



# 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  $\mu\text{g kg}^{-1}$  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  $\mu\text{g kg}^{-1}$  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  $\mu\text{g kg}^{-1}$  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

(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-of-life 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  $\alpha$ -HCH,  $\beta$ -HCH,  $\gamma$ -HCH,  $\delta$ -HCH,  $p,p'$ -DDE,  $p,p'$ -DDD,  $p,p'$ -DDT, heptachlor, heptachlor epoxide,  $\alpha$ -endosulfan,  $\beta$ -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 purchased from AccuStandard. 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 ( $\text{Na}_2\text{SO}_4$ ) 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)  $\text{H}_2\text{SO}_4$  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 packed in sealed bags and transferred to the lab where they 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  $\text{Na}_2\text{SO}_4$ , 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  $\mu\text{L}$  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  $\mu\text{m}$ ) was used to separate target analytes. The carrier helium gas flow was 1.2 mL  $\text{min}^{-1}$ . The injection volume was 1  $\mu\text{L}$  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 °C  $\text{min}^{-1}$ , then to 220 °C (5 min) at 8 °C  $\text{min}^{-1}$ , and finally to 300 °C (7 min) at 10 °C  $\text{min}^{-1}$ . 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  $\text{g}^{-1}$ , 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  $\text{g}^{-1}$ , respectively. A solvent blank was monitored for every 10 samples, and a pair of matrix blank and spike experiments at 10 ng  $\text{g}^{-1}$  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  $\text{g}^{-1}$  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  $\text{g}^{-1}$  and 0.5–20 ng  $\text{g}^{-1}$ , 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  $\text{g}^{-1}$  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

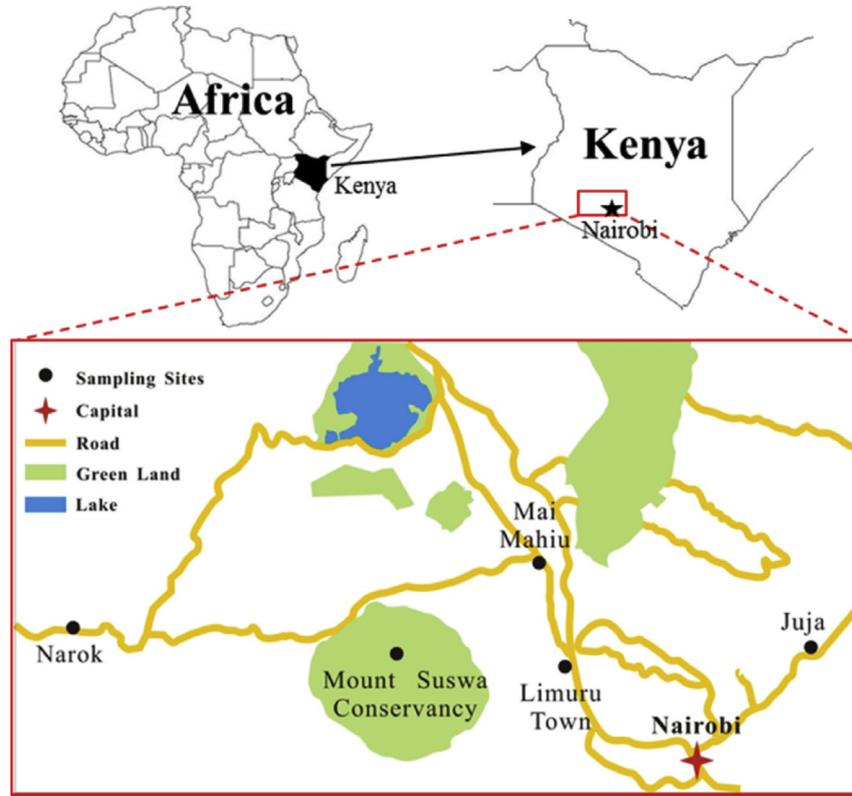


Fig. 1. Map of sampling sites in Kenya, Eastern Africa.

non-carcinogenic hazard quotient (TnHQ, for child) were calculated for these pollutants in the present study, covering the exposure routes of incidental ingestion of soil, inhalation of volatiles and particulates emitted from soil and dermal contact with soil (USEPA, 2015). To be specific, the TLCR was calculated according to Eqs. (1)–(6), and the TnHQ was calculated following Eqs. (7)–(10).

$$LCR_{Ingestion} = \frac{C_s \times CSF \times IFS}{AT \times LT} \quad (1)$$

$$LCR_{Dermal\ adsorption} = \frac{C_s \times CSF \times DFS \times ABS_d}{GIABS \times AT \times LT} \quad (2)$$

$$LCR_{Inhalation} = \frac{C_s \times IUR \times ED \times ET \times EF \times (1/VF_s + 1/PEF_w)}{AT \times LT} \quad (3)$$

$$TLCR = LCR_{Ingestion} + LCR_{Dermal\ adsorption} + LCR_{Inhalation} \quad (4)$$

where

$$IFS = \frac{IRS_c \times EF \times ED_c}{BW_c} + \frac{IRS_a \times EF \times (ED - ED_c)}{BW_a} \quad (5)$$

