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Concentrations, distribution, sources and risk assessment of organohalogenated contaminants in soils from Kenya, Eastern Africa

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ABSTRACT

The organohalogenated contaminants (OCs) including 12 organochlorine pesticides (OCPs), 7 indicator polychlorinated biphenyls (PCBs) and 7 polybrominated diphenyl ethers (PBDEs) were determined in soils collected from Kenya, Eastern Africa. The total OCPs fell in the range of n.d–49.74 μ g kg⁻¹ dry weight (dw), which was dominated by DDTs and endosulfan. Identification of pollution sources indicated new input of DDTs for malaria control in Kenya. The total PCBs ranged from n.d. to 55.49 μ g kg⁻¹ dw, dominated by penta- and hexa-PCBs, probably associated with the leakage of obsolete transformer oil. The soils were less contaminated by PBDEs, ranging from 0.19 to 35.64 μ g kg⁻¹ dw. The predominant PBDE congeners were penta-, tri- or tetra-BDEs, varying among different sampling sites. Risk assessment indicated potential human health risks posed by OCs in soils from Kenya, with PCBs as the most contributing pollutants. The local authorities are recommended to make best efforts on management of OC pollution, particularly from DDTs and PCBs to meet the requirement of Stockholm Convention.

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1. Introduction

Organohalogenated contaminants (OCs) such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), also known as persistent organic pollutants (POPs) regulated by the Stockholm Convention (UNEP, 2009), are a group of hydrophobic compounds which were highly toxic, environmental persistent and long distance transportable (Gouin et al., 2004; Wania and MacKay, 1996). OCPs such as DDT and HCH were once extensively used in agriculture worldwide. Despite the ban of production and application of OCPs since the 1970s in many countries, OCPs residues have been reported in water, air and soils around the world (Ge et al., 2013; Rios et al., 2010; Whitehead et al., 2015; Yang et al., 2014). PCBs had been used as transformer dielectric fluids, flame retardants, plasticizers and pesticide additives since the 1930s, and have also been identified in various environmental compartments and human tissues

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(Everaert et al., 2015; Koh et al., 2015; Wang et al., 2011). PBDEs have been widely used as flame retardants, and detected ubiquitously in the environment (Guan et al., 2007; Jiang et al., 2010). PBDEs were considered as emerging POPs under Annex A of the Stockholm Convention.

Kenya is located within the eastern side of the vast continent of Africa. Kenya became a contracting party to Stockholm Convention in 2004. Kenya had a population of approximately 32 million in 2003, about 70% of which lived in rural areas. Greater than two thirds of the land in Kenya is either desert or semi-desert, with only approximately 18% of the land suitable for agricultural use (Kenya NIP, 2007). Besides, unpredictable climate changes such as floods and droughts occurred frequently. Large efforts have therefore been made to enhance the food security, and the agricultural chemicals including obsolete OCPs have been consequently overused and even misused to control pests and boost agricultural productivity (UNEP, 2015). Although DDTs were banned for agricultural use in 1986 in Kenya, the other OCPs were not banned until 2004 (Kenya NIP, 2007). Some OCPs are still in use for public health purposes (Kenya NIP, 2007). PCBs can be released into the environment due to improper disposal of the waste equipment (Kenya NIP, 2014). Similarly, PBDEs contamination can be caused by poor





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management of waste electrical and electronic equipment, end-oflife vehicles and other articles such as fire-fighting foams, aviation hydraulic fluids, and textiles (Arinaitwe et al., 2014; Odusanya et al., 2009). Nevertheless, environmental contamination by POPs in Kenya has not been well studied. Only a few reports were found in the literature (Everaarts et al., 1998; Omwoma et al., 2015; Oyugi et al., 2003). To our knowledge, POPs in the Kenyan soils were not reported.

Therefore, this study was designed to investigate the occurrence of OCs including OCPs, PCBs and PBDEs in soils from southern Kenya, the most flourish areas around Nairobi. The OCs pollution characteristics including concentrations, composition, and spatial distribution were analyzed, as well as possible sources of the detected pollutants. Especially, we were interested in potential adverse effects on human health caused by OCs in soils. The results could expand our knowledge about the POPs contamination in this area, and provide useful information to the local authority for risk management of POPs pollution and implementation actions of the Stockholm Convention.

2. Method and materials

2.1. Chemicals and materials

The mixed standard solution of 12 OCPs including α -HCH, β -HCH, γ -HCH, δ -HCH, p,p'-DDE, p,p'-DDD, p,p'-DDT, heptachlor, heptachlor epoxide, α -endosulfan, β -endosulfan, and endosulfan sulfate were purchased from AccuStandard (New Haven, CT, USA). Seven indicator PCB (PCB-28, 52, 101, 118, 138, 153 and 180), seven PBDE congeners (BDE-28, 47, 99, 100, 153, 154 and 183) and the internal standard pentachloronitrobenzene (PCNB) were also pur-AccuStandard. chased from The recovery surrogate 2,2',3,3',4,4',5,5',6,6'-decachlorobiphenyl (PCB209) was obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany). The solvents used such as dichloromethane (DCM) and n-hexane were of HPLC grade (Fisher Scientific, USA). The other reagents were analytically pure. Anhydrous sodium sulfate (Na₂SO₄) was baked at 450 °C for 4 h. Florisil (60-100 mesh) were activated at 150 °C for 10 h. Silica gel of 100-200 mesh was activated at 180 °C for 4 h, and acidified with 30% (v/m) H₂SO₄ before use. Copper powder was activated with 2 N hydrochloric acid (HCl), washed with distilled water and stored in acetone until use.

2.2. Sample collection

Soil samples were collected in January 2015 from the rural and suburban area surrounding Nairobi, the capital of Kenya (Fig. 1). This area was one of the most developed regions in eastern Africa, with fertile land for agriculture, convenient transportation for industry and tourism. To be specific, 16 samples were from the rural area of Mai Mahiu, 13 samples from the suburban area of Narok, 14 samples from the Mount Suswa Conservancy, 10 samples from the rural area of Juja, and 6 samples from the rural area of Limuru Town. Mount Suswa Conservancy was a volcano in the Great Rift Valley, and was famous for the wildlife tours. In our study, this site was set as a background control. The top soils of 0-5 cm were collected with a stainless steel shovel in 3 subsamples which were combined as one at each site. The soil samples were lyophilized and homogenized for extraction of target analytes.

2.3. Sample extraction and instrumental analysis

The extraction procedure of OCPs, PCBs and PBDEs in soils was similar with our previous reports (Yun et al., 2014, 2015), with

minor modification. One gram of the soil sample was spiked with a known amount of PCB209, and ground together with 3 g C18 (Silicycle, Inc., Québec, Canada) thoroughly for 5 min. The homogeneous mixture was then transferred onto the top of a multilayer cartridge packed in a polyethylene syringe barrel column (10 mL). The cartridge consisted of 0.5 g Na₂SO₄, 1 g florisil, 1 g acidic silica gel, and 0.5 g activated copper powder, from bottom to top successively. The target analytes were eluted with 15 mL of DCM. The eluent was dried under a gentle nitrogen flow. The residues were re-dissolved with 100 μ L of n-hexane. A known amount of PCNB was spiked as the internal standard before instrumental quantification.

Qualitative and quantitative analysis of OCPs, PCBs and PBDEs were carried out on an Agilent 7890A gas chromatograph equipped with an electron capture detector (GC-ECD) and a Model 5975 mass spectrometer (MS) using electron-ionization ion source (EI) in the selected ion monitoring (SIM) mode (Agilent Technologies, Santa Clara, CA, USA). A capillary column HP-5MS (Agilent Technology, 30 m \times 0.25 mm i.d. \times 0.25 $\mu m)$ was used to separate target analytes. The carrier helium gas flow was 1.2 mL min⁻¹. The injection volume was 1 µL in splitless mode. The temperatures of the inlet and detector were 280 °C and 300 °C, respectively. The oven temperature was programmed as follows: 80 °C for 1 min, to 190 °C (2 min) at 15 $^{\circ}$ C min⁻¹, then to 220 $^{\circ}$ C (5 min) at 8 $^{\circ}$ C min⁻¹, and finally to 300 °C (7 min) at 10 °C min⁻¹. The total time required for analyzing one single sample was approximately 34 min. The ion source temperatures were set to 300 °C. The data were acquired and processed with Chemstation software (Hewlett–Packard).

The total organic carbon (TOC) fraction of the soil samples were analyzed on a solid TOC analyzer (Elementar, Germany), consuming approximately 5 mg soil wrapped in aluminum foil for each assay.

2.4. Quality assurance and quality control (QA/QC)

Various control experiments were carried out to validate the analytical processes. Solvent blanks and solvent spike experiments at three levels were conducted, and the average recoveries of the 26 compounds were 102 \pm 36%, 92 \pm 19% and 93 \pm 15% for spiking levels of 5, 10 and 20 ng g^{-1} , respectively. The average recoveries of 26 compounds in matrix blank and spike experiments were $96 \pm 24\%$, $92 \pm 18\%$ and $80 \pm 13\%$ for 5, 10 and 20 ng g⁻¹, respectively. A solvent blank was monitored for every 10 samples, and a pair of matrix blank and spike experiments at 10 ng g^{-1} were analyzed for every 20 samples, and the average recoveries of the 26 compounds ranged from 70 \pm 15% to 114 \pm 5%. The surrogate standard (PCB209) was spiked to all samples before extraction, and the average recovery of PCB209 in all samples was 78 \pm 12%. A standard solution with 5 times the signal-to-noise ratio (S/N) was analyzed for 6 runs, and the limit of detection (LOD) was defined as 3.36 times of the deviation, which ranged from 0.001 to 0.025 ng g^{-1} for the 26 compounds. The degradation ratio of DDT at the GC inlet was less than 15%, as suggested by USEPA Method 8081B. The calibration curves were established for each compound with concentrations ranging from the LOD to 0.5 ng g^{-1} and 0.5–20 ng g⁻¹, respectively, with $R^2 > 0.99$ for all compounds in both concentration intervals. The quantification was performed based on the internal standard normalization. The concentrations reported were in ng g^{-1} dw.

2.5. Risk assessment

Potential human health risks caused by OCs in soils were assessed based on the Regional Screening Levels (RSL) for residential soils proposed by the US Environmental Protection Agency (EPA). The total lifetime span carcinogenic risk (TLCR) and total Download English Version:

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