju2 but less than in Maho2 (Fig. S10) as expected from the % OC profiles of these cores.

The post-1990 sediment in SC1 had higher ΣDDT subsurface concentrations than a sediment core collected in 1990 from a near shore depositional site on the eastern side of Lake Victoria (Lipiatou et al., 1996) although the concentration in the surface sediment in SC1 was lower.

3.3.2. Endosulfan residues

Historical deposition of total endosulfan (Σ Endo) in the three sediment cores was in the order Buju2 > Maho2 > SC1 (Fig. S12). From the late-1970s, there was a general increase in Σ Endo in Buju 2, peaking in the late 1990s. It is around this time that endosulfan residues are first detected in SC1 from where they proceed to increase, a sharp rise being observed near the sediment–water interface. Endosulfan sulfate was generally predominant in all cores (Fig. S11, Fig. S13). In Buju2, endosulfan sulfate was higher than the endosulfan isomers in all the sediment dated to the 1970–2003 period while the α -isomer also exceeded the β -isomer during this period, consistent with atmospheric deposition of “unweathered” endosulfan. For the period 1974–2003, the ranges of the ratios of endosulfan sulfate to α - and β -endosulfan were 1.2–4.3 and 2.7–6.6 respectively whereas the α -/ β -ratio for the same period had a range of 1.0–3.4, suggesting a short environmental residence time prior to deposition. The deposition profiles for the three compounds were strongly correlated. For instance, for the period 1963–2003, the correlation coefficients for endosulfan sulfate with the α - and β -isomers ($r = 0.859$ and 0.965 , respectively) and between the two isomers ($r = 0.856$) were significant (Table S4). These profiles are consistent with atmospheric deposition of endosulfan followed by biotransformation at the sediment–water interface. The deposition trends in Buju2 are also consistent with global historical use of endosulfan which has been on the rise since the 1960s (Weber et al., 2010). The average organic-carbon normalized endosulfan concentrations for Buju2 were about 10 times the ones for Maho2 and much higher than for SC1 (Fig. S12).

The general profile of Σ Endo in Maho2 is inconsistent with the one in Buju2; there is no clear general increase in endosulfan residues up the core and the β -isomer was detected only in sediment dated to 1990. There was no significant correlation between the profiles of endosulfan sulfate and the α -isomer. This indicated possible influence of other factors, in addition to direct atmospheric deposition, on the observed profile in Maho2. The canopy of the surrounding forest vegetation may scavenge the residues from the atmosphere. Re-volatilization from vegetation, “wash-off” of residues from the canopy during rain events and possible β -to α -conversion on vegetation or lake sediment (Weber et al., 2010) may eventually contribute to the overall “distortion” of the sedimentation profile of endosulfan in Maho2.

In the case of SC1, only β -endosulfan was detected in the top sediment dated 2006–2008 and the concentration exceeded that of endosulfan sulfate. β -endosulfan has been reported to be more susceptible to wet deposition than the α -isomer due to its higher solubility and lower Henry's Law Constant (Weber et al., 2010). This has also been observed in precipitation samples collected at the north shore of Lake Victoria between October 2008 and April 2010 in which the proportion of the β -isomer was higher in precipitation samples than in the air samples (Arinaitwe et al., in press). The depletion of α -endosulfan in sediment close to the sediment–water interface may also be explained by any (or a combination) of the following from other studies (Weber et al., 2010): (a) in aquatic systems, the α -isomer is preferentially metabolized over the β -isomer to produce endosulfan sulfate, especially in sediment rich in microbial populations even under anaerobic conditions; (b) the α -isomer volatilizes from aqueous surfaces to a greater extent than the β -isomer; this is significant for large water bodies like Lake Victoria where travel distances before settling in benthic sediment are long and environmental temperatures are elevated throughout the year and (c) in aquatic systems, the β -isomer was more likely to associate with sediment ($\log K_{OC\alpha} 3.6$; $\log K_{OC\beta} 4.3$) (Weber et al., 2010) while the α -isomer was more likely to be remobilized from sediment, resulting in its increased bioavailability and transformation to endosulfan sulfate. The depletion of the β - and α -

isomers in the sediments deposited before 2006 could be attributed to slow long term degradation of old endosulfan deposits to endosulfan sulfate by the anoxia-tolerant microbes in Lake Victoria.

3.3.3. Hexachlorocyclohexanes (HCHs)

The average FFFs of total HCHs (Σ HCHs) in the post-1940 sediment were 210, 130, and 110 $\text{ng m}^{-2} \text{y}^{-1}$ for SC1, Buju2 and Maho2, respectively (Table S2). This represents a very high loading of HCHs in Lake Victoria given its large surface area (about 68, 800 km^2) in contrast to the small surface areas of Lakes Bujuku and Mahoma (combined area less than 1 km^2) (Yang et al., 2010). The average FFFs of all the HCH isomers in SC1 were the highest. The FFFs for γ - and β -HCH in Buju2 were higher than in Maho2 while the latter had a higher FFF of α -HCH than the former.