$$DFS = \frac{SA_c \times EF \times AF_c \times ED_c}{BW_c} + \frac{SA_a \times EF \times AF_a \times (ED - ED_c)}{BW_a} \quad (6)$$

and  $C_s$  was the concentrations of POPs in soil,  $\mu\text{g kg}^{-1}$  dw; CSF in  $(\text{mg kg}^{-1} \text{d}^{-1})^{-1}$  was the carcinogenic slope factor of the specific compound; IFS in  $\text{mg kg}^{-1}$  was the age adjusted resident soil ingestion rate; AT was the averaging time, 365 d; LT was the

lifetime, 70 years; DFS in  $\text{mg kg}^{-1}$  was the age adjusted resident soil dermal contact factor;  $ABS_d$  was the fraction of contaminant absorbed dermally from soil, unitless; GIABS was fraction of contaminant absorbed in gastrointestinal tract (unitless); IUR in  $(\mu\text{g m}^{-3})^{-1}$  was the chronic inhalation unit risk;  $PEF_w$  was the particulate emission factor,  $1.36 \times 10^9 \text{ m}^3 \text{ kg}^{-1}$ ;  $VF_s$  was the volatilization factor; EF was the exposure frequency, 350 days  $\text{y}^{-1}$ ; ED was the exposure duration, 26 yr; ET was the exposure time, 24 h  $\text{d}^{-1}$ ;  $ED_c$  was the ED of child, 6 yr;  $IRS_c$  was the soil ingestion rate of child, 200  $\text{mg d}^{-1}$ ;  $IRS_a$  was the soil ingestion rate of adult, 100  $\text{mg d}^{-1}$ ;  $BW_c$  was the body weight of child, 15 kg;  $BW_a$  was the body weight of adult, 80 kg;  $SA_c$  was the surface area of child, 2373  $\text{cm}^2$ ;  $SA_a$  was the surface area of adult, 6032  $\text{cm}^2$ ;  $AF_c$  was the soil adherence factor of child, 0.2  $\text{mg cm}^{-2}$ ;  $AF_a$  was the soil adherence factor of adult, 0.07  $\text{mg cm}^{-2}$ .

$$nHQ_{Ingestion} = \frac{C_s \times EF \times IRS_c}{AT \times BW_c \times RfD} \quad (7)$$

$$nHQ_{Dermal\ adsorption} = \frac{C_s \times EF \times SA_c \times AF_c \times ABS_d}{GIABS \times AT \times BW_c \times RfD} \quad (8)$$

$$nHQ_{Inhalation} = \frac{C_s \times ET \times EF \times (1/VF_s + 1/PEF_w)}{AT \times RfC} \quad (9)$$

$$TnHQ = nHQ_{Ingestion} + nHQ_{Dermal\ adsorption} + nHQ_{Inhalation} \quad (10)$$

where RfD refers to the chronic oral reference dose of specific contaminant,  $\text{mg kg}^{-1} \text{d}^{-1}$ ; RfC in  $\text{mg m}^{-3}$  was the chronic inhalation reference concentration of specific contaminant. The parameters involved were provided in Table S1 of the Supplementary Material.

## 2.6. Statistical analysis

Statistical analysis was performed with SPSS software (IBM, USA). Pearson correlation analysis was performed to investigate the relationship between TOC and OCs concentrations. A  $p$ -value  $\leq 0.05$  was considered statistically significant.

## 3. Results and discussion

### 3.1. Profiles of OCPs contamination and potential sources

Table 1 shows the concentration profiles of OCPs in soils. All OCPs species except  $\beta$ -HCH were frequently detected, with  $\Sigma$ OCPs concentrations in the range of n.d.–14.22, 2.91–23.93, 2.01–7.77, 0.10–35.41 and 10.61–49.74  $\mu\text{g kg}^{-1}$  dw for Mai Mahiu, Narok, Mount Suswa Conservancy, Juja and Limuru Town, respectively. Fig. 2 shows the spatial distribution of OCPs. The highest  $\Sigma$ OCPs concentrations were found at Limuru Town, with an average as high as 24.20  $\mu\text{g kg}^{-1}$  dw, while the lowest  $\Sigma$ OCPs concentrations were detected at Mount Suswa Conservancy, with an average concentration of 3.88  $\mu\text{g kg}^{-1}$  dw. Limuru Town was well known in Kenya for tea production. Tea was among the most important export products in Kenya. The relatively higher levels of OCPs contamination could arise from historical and current applications of pesticides in tea plantations. In addition, the OCPs contaminated soils might cause OCPs residues in the tea leaves. Compared with the soil OCPs concentrations in literature (Table 2), the OCPs contamination levels in Kenya was close with those in China (mean value of 12.20  $\mu\text{g kg}^{-1}$  dw) and Brazil (mean value of 6.08  $\mu\text{g kg}^{-1}$  dw) (Rissato et al., 2006; Yu et al., 2013), lower than those in India (0.36–104.50  $\mu\text{g kg}^{-1}$  dw) and Uganda (mean value of 84.60  $\mu\text{g kg}^{-1}$  dw) (Singh et al., 2007; Ssebugere et al., 2010).

