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Cancer risk assessment of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) in former agricultural soils of Hong Kong

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ABSTRACT

The major objective of this study was to evaluate the carcinogenic risk posed to humans through PBDEs and PCBs of changing agricultural land use for recycling of e-waste and open burning of municipal waste. Nine locations were selected to represent 6 different types of land use such as e-waste dismantling workshop (EW (DW)) and e-waste open burning site (EW (OBS)). The total concentrations for PBDEs and PCBs, and the bioaccessibility of PCBs were determined using Soxhlet extraction and *in vitro* simulated gastric solution, respectively. Both total and bioaccessible concentrations were subsequently used to establish the cancer risk probabilities in humans via ingestion, dermal contact and inhalation of soil particles. It was found that very low cancer risk in all 6 types of different land use was caused by BDE-209. Nevertheless, at the 95th centile, the concentration of PCBs in EW (DW) and EW (OBS) indicate a low cancer risk to humans of 40 and 2.1 in a million, respectively, while the same was also observed for the bioaccessible PCBs in EW (DW) of 1.71 ± 2.96 in a million.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) are both persistent organic pollutants (POPs), which are commonly found at e-waste recycling sites [1,2]. Polybrominated diphenyl ethers and PCBs tend to readily accumulate in the fats of organisms and get passed along the food chain due to their high lipophilicity [3,4]. For example, the former were detected in Indo-Pacific humpback dolphins (*Sousa chinensis*) and finless porpoises (*Neophocaena phocaenoides*) in Hong Kong [5], while the latter were identified in human milk [6]. Polybrominated diphenyl ethers are believed to act as endocrine disruptors that affect hormone regulation [7]. It has been shown that BDE-209, the major ingredient of commercial brominated flame retardants can cause neurobehavioral derangements in adult mice [8]. Animal studies also revealed that PBDEs can cause other health problems such as thyroid hormone disruption, and possibly cancer [9–11].

Polybrominated diphenyl ethers are commonly used as flame retardants, while PCBs are components of transformers and capacitors, as well as, hydraulic and heat exchange fluids [12], which may explain why they can always be found at electronic waste (e-waste) recycling sites [1,2]. In Hong Kong, there are sites for e-waste recycling and open burning that represent a significant number of potential releasing sources of PBDEs and PCBs. These sites are former agricultural lands in which their initial purpose has been changed to other forms of land use. The reasons behind this may be clarified by the burgeoning urbanization leading to the disappearance and fragmentation of large areas of farmland, which in turn depreciated the value of the land and thus further exacerbating the problem. Large-scale cultivation was no longer feasible after the "critical mass" of farmland had been destroyed [13,14]. In the 1980s, both abandoned and existing farmlands were dramatically converted for other purposes, directly as a result of rapid economic development and fragmentation [13,15,16]. Consequently, substantial areas of agricultural land were changed to greater profit generating applications such as storage sites (for container), car dismantling workshops, and more recently, for the storing, dismantling, recycling, and open burning of e-waste.

Most of the previous studies focused on the concentrations of PBDEs and PCBs in biota samples [17–19], where no studies relating to their concentrations in former agricultural soils of Hong Kong could be found. Therefore, it was crucial to perform an investigation on estimating the risks and potential health effects of these POPs in soils after land use changes in Hong Kong. The health risks exerted on humans was generally overestimated in other studies when risk assessments were conducted by means of total pollutant

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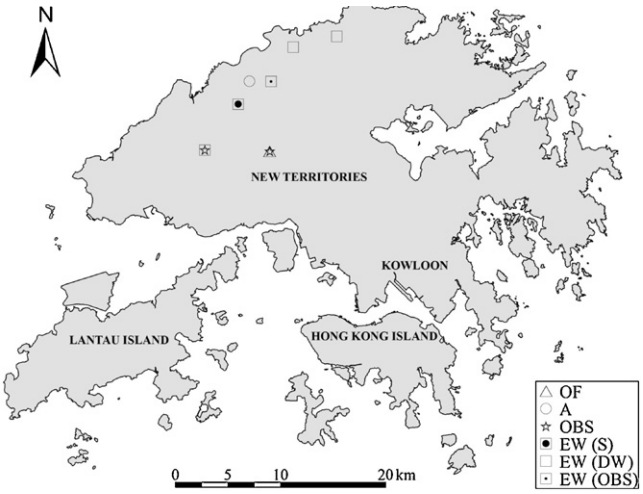