The profile in Buju2 shows a general decline after the 1970s while HCH levels in SC1 rose during the same period (Figs. S14–16). This indicates significant contribution from sources, other than atmospheric deposition, to the HCH profile in SC1. The Buju2 sediment dated to the period covering the 1940s – early 1960s and the 1970s had high HCH concentrations. β -HCH had its peak concentration in the sediment dated early 1970s while α -HCH had its peak in the early-1960s. Having reached a maximum in the late-1950s, γ -HCH gradually decreased in recent sediment deposits reaching undetectable levels from the 1980s onwards. It is possible that the high fraction of γ -HCH in the pre-1960s period is probably due to increased local and global use of lindane (Breivik et al., 1999; Vijgen et al., 2011). Technical HCH (a mixture of HCH isomers) was used from the 1940s while global commercial distribution of lindane (γ -HCH up to 99%) started in the 1950s (Vijgen et al., 2011). The three cores show that the late 1970s – early 1980s period coincided with elevated HCH deposition. During this period, Maho2 and SC1 had peak deposits of γ -HCH. Maho 2 shows a general decline of HCHs from the 1950s to the late 1980s but differs from Buju 2 in two key ways: (a) that γ -HCH was undetected in Maho 2 in the 1940s and 1950s, but remains detectable in later sediment deposits; and (b) that there is a sharp increase in HCHs at the benthic sediment–water interface, dominated by β - and α -HCH. The high proportions of β - and α -HCH in the pre-1990 sediment in Maho2 reflects the historical use of technical HCH, which had higher proportions of these isomers, while the high concentrations of these isomers in the surface sediment of Maho2 are more likely to be from recent secondary release from the forested catchment. In SC1, the sediment dated to 1990–2008 showed a large increase in HCHs, over pre-1990 levels. This increase, characterized by the dominant β -HCH isomer, indicates an increased influence of legacy secondary sources on the loading profile of HCHs in Lake Victoria. Of the three HCH isomers, β -HCH has the lowest Henry's Law constant (Sahsuvur et al., 2003) and is, therefore, the most likely to be loaded into Lake Victoria through the aqueous phase. The Lake Victoria basin has high agricultural activity with a history of HCH use since the 1940s (Wandiga, 2001). This would have been largely technical HCH (with 5–14% being β -HCH) (Breivik et al., 1999; Vijgen et al., 2011), while contamination of aquatic systems from poorly managed obsolete pesticide storage has also been observed in the region (Elfvendahl et al., 2004; Kishimba et al., 2004). In addition, the Henry's Law constant of α -HCH is over 10 times that of β -HCH and this implies that the former isomer has a higher propensity to volatilize from the aqueous phase enhancing the proportion of β -HCH in SC1. The potential for the enhancement of β -HCH from isomerization has also been suggested but it is not a straight-forward occurrence, especially under field conditions (Phillips et al., 2005). Isomerization of γ - and α -HCH to β -HCH in both oxic and anoxic conditions, enhanced by organic nutrient enrichment has been reported although β -to γ - and α -conversions have also been observed in sterile deionized water (Phillips et al.,

2005). Most studies have instead reported γ - to α -HCH conversions in sediment/water systems (Phillips et al., 2005).

3.3.4. Polychlorinated biphenyls (PCBs)

In general, the total PCB (Σ PCB) fluxes were highest in Buju2 (Figs. S17 and Table S2). The average FFFs of Σ PCBs in the post-1940 sediment were 4060, 770 and 230 $\text{ng m}^{-2} \text{yr}^{-1}$ for Buju2, Maho2 and SC1, respectively. For each lake, the two homologues with the highest average FFF were Σ di- and Σ tri-PCBs for Buju2 and Maho2, and Σ penta- and Σ hexa-PCBs for SC1. The abundance of highly chlorinated congeners in SC1 indicates an influence of sources other than the atmospheric deposition of re-volatilized PCBs.