The compositions of OCPs residues in soils were illustrated in Fig. 3. The OCPs were dominated by DDTs and endosulfans in most sites. Heptachlor was not considered as a predominant OCPs species because of its low detection frequencies at all sites (Table 1). Concentrations of HCHs were rather low at all investigated sites,

accounting for less than 15% of total OCPs. The isomer ratio of  $\alpha$ -HCH and  $\gamma$ -HCH was generally adopted as an indicator of the HCHs sources. When  $\alpha$ -HCH/ $\gamma$ -HCH is less than 1, HCHs may be originated from the use of lindane. When  $\alpha$ -HCH/ $\gamma$ -HCH is between 3 and 7, HCHs may be related with the input of technical HCHs (Jiang et al., 2009). Fig. S1 showed that at most sites, HCHs were dominantly lindane originated. This finding can be supported by the fact that five isomers of HCHs were banned at 1986, whereas lindane was not banned until 2011 (PCPB, 2015). And there were still 40,000 kg of lindane imported during 2001 and early 2004 according to the record of Kenyan Pesticides Control Product Board (PCPB, 2004–2005).

DDTs accounted for 20–50% of the total OCPs contents. The highest DDTs level was at Limuru town with a mean concentration of 12.49  $\mu\text{g kg}^{-1}$  dw.  $p,p'$ -DDT dominated among DDTs species at all sites. The ratios of (DDD + DDE)/ $\Sigma$ DDTs were generally less than 0.5 (Fig. S2), which implied new input of  $p,p'$ -DDT (Luo et al., 2004). It was reported that approximate 70 tons of DDT was used annually before it was banned for agricultural use in 1986 in Kenya. DDTs are still in use for the public health, for instance, to control malaria. Unfortunately, DDT are still available on the market despite the official ban, due to the insufficient awareness of the hazardous effect of the chemical. In a case, for example, a total of 1338 kg DDT was still held by Kenyan farmers (Kenya NIP, 2007). In addition, 13,800 L of dicofol, which contains DDTs as formulation impurities (Qiu et al., 2005), were imported during 2001 and early 2004 (PCPB, 2004–2005). Therefore, the local government may seek for safer alternatives of DDT for malaria control, strictly ban DDT usage and restrict dicofol import.

Endosulfan had been heavily used in Kenya against cotton boll-borer and maize stalk-borer until 2011. The use of endosulfan and its isomers was banned in 2011 (PCPB, 2015). During 2006, 2010, 17,480 tons of endosulfan were imported into Kenya (PCPB, 2009–2010). Commercial technical endosulfan consists of  $\alpha$ -endosulfan (70%) and  $\beta$ -endosulfan (30%) (Sun et al., 2014).  $\alpha$ -/ $\beta$ -endosulfan isomers can be easily transformed into endosulfan sulfate by microbes in the soil, which is difficult to be further

**Table 1**  
The concentrations of 12 OCPs in soils from Kenya ( $\mu\text{g kg}^{-1}$  dw).

	Mai Mahiu (n = 16)			Narok (n = 13)			Mount Suswa Conservancy (n = 14)			Juja (n = 10)			Limuru (n = 6)		
	Range	Mean <sup>a</sup>	Det. Freq.	Range	Mean <sup>a</sup>	Det. Freq.	Range	Mean <sup>a</sup>	Det. Freq.	Range	Mean <sup>a</sup>	Det. Freq.	Range	Mean <sup>a</sup>	Det. Freq.
$\alpha$ -HCH	n.d.–1.30	0.38	88%	n.d.–1.62	0.40	85%	n.d.–0.37	0.10	71%	n.d.–0.09	0.07	50%	n.d.–0.19	0.13	67%
$\beta$ -HCH	n.d.	n.d.	0%	n.d.	n.d.	0%	n.d.	n.d.	0%	n.d.	n.d.	0%	n.d.	n.d.	0%
$\gamma$ -HCH	n.d.–1.31	0.36	94%	n.d.–1.50	0.39	85%	n.d.–0.36	0.21	71%	n.d.–0.32	0.11	80%	n.d.–0.58	0.37	67%
$\delta$ -HCH	n.d.–3.17	0.94	88%	n.d.–4.26	1.03	92%	n.d.–1.07	0.57	79%	n.d.–1.18	0.61	90%	0.01–1.77	0.61	100%
$\Sigma$ HCHs	n.d.–5.78	1.50	94%	n.d.–7.38	1.62	92%	n.d.–1.54	0.66	93%	n.d.–1.36	0.67	90%	0.01–2.54	0.95	100%
$p,p'$ -DDE	n.d.–1.84	0.35	81%	0.04–1.62	0.49	100%	n.d.–1.45	0.30	71%	n.d.–1.30	0.40	70%	0.62–1.74	0.97	100%
$p,p'$ -DDD	n.d.–1.18	0.40	81%	n.d.–1.72	0.37	69%	n.d.–0.36	0.23	50%	n.d.–3.31	0.51	80%	0.14–4.87	1.71	100%
$p,p'$ -DDT	n.d.–5.51	3.13	94%	1.53–9.13	3.96	100%	n.d.–5.36	1.65	93%	n.d.–15.99	4.54	60%	n.d.–23.43	11.76	83%
$\Sigma$ DDTs	n.d.–6.72	3.51	94%	1.88–11.33	4.71	100%	0.40–5.92	1.86	100%	n.d.–20.60	3.42	90%	3.07–30.04	12.49	100%
Heptachlor	n.d.–11.14	6.73	25%	n.d.–7.30	3.35	54%	n.d.–4.31	1.75	29%	n.d.–1.32	1.15	30%	n.d.–0.65	0.65	17%
Heptachlor epoxide	n.d.–0.80	0.29	75%	n.d.–0.45	0.21	31%	n.d.–0.21	0.08	57%	n.d.–0.85	0.31	70%	0.37–1.31	0.63	100%
$\alpha$ -endosulfan	n.d.–0.47	0.31	75%	n.d.–0.53	0.20	92%	n.d.–0.35	0.18	50%	n.d.–1.35	0.34	80%	0.44–1.91	0.93	100%
$\beta$ -endosulfan	n.d.–0.91	0.47	69%	0.13–2.56	0.85	100%	n.d.–1.27	0.50	43%	n.d.–1.32	0.74	20%	0.31–1.86	1.29	100%
Endosulfan sulfate	n.d.–5.46	2.24	88%	n.d.–6.07	2.41	92%	n.d.–2.36	1.18	86%	n.d.–10.32	2.55	90%	3.48–12.18	7.91	100%
$\Sigma$ Other OCPs	n.d.–7.10	2.73	88%	0.45–7.63	3.32	100%	0.04–3.70	1.36	100%	n.d.–13.85	2.92	90%	4.99–17.17	10.76	100%
$\Sigma$ OCPs	n.d.–14.22	7.74	94%	2.91–23.93	9.65	100%	2.01–7.77	3.88	100%	0.10–35.41	7.01	100%	10.61–49.74	24.20	100%