Fig. 1. The location of 9 soil sampling sites in Hong Kong. OF = organic farm, A = agricultural, EW (S) = e-waste storage, OBS = open burning site, EW (DW) = e-waste dismantling workshop, EW (OBS) = e-waste open burning site and CDW = car dismantling workshop.

concentrations [20]. Therefore, this study included the analysis of bioaccessible PBDE and PCB concentrations to combat this issue, thus ensuring that the resultant cancer risk assessment presented was a realistic portrayal of events.

The objectives of this study were to determine the concentrations of PBDEs and PCBs, especially their bioaccessible concentrations in current and former agricultural soils of Hong Kong, as well as, to conduct a human health risk assessment on cancer, in order to evaluate the potential risks based on their concentrations within soils.

2. Materials and methods

2.1. Sampling, preparation and analysis

The PBDE and PCB concentrations were previously reported in a study by Lopez et al. [21], where its major difference with this study is that it involved the addition of 2 open burning sites, in which bioaccessible PCB concentrations were analyzed and the pollutant concentrations subsequently used for human cancer risk evaluation. Fig. 1 briefly shows the location of the 9 sampling sites, whereby a total of 45 composite soil samples were collected from these sites within the existing and former agricultural lands, located to the north west of the New Territories in Hong Kong. The sampling sites were grouped into 6 soil types according to their current land use: agricultural (A); organic farm (OF); e-waste storage (EW (S)); e-waste dismantling workshop (EW (DW)); e-waste open burning site (EW (OBS)); and open burning site (OBS). Descriptions of each type of land use and their number of sites are given in Table 1. The size of each location totaled approximately 100 m × 100 m, which were divided into 5 equal sub-areas, where each had 5 random soil samples (0–5 cm, 0.5 kg) assembled using a stainless steel spade. Consequently, leading to the formation of individual composite samples, hence, each site contained 5 of these. The soil samples from EW (S) were taken from the surrounding agricultural area as the floor of the EW (S) was concerted. Half of the soil samples were air-dried for two weeks, while the other half was freeze-dried for at least two weeks and sieved through a 2-mm mesh. Soil texture and soil organic matter (SOM) were determined by the Bouyoucos Soil Hydrometer Method [22] and the method for analyzing the total organic carbon (TOC) in soils and sediments [23], respectively. Every 5th soil sample was tested in duplicate to check the consistency of the data.

2.2. The extraction and analysis of total PBDEs and PCBs

The extraction of PBDEs and PCBs from the soil samples was conducted by using the Standard Method 3540C [24]. Five grams (g) of each soil sample was spiked with PCB standard solutions (28 PCB congeners mixture, Standard Reference Materials Group of NIST, USA), native ($^{12}C_{12}$) PBDE solution/mixture (BDE-MXE) for precision and recovery (PAR) and mass-labelled ($^{13}C_{12}$) solution/mixture (MBDE-MXE) (Wellington Laboratories Inc., Canada) for PBDE recoveries. The samples were then Soxhlet extracted with 150 mL acetone (pesticide grade, Tedia), dichloromethane (DCM) (pesticide grade, Tedia) and n-hexane mixture (1:1:1, v:v) in a 65 °C water bath for 18 h with 3 g of anhydrous sodium sulphate for moisture removal during extraction. One g of activated copper granules (Riedel-de Haën) was added to remove the sulphur during the process (activated copper granules was prepared by mixing with hydrochloric acid (1 N) then washed with distilled water and DCM). The extracts were then concentrated to roughