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Prevalence and sources of polychlorinated biphenyls in the atmospheric environment of Lake Victoria, East Africa

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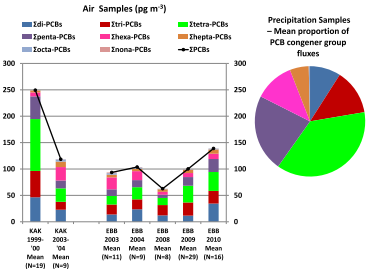
HIGHLIGHTS

- Samples collected at Entebbe showed a general increase in airborne ΣPCBs from 2003 to 2010.
- Combustion sources influenced the PCB profiles in Entebbe 2008–2010 samples.
- Densely populated areas in the region were likely major sources PCBs.
- Tetra- and penta-PCBs were predominant in precipitation samples.
- Generally, the same congeners were abundant in air and precipitation samples.

GRAPHICAL ABSTRACT



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ABSTRACT

The large surface area of Lake Victoria (about 68,800 km^2) makes it vulnerable to high atmospheric deposition of chemical pollutants. We present measurements of polychlorinated biphenyls (PCBs) from the lake's atmospheric environment. High volume air (24 h) samples were collected within the northern Lake Victoria watershed in Uganda over two periods; 1999–2004 [at Kakira (KAK) and Entebbe (EBB)] and 2008–2010 (at EBB only). Precipitation samples were also collected monthly during the 2008–2010 period at EBB. Analysis for PCBs was done using GC-μECD in a dual column approach. The ranges of ΣPCB concentrations in the KAK air samples were 154–462 pg m^{-3} (KAK 1999–2000), 26.7–226 pg m^{-3} (KAK 2003–2004), 27.0–186 pg m^{-3} (EBB 2003), 46.8–174 pg m^{-3} (EBB 2004), 19.2–128 pg m^{-3} (EBB 2008), 45.8–237 pg m^{-3} (EBB 2009) and 65.6–244 pg m^{-3} (EBB 2010). The di-, tri-, tetra- and penta-PCBs were predominant in air sample sets while the tetra- and penta-PCBs were predominant in precipitation samples. The mean flux of ΣPCBs in the precipitation samples was 26.9 ng m^{-2} (range of 14.8–41.5 and median of 27.5). Concentrations at EBB were lower than those reported elsewhere for urban sites in the East and Central African region. Multivariate analysis and analysis of air mass movements suggested influence of combustion sources on the PCB profiles from the region, especially, from the major East African urbanized regions.

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1. Introduction

PCBs were produced from the 1920's to 1980's and used in various industrial applications, such as coolants, dielectrics, reactive flame retardants and as plasticizers. Industrial production of PCBs was banned in the US, Western Europe and Japan in the 1970's and later, globally, in 2004 under the Stockholm Convention. In order to facilitate the evaluation of the effectiveness of the Stockholm Convention in reducing the global proliferation of POPs, the Global Monitoring Programme was put in place, requiring member states to monitor POPs in three key indicator matrices; human milk, human blood and air (UNEP, 2004). Various studies conducted in regions of historically high PCB production and use have shown a decline in air concentrations since they were banned (Buehler et al., 2002; Sun et al., 2007; Hung et al., 2010; Schuster et al., 2010; Venier and Hites, 2010; Tørseth et al., 2012; Salamova et al., 2013; Hung et al., 2016). These studies span long periods of time under national and international monitoring programmes. Long time series of data from such monitoring on the African continent are lacking. Only a few studies have reported outdoor atmospheric measurements of PCBs with some suggesting increasing emissions of PCBs into the atmosphere (Gioia et al., 2014). Old PCB-containing technologies (such as old electric transformers, capacitors and other old electrical equipment) remain in use or are still being imported in the developing countries, and are likely sources of major emissions of these chemicals in the environment (Gioia et al., 2014). Various emission processes have been suggested, including dismantling of PCB containing, illegal or improper dumping of PCB containing wastes, leaks or releases from electrical transformers containing PCBs and open low-temperature burning of PCB containing equipment. The atmospheric prevalence of lower PCB congeners has been associated with continuous volatilization while combustion (such as open burning of PCB containing materials, waste incineration and accidental fires) is a source of higher PCB congeners in the atmosphere (Breivik et al., 2002). The emitted PCBs are dispersed to non-source areas by various means, long range atmospheric transfer being the major route of global contamination, especially for large water bodies. The large surface area of Lake Victoria (about 68, 800 km^2) and a watershed area of about 284,000 km^2 potentially expose the lake to significant atmospheric deposition of chemical pollutants. A few studies have reported PCBs in the fish (Ssebugere et al., 2013a) and sediments (Lipiatou et al., 1996; Ssebugere et al., 2013b; Arinaitwe et al., 2016) from Lake Victoria, but there remains lack of adequate information on exposure of the lake to PCBs from atmospheric input.