n.d. = not detected. The concentrations below the LODs were reported as not detected.

Det. Freq. = detection frequency.

<sup>a</sup> The concentrations below the LODs (n.d.) were not included when calculating the mean values.

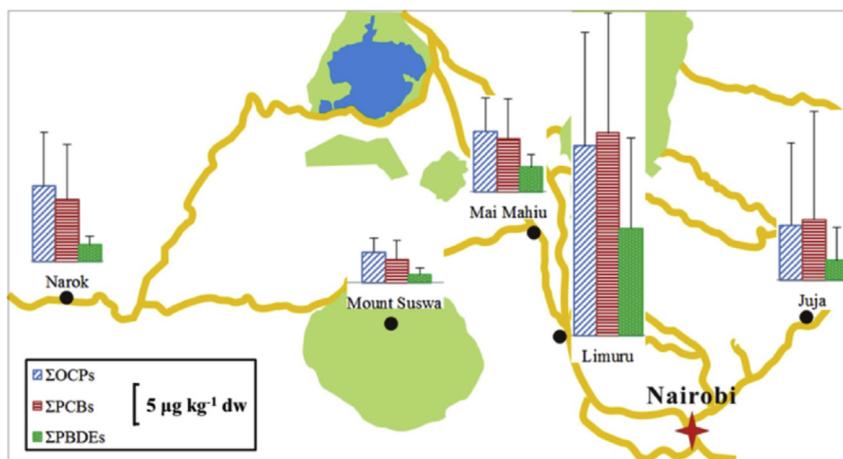


Fig. 2. Spatial distribution of OCPs in soils around Nairobi, Kenya.

Table 2 Comparison of OCPs concentrations ( $\mu\text{g kg}^{-1} \text{ dw}$ ) with those reported in the literature.

Location	Sampling year	Compounds	Concentration	Reference
<b>OCPs</b>				
Kenya	2015	12 OCPs	n.d.–49.74 (8.80) <sup>a</sup>	Present study
Pearl River Delta, China	2006	3 DDTs + 4 HCHs	(12.20)	(Yu et al., 2013)
Gangetic Alluvium, India	2003	21 OCPs	0.36–104.50	(Singh et al., 2007)
São Paulo State, Brazil	2005	12 OCPs	(6.08)	(Rissato et al., 2006)
Kihiihi, Uganda	not available	9 OCPs	(84.60)	(Ssebugere et al., 2010)
<b>PCBs</b>				
Kenya	2015	7 PCBs	n.d.–55.49 (8.23) <sup>a</sup>	Present study
Europe	1998–2005	29 PCBs	0.047–97 (7.5)	(Li et al., 2010)
North America	1998	29 PCBs	0.11–25 (4.3)	
South America	1998	29 PCBs	0.061–9.5 (1.4)	
Asia	1998–2005	29 PCBs	0.12–2.9 (0.58)	
Africa	1998	29 PCBs	0.094–0.62 (0.39)	
Australia	1998	29 PCBs	0.14–0.54 (0.28)	
<b>PBDEs</b>				
Kenya	2015	7 PBDEs	0.19–35.64 (3.40) <sup>a</sup>	Present study
Pearl River Delta, China	2002–2005	9 PBDEs	0.13–3.81 (1.02)	(Zou et al., 2007)
Indus River Basin, Pakistan	not available	8 PBDEs	0.047–2.377 (0.272)	(Ali et al., 2015)
UK & Norway	1998	20 PBDEs	0.065–12.000	(Hassanin et al., 2004)
Izmir, Turkey	2004–2005	7 PBDEs	0.504–2840	(Cetin and Odabasi, 2007)

<sup>a</sup> Minimum-maximum (mean): calculated with OCPs concentrations at all the five sites.