The general order of Σ PCB concentrations in the three sediment cores was Maho2 > Buju2 > SC1 with average post-1940 Σ PCB concentrations (ng/g dw) of 49, 18 and 2.5 and sub surface peak concentrations of 110, 67 and 19 ng/g dw , respectively (Table S2, Figs. S18–20). PCBs have high K_{OC} values and, therefore, are largely associated with particulate matter. This could explain the high concentrations of PCBs in Maho2 which was rich in organic matter. However, Buju2 shows much higher organic carbon normalized PCB concentrations (Fig. S21). In Buju 2, there was a general increase in Σ PCB levels from the late-1940s to early-1960s before a rapid decrease in the 1960s. The Σ PCB deposition then increased from the 1970s, with a brief decline towards the end of the same decade, before going on to peak in the sediment dated to early-1990s. The Σ PCB concentration then gradually decreased in the sediment deposited later. A similar trend is observed in Maho2, with the exception of the surface sediment. As with HCHs, the high Σ PCB signal in the Maho2 surface sediment could be due to recent secondary release of PCBs from the catchment by rain events. In Maho2, peak deposition was in the 1940s and minimum deposition in the 1960s before a gradual increase from early-1970s. A second peak in deposition is observed in the 1990s before falling in the early-2000s. In SC1, the Σ PCB concentrations in the sediment dated to the 1960s–1970s are generally higher than the ones observed in newer sediment deposits, the exception being the sediment dated to 1986 ± 2 yrs. This sediment had a Σ PCB concentration of 19 ng/g dw while the sediment dated to 1973 ± 3 yrs had the second highest concentration of 4.6 ng/g dw . Peak concentrations of Σ PCB in Buju2 and Maho2 were in the sediment dated to early-1990s (67 ng/g dw) and 1946 ± 3 (110 ng/g dw), respectively. The di- and tri-PCBs dominated the PCB profile in Buju2 (Fig. S18) as would be expected from atmospheric deposition given their low molecular weights relative to other PCB homologues. The Σ PCB profile in Buju2 was significantly correlated to the profiles of PCBs 15 + 18 ($r = 0.779$) and PCBs 33 + 53 ($r = 0.964$) (Table S5), indicating that these congeners were the best descriptor congeners for atmospheric deposition (the “+” sign denotes co-elution). In Maho2, the di-, tri-, tetra- and penta-PCBs were the predominant homologues (Fig. S19). The concentration of hexa-PCBs was higher in Maho2 than in Buju2, probably due to association with particulate organic matter released from Lake Mahoma's forest catchment. The Σ PCB profile was significantly correlated with PCBs 5 + 8, 33 + 53, 49, 52, 70, 101, 95, 149 + 118, and 153 + 132 (range of $r = 0.885$ –0.949), the highest coefficient being the one for PCB 33 + 53 (Table S6). These significant correlations involving the penta- and hexa-PCBs are an indication of factors additional to atmospheric deposition influencing the profile and could include rainfall-driven secondary release from the forest. In SC1, the sediment dated to the pre-1990s was richer in the di- and tetra-PCBs than post-1990 sediment (Fig. S20). The concentrations of the tri-, penta- and hexa-PCBs in SC1 were consistently high and were the dominant homologues from the late-1980s onwards. The hepta-PCB profile shows maxima in the early 1980s, at the turn of the century and in the mid 2000s. Whereas Buju2 and Maho2 had some significant correlations

involving di- and tri-PCBs, significant correlations in SC1 were between tetra- and penta-PCBs as well as between PCB 199 and 170 + 190 ($r = 0.818$) (Table S7). These correlations in SC1 underscore the significance of particle-bound PCB exposure to Lake Victoria. The lake is exposed to industrial and domestic waste via both waste water treatment plants and urban drainage channels from surrounding urban centers. It is also exposed to significant precipitation. These exposure routes could account for the elevated proportion of highly chlorinated congeners in SC1.

3.3.5. Chlordanes (CHLs)

The average FFFs of Σ CHLs for the post-1940 sediment for the three cores was in the order Buju2 > Maho2 > SC1 (Fig. S22, Table S2). Buju2 had the highest average FFFs for all chlordanes except oxychlordane, heptachlor and heptachlor epoxide. The highest average FFFs for heptachlor and heptachlor epoxide were observed in Maho2 while SC1 had the highest FFFs of oxychlordane and this indicated more chlordane degradation in Maho2 and SC1 than in Buju2.

The concentration of total chlordane (Σ CHLs) in the three cores was in the order Maho2 > SC1 > Buju2 (Fig. S23) with post-1940 average concentrations of 11, 1.4 and 1.0 ng/g dw , respectively. The concentration ranges were from below detection limit to 5.35 ng/g dw for SC1, 0.21–3.6 ng/g dw for Buju2 and 2.3–27 ng/g dw for Maho2. The Σ CHL concentration level in Buju2 gradually decreased from peak values in the sediment dated to the 1950s and 1960s to minimum values in the late-1980s before gradually increasing again in more recently deposited sediment. A similar Σ CHL trend is generally observed in Maho2. The decrease in Buju2 Σ CHL was accompanied by a significant decrease in the concentrations of the other chlordane related compounds, except α -chlordane, reaching non-detectable levels in the 1980s. Σ CHL concentration peaked in the three cores in the late-1970s. The surface sediment in Maho2 was richer in chlordane deposits than the other two cores, probably due to sediment mixing and recent secondary deposition from the catchment.