This study aimed at generating baseline prevalence data of PCBs in Lake Victoria's atmospheric environment. We present results of PCB analysis of high-volume air and precipitation samples collected from the northern shoreline of Lake Victoria, East Africa.

2. Materials and methods

2.1. Sampling and sample preparation

High Volume air samples (24 h, 283 m^3 each) were collected with a TE-1000 PUF high volume air sampler (Tisch Environmental Inc., OH, USA) at Kakira sugarcane plantation in Jinja (KAK, 0°30′40.53′ N, 33°16′48.97′ E) and at the Directorate of Water Resources Management (DWRM) at Entebbe (EBB, 0°02′55.38′ N, 32°28′19.50′ E), both located near the northern shoreline of Lake Victoria. The Kakira (KAK) and Entebbe (EBB) sampling sites and sampling procedures have been described in detail elsewhere (Arinaitwe et al., 2012, 2014). In summary, the samples were collected over two sampling campaigns. In the first campaign, samples were collected intermittently at KAK from 1999 to 2003 and at EBB from 2003 to 2004. In the second campaign, samples were collected from EBB from October 2008 to July 2010. The samples were initially stored (−16 °C - 20 °C) at Makerere University and later analyzed in Canada. The samples were extracted (combined PUF + filter) and analyzed for PAHs (Arinaitwe et al., 2012) at AirZoneOne Laboratories (Mississauga, ON) before further processing and analysis for POPs in a clean room (positively pressured HEPA™ and carbon filtered air) at the Canada Centre for Inland Waters (CCIW) in Burlington, Ontario.

The precipitation samples were collected at EBB between October 2008–April 2010 using a Meteorological Instruments of Canada (MIC) type sampler for automated precipitation sampling. During rain events, the collected precipitation drained into a calibrated container through a column packed with an XAD-2 resin. Every resin column was deployed for a period of one month. The detailed description of the precipitation sample collection, including description of the sampler and the individual resin column deployment period have been previously reported (Arinaitwe et al., 2014). The resin columns were then refrigerated and later sent to CCIW for analysis.

The sample extraction and cleanup procedures have been previously described in detail (Arinaitwe et al., 2014). In summary, the air samples were extracted by pressurized fluid extraction (ASE 200, Dionex Instruments) with Hexane/Acetone 70:30 v/v. The concentrated extract was fractionated on an activated silica column using hexane (fraction A) and hexane/DCM 1:1 v/v (fraction B). Fraction A was analyzed for PCBs. For precipitation samples, the XAD resin was successively eluted with methanol and DCM. The organic layer was separated, dried over anhydrous Na_2SO_4 then processed further for PCB analysis as was with the air sample extracts.

2.2. Quality control

The high volume air samplers were regularly calibrated following the manufacturer's manual. Field PUF/filter and XAD resin column blanks, laboratory procedure and recovery blanks were analyzed. All the samples were spiked with internal standards during sample preparation. The internal standards used were PCBs 30, 166 and 204 for air samples and PCBs 30 and 204 for precipitation samples. The average recoveries of these standards were 106%, 102%, 76% (for PCBs 30, 166 and 204, respectively, in air samples) and 78%, 99% (for PCBs 30 and 204, respectively, in precipitation samples). All data were blank corrected using the average blank concentration. For compounds not detected, one-half of the instrument detection limit (IDL) (Table S1) was applied during statistical analyses. The IDL was determined as the concentration of the target analyte giving rise to a signal/noise ratio in the range of 2.5–5 (EPA, 2011).

2.3. Sample and data analysis

PCB analysis for the KAK and EBB (2003–2004) samples was done by gas chromatography with electron capture detection (GC ECD) using an Agilent 5890 GC equipped with dual ECDs. For the EBB 2008–2010 air and rain samples, an Agilent 6890 GC-ECD was used. Dual column analysis was done using HP-5, HP-5MS, HP-5 and HP-1MS columns (30 m × 0.25 mm x 0.25 μm, Agilent Technologies Inc., U.S.A). Hydrogen was used as carrier gas at a flow rate of 1 mL/min. The injector temperature was 220 °C while detector temperatures were 350 °C and 300 °C for the HP and Agilent GCs respectively. The oven temperature program was: 80 °C for 2 min, 10 °C/min to 150 °C, 2 °C/min to 280 °C, and hold for 5 min (total run time of 79 min). Data processing was done using Agilent GC