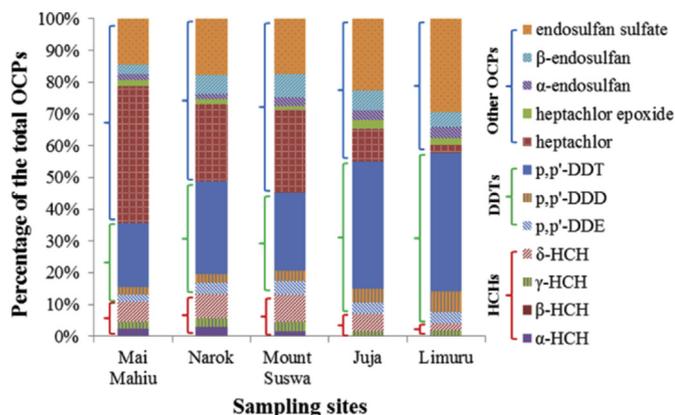


Fig. 3. Composition profiles of OCPs in soils from Kenya, Eastern Africa.

degraded (Qiao et al., 2010). In the present study, endosulfan sulfate accounted for greater than 60% of the total endosulfans ( $\alpha$ -/ $\beta$ -endosulfan and endosulfan sulfate) at all sites. However, it is difficult to sort out whether new input of endosulfan exists.

Nevertheless, it is still important for the government to monitor endosulfan residues.

### 3.2. Profiles of PCBs contamination and possible sources

Seven indicator PCBs are identified as priority food contaminants by Global Environmental Monitoring System/Food (WHO Regional Office for Europe, 2004) because they are ubiquitously present in various environmental matrices and account for more than 50% of the total non-dioxin-like PCBs (European Food Safety Authority, 2005; Liu et al., 2011). The total concentrations of 7 PCBs in Kenyan soils were in the range of n.d.–17.80, 1.99–19.99, 0.04–9.64, 0.09–46.00 and 12.82–55.49  $\mu\text{g kg}^{-1} \text{ dw}$  for Mai Mahiu, Narok, Mount Suswa Conservancy, Juja and Limuru Town, respectively (Table 3). Similar with the case for OCPs, the highest  $\Sigma\text{PCBs}$  concentrations were also found at Limuru Town (mean value 25.84  $\mu\text{g kg}^{-1} \text{ dw}$ ), while the lowest  $\Sigma\text{PCBs}$  concentrations were also detected at Mount Suswa Conservancy, with mean value of 2.95  $\mu\text{g kg}^{-1} \text{ dw}$  (Fig. 2). Limuru was home to the largest shoe factory in East and Central Africa, and there was a railway station on the Uganda Railway. The PCBs were possibly from the historical use of PCBs containing equipment for power supplies. According to the

**Table 3**  
The concentrations of 7 PCBs and 7 PBDEs in soil samples from Kenya ( $\mu\text{g kg}^{-1}$  dw).

	Mai Mahiu (n = 16)			Narok (n = 13)			Mount Suswa Conservancy (n = 14)			Juja (n = 10)			Limuru (n = 6)		
	Range	Mean <sup>a</sup>	Det. Freq.	Range	Mean <sup>a</sup>	Det. Freq.	Range	Mean <sup>a</sup>	Det. Freq.	Range	Mean <sup>a</sup>	Det. Freq.	Range	Mean <sup>a</sup>	Det. Freq.
PCB-28	n.d.–4.07	0.92	88%	0.28–9.83	1.46	100%	n.d.–0.69	0.45	71%	n.d.–0.26	0.12	40%	n.d.–0.19	0.19	17%
PCB-52	n.d.–4.68	1.46	63%	n.d.–7.86	2.30	62%	n.d.–2.47	0.54	79%	n.d.–2.55	1.06	90%	0.38–6.85	2.43	100%
PCB-101	n.d.–4.69	1.97	81%	0.33–6.15	1.86	100%	n.d.–2.88	1.18	50%	n.d.–11.49	3.04	60%	3.34–13.83	6.66	100%
PCB-118	n.d.–2.63	0.87	81%	0.09–1.89	0.68	100%	n.d.–2.10	0.74	43%	n.d.–6.06	1.17	70%	1.17–9.72	3.82	100%
PCB-138	n.d.–2.25	0.66	88%	n.d.–1.38	0.47	77%	n.d.–1.44	0.42	64%	n.d.–4.81	1.05	80%	n.d.–2.77	1.88	33%
PCB-153	n.d.–4.80	1.82	81%	n.d.–4.76	1.57	92%	n.d.–2.68	0.90	71%	n.d.–12.70	2.39	90%	4.04–21.95	10.35	100%
PCB-180	n.d.–2.08	0.77	94%	n.d.–2.03	0.79	85%	n.d.–0.70	0.49	79%	n.d.–8.13	1.56	70%	1.26–3.13	1.92	100%
$\Sigma$ PCBs	n.d.–17.80	6.81	94%	1.99–19.99	7.89	100%	0.04–9.64	2.95	100%	0.09–46.00	7.73	100%	12.82–55.49	25.84	100%
BDE-28	n.d.–1.92	0.70	81%	0.10–1.37	0.57	100%	n.d.–1.32	0.55	43%	n.d.–3.64	0.77	70%	0.86–5.81	2.43	100%
BDE-47	n.d.–2.79	1.03	81%	0.36–2.22	0.87	100%	n.d.–0.77	0.36	86%	n.d.–2.45	0.50	90%	0.69–3.84	1.80	100%
BDE-99	n.d.–1.23	0.55	94%	n.d.–1.67	0.55	69%	n.d.–0.53	0.26	79%	0.05–5.46	0.88	100%	1.22–14.02	4.31	100%
BDE-100	n.d.–1.01	0.45	88%	n.d.–0.66	0.17	62%	n.d.–0.40	0.11	64%	n.d.–2.04	0.43	90%	0.83–11.88	4.36	100%
BDE-153	n.d.–1.29	0.51	50%	n.d.–0.21	0.14	15%	n.d.–0.13	0.07	36%	n.d.–0.16	0.07	80%	n.d.–1.58	0.56	50%
BDE-154	n.d.–0.84	0.35	63%	n.d.–0.15	0.15	8%	n.d.–0.10	0.08	29%	n.d.–0.51	0.18	80%	n.d.–1.10	1.10	17%
BDE-183	n.d.–1.25	0.49	81%	n.d.–0.35	0.26	92%	n.d.–0.35	0.19	86%	0.01–0.23	0.08	100%	0.09–0.62	0.28	100%
$\Sigma$ PBDEs	0.93–6.60	3.15	100%	1.12–4.20	2.19	100%	0.19–3.13	1.03	100%	0.25–14.12	2.54	100%	4.68–35.64	13.65	100%