The Σ CHL concentrations in SC1 increased from the early-1990s reaching peak levels at the turn of the century. The 1970s, the early-1990s and the turn of the century stand out as the periods of relatively elevated deposition of Σ CHL in SC1. α -Chlordane, oxychlordane and cis-nonachlor were more predominant in the 1990s. Heptachlor was the most frequently detected and peaked in 1980 ± 2 yrs. Over all, there was a fair agreement in the profiles of Σ CHL of Maho2 and SC1 with similar periods of peak deposition although concentrations in Maho2 were much higher than in SC1. The difference in concentrations could be attributed to dilution in Lake Victoria while the similarity in peak periods could indicate long range atmospheric transfer to the region. The post-1940 FFFs Σ CHL of these two cores were generally on the same scale.

In the post-1980 Buju2 sediment, α -chlordane was the most frequently detected isomer and showed an increase dated to the early-2000s. Oxychlordane, heptachlor and the latter's oxidation product, heptachlor epoxide, were detected in 1961 ± 2 yrs which is the period of peak chlordane deposition for this core. It was also frequently detected in Maho2 but less so in Buju2. In Maho2, heptachlor and heptachlor epoxide were frequently detected while oxychlordane was observed in fewer sediment sub-samples. The absence of detectable oxychlordane in Buju2 relative to Maho2 indicates possible high incidence of chlordane degradation in the latter core although the chlordane concentration in Maho2 remains higher probably due to secondary release from its catchment. This could also explain the notably higher depletion of γ -chlordane in Maho 2 and SC1, especially in the sediment dated pre-1980. The absence of detectable heptachlor epoxide despite the frequent detection of heptachlor in SC1 is likely due to anoxia in this part of

the lake sediment. The proportion of trans-nonachlor in the alpine cores was generally higher than in SC1 although it was undetected in recent sediment deposits in Buju2.

3.3.6. Hexachlorobenzene (HCB) residues

The profiles of the three cores (Fig. S24) showed an increase in HCB deposition in sediment dated to the late-1980s and peaking in the early-1990s although an even larger peak was observed in the SC1 sediment at the turn of the century. This peak was also reflected in Maho2 and Buju2, albeit on a smaller scale. This indicates atmospheric deposition as the major route of HCB loading in Lake Victoria during this period. Buju2 showed peak deposition of HCB in the 1970s but this was not reflected in the other two cores. The average FFFs of HCB ($\text{ng m}^{-2} \text{yr}^{-1}$) for the post-1940 period were 120 for Buju2, 7.4 for Maho2 and 7.6 for SC1 (Table S2). In Buju2 the average FFF of HCB was higher than that of individual endosulfans and chlordanes. It was also higher than for individual endosulfans in SC1, indicating higher historical atmospheric transfer and deposition of HCB in the study region.

4. Conclusions

The deposition profiles of POPs in Buju2 were less altered by surrounding vegetation than profiles in Maho2. With reference to these two cores, it is evident that atmospheric deposition was the major historical loading route of p,p'-DDE, Σ CHLs and HCB in Lake Victoria. The PCB profile in SC1 also indicated additional exposure routes associated with the loading of particle-bound POPs. These routes include precipitation, inflowing rivers, urban centre drainage channels and run-off from near-shore land surface. These routes likely account for the observed HCH profile in SC1. Other factors that influence the identity and quantity of the contaminants were anoxia in that part of the lake where SC1 was collected, dilution due to the large size of Lake Victoria and sediment focusing. The latter two factors support the assertion that for a large lake like Lake Victoria, some of the near-shore areas may also be depositional zones with relatively higher historical and current depositions of contaminants. For instance, PCB concentrations (dry weight based) in surface sediments taken from Murchison Bay, Lake Victoria, in 2013, (Ssebugere et al., 2014) were generally higher than in surface sediment in SC1. Also, in a sediment core taken from the near-shore Kenyan part of the lake in 1990 (Lipiatou et al., 1996), the total PCBs in the surface sediment (representing deposition in the late 1980s) was more than ten times higher than the concentration in the surface sediment of SC1 as well as in the SC1 sediment horizon dated to 1990 ± 2 y. Therefore, these observations warrant further investigation of deposition profiles in near-shore areas to generate better spatial resolution. It would also be valuable to extend the study to other lakes in the region for comparative investigation. Activities within Lake Victoria's catchment have implications for the chemical profiles in the lake sediment. Therefore, effort should be made to promote practices that reduce the lake's risk to additional chemical loadings given its economic importance to the region.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.chemosphere.2015.10.061>.

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