n.d. = not detected. The concentrations below the LODs were reported as not detected.

Det. Freq. = detection frequency.

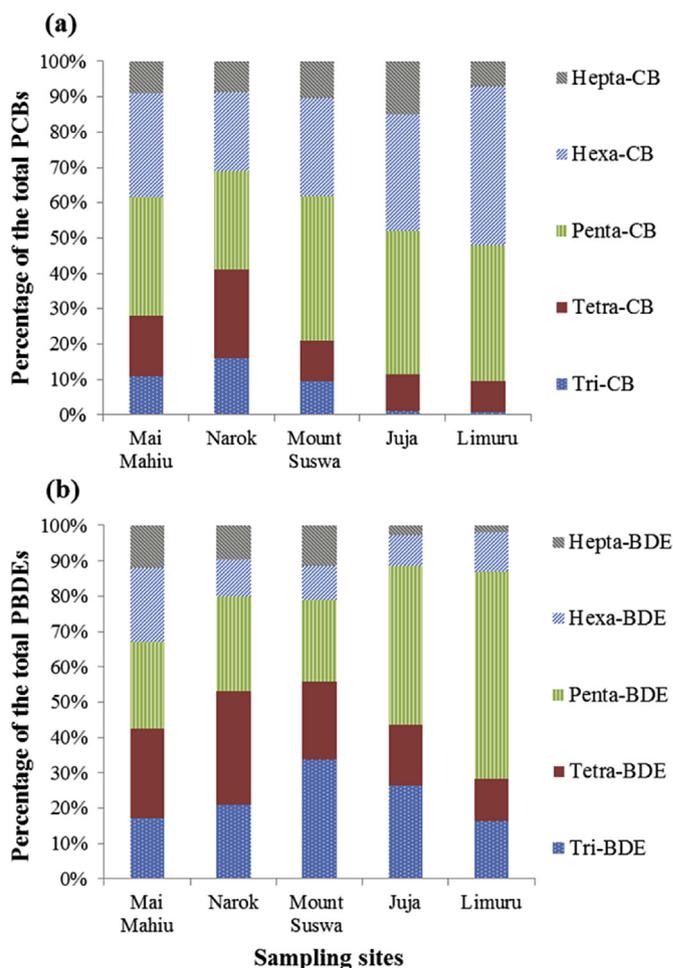
<sup>a</sup> The concentrations below the LODs (n.d.) were not included when calculating the mean values.

previous report on global PCBs levels in background soils (Table 2), the  $\Sigma$ PCBs concentration at the control site Mount Suswa Conservancy was higher than that in South America, Asia, Australia and Africa, but lower than that in Europe and North America (Li et al., 2010). PCBs contamination may be therefore of concern in the studied area of the present study.

Fig. 4a shows the composition of PCBs homologs detected, which the pattern of homolog dominance was penta-CB > hexa-CB > tetra-CB > tri-CB. This pattern was consistent with some earlier reports, for example, the surface sediment of Lake Victoria (Omwoma et al., 2015) and the breast milk samples of Kenyan mothers (Kenya NIP, 2014). In Kenya, the most important sources of PCBs contamination was believed to be the leaking electrical transformers and capacitors that were manufactured and imported before 1985. The PCBs isomer profile in PCBs polluted transformer oil was similar with that found in the present study (Takasuga et al., 2006). In 1985, lots of leaking capacitors were buried at the Juja substation near Nairobi, polluting a large area of land (Kenya NIP, 2007). It is widely believed that the transformer oil was generally sought after by the public as coolant or ointment (Kenya NIP, 2007). Open burning of PCBs-containing wastes and improper dismantling of e-waste for valuable metal recycle over open fire and acid bath have also been recognized as emerging sources of PCBs release (Gioia et al., 2014). According to the request of Stockholm Convention, Kenya has to eliminate the use of PCBs-containing equipment by 2025, and establish the detailed inventory of PCBs sources (UNEP, 2009). Thus, efforts are required to identify, label and dispose equipment containing PCBs >50 ppm, which is the limit that USEPA specified above which the material is considered to be PCB bulk product waste (USEPA, 1998). In addition, efforts are needed to determine the location of unmarked graves of obsolete transformers; to minimize open burning of wastes; and to monitor the PCBs levels in the environment.

### 3.3. Profiles of PBDEs contamination and possible sources

The seven PBDEs determined in this work were defined as “PBDEs of primary interest” by the USEPA Method 1614. The total concentrations of the 7 PBDEs in soils fell in the range of 0.93–6.60, 1.12–4.20, 0.19–3.13, 0.25–14.12 and 4.68–35.64  $\mu\text{g kg}^{-1}$  dw at the sampling sites Mai Mahiu, Narok, Mount Suswa Conservancy, Juja



**Fig. 4.** Composition profiles of (a) PCBs and (b) PBDEs in soils from Kenya, Eastern Africa. (Tri-CB = PCB-28, Tetra-CB = PCB-52, Penta-CB = PCB-101 + PCB-118, Hexa-CB = PCB-138 + PCB-153, Hepta-CB = PCB-180; Tri-BDE = BDE-28, Tetra-BDE = BDE-47, Penta-BDE = BDE-99 + BDE-100, Hexa-BDE = BDE-153 + BDE-154, Hepta = BDE-183).

and Limuru Town, respectively (Table 3). The  $\Sigma$ PBDEs levels at Limuru Town was significantly higher than the other sites, with

mean value of  $13.65 \mu\text{g kg}^{-1} \text{ dw}$  (Fig. 2), perhaps related with the local shoe factory production, considering that PBDEs were used as additives in resins, rubbers, foams and textiles (Asante et al., 2011). As expected, the  $\Sigma\text{PBDEs}$  levels at Mount Suswa Conservancy was the lowest, with mean value of  $1.03 \mu\text{g kg}^{-1} \text{ dw}$ . The PBDEs present at this site seemed to arise from the transportation of contaminants from other places, for example, through aerosol condensation and deposition (Ali et al., 2015; Cetin and Odabasi, 2007). Compared with the previous reports on PBDEs levels in other regions of the world (Table 2), the  $\Sigma\text{PBDEs}$  concentration in Kenya was higher than those in China (Zou et al., 2007), Pakistan (Ali et al., 2015), UK and Norway (Hassanin et al., 2004), but lower than that in Turkey (Cetin and Odabasi, 2007).

The homolog profiles of PBDEs in soils were shown by Fig. 4b, and the composition patterns differed slightly among the five sampling sites. For Juja and Limuru Town, the homolog dominance was penta-BDE > tri-BDE > tetra-BDE > hexa-BDE > hepta-BDE, whereas the proportion of tetra-BDE increased at Mai Mahiu and Narok. Tri-BDE was the dominant homolog at Mount Suswa Conservancy, which might be interpreted that the less brominated homolog are more volatile and transportable (Gouin et al., 2004). However, it is hard to elucidate the exact cause of the different homolog patterns because Kenya was not a producer but an importer of PBDEs-containing products, and detailed information about their production are not available (Kenya NIP, 2014). Nevertheless, the leakage during robust dismantling and incineration of e-waste was considered as an important source of PBDEs in Kenya. It is estimated that 7350 tons of e-waste were generated annually, including fridges, televisions, computers and mobile phones, which would release 143.19 tons of PBDEs into the environment per year (Kenya NIP, 2014). Measures need to be taken to regulate the recycling of e-waste, for example, to promote public awareness of e-waste, to develop an organized collection and disposal system, and to raise funds for e-waste management (Waema and Mureithi, 2008).

#### 3.4. Relationship between OCs concentrations and TOC in soils from Kenya

The Pearson correlations between soil TOC, concentrations of

$\Sigma\text{HCHs}$ ,  $\Sigma\text{DDTs}$ ,  $\Sigma\text{other OCs}$ ,  $\Sigma\text{PCBs}$  and  $\Sigma\text{PBDEs}$  were performed (Table S2). TOC was not significantly related with any of the pollutants except at Narok, where significant positive relationship was found between TOC and  $\Sigma\text{HCHs}$ , as well as  $\Sigma\text{DDTs}$  ( $p < 0.05$ ). This might imply that HCHs and DDTs were mainly historical residues at Narok, whereas at the other sampling sites, OCs have been introduced into the soils continuously, and the equilibrium partition of OCs between soil and other environmental compartments such as air and water has not been well established (Nam et al., 2008). The total concentrations of HCHs were positively related with  $\Sigma\text{DDTs}$  at Mai Mahiu ( $r = 0.602$ ,  $p < 0.05$ ) and Narok ( $r = 0.810$ ,  $p < 0.01$ ), indicating their common sources as agricultural pesticides. The total concentrations of other OCs (mainly endosulfans) was positively correlated with  $\Sigma\text{DDTs}$  at Juja ( $r = 0.953$ ,  $p < 0.01$ ). Considering that endosulfan was not banned for agricultural use until 2011, DDTs (perhaps in form of dicofol) were probably used together with endosulfans at Juja before 2011. The total concentrations of PCBs were found to be positively correlated with both  $\Sigma\text{DDTs}$  and  $\Sigma\text{other OCs}$  except at Limuru Town ( $p < 0.05$ ), which seemed difficult to explain. As expected, the positive correlation between  $\Sigma\text{PCBs}$  and  $\Sigma\text{PBDEs}$  was obtained at most sites ( $r > 0.58$ ,  $p < 0.05$ ) except at Narok, suggesting similar sources and processes for PCBs and PBDEs, for example, open burning of waste and dismantling of e-waste (Gullett et al., 2010; Leung et al., 2006).

#### 3.5. Human health risk assessment

The total lifetime carcinogenic risk (TLCR) of pollutants in the Kenyan soils through ingestion, inhalation and dermal contact was shown in Fig. 5a. PBDEs were not included because of lack of data such as the chronic oral slope factor. As can be seen, the TLR at all the five sampling sites fell in the range of  $10^{-7}$ – $10^{-6}$ , but did not exceed the target risk level of  $10^{-6}$ , indicating that the lifetime cancer risk was acceptable, but not low enough. PCBs accounted for more than 90% of the TLR at all sites, due to both the high contamination levels and toxicity of PCBs. The non-carcinogenic risk was quantified in the form of the total non-carcinogenic hazard quotient (TnHQ) for child, as illustrated by Fig. 5b. TnHQ at most sites was between 0.01 and 0.06, close to the target level of 0.1

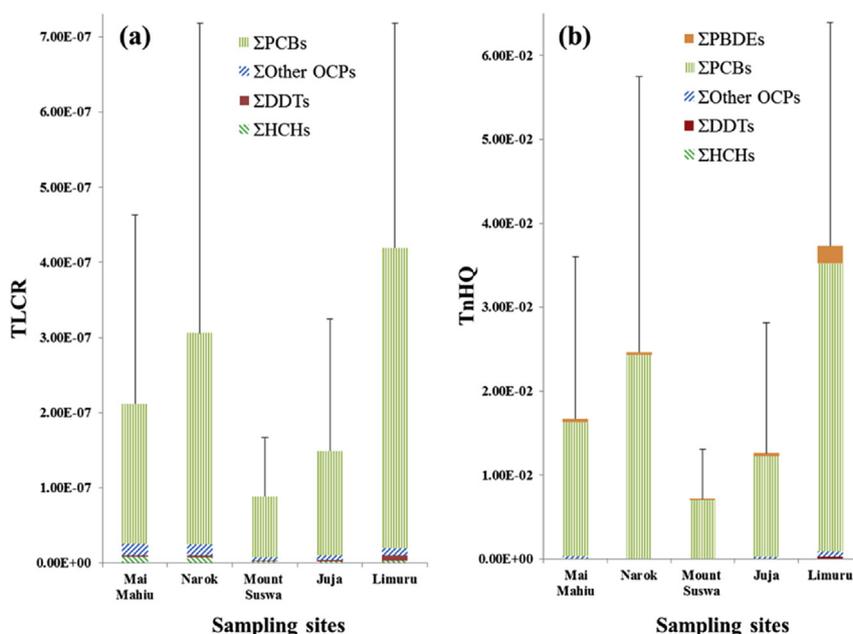


Fig. 5. The human health risk assessment of detected pollutants in soils from Kenya. (a) the total lifetime carcinogenic risk (TLCR) and (b) the total non-carcinogenic hazard quotient (TnHQ) through ingestion, inhalation and dermal contact exposure routes.

suggested by USEPA (2015), which might imply that the chronic non-carcinogenic risk of PCBs in soils was not low enough for the safety of children. Similarly with the case in TPCR, most content of the TnHQ was also contributed by PCBs at all sites. Therefore, the local authorities should make best efforts to reduce PCBs contamination in soil, as suggested in section 3.2.

#### 4. Conclusion

The soils in Kenya were contaminated by OCs, especially at the Limuru Town site. New input of DDTs was still present, possibly due to the insufficient execution of the ban and application of DDTs for malaria control. High residue levels of endosulfans as well as DDTs in soils also called for the exploration of their alternatives both in agricultural and public health applications. The total concentrations of PCBs residue reached a mean of  $8.23 \mu\text{g kg}^{-1}$  dw, which accounted for most proportion of the human health risk posed by all the detected pollutants. The soils were less contaminated by PBDEs, and health risk caused by PBDEs was negligible. Therefore, the local authorities should focus on the PCBs pollution when implementing the Stockholm Convention recommendation, including accomplishing the detailed inventory of PCBs sources and capacity building for sustainable PCBs management.

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#### Appendix B. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2015.11.040>.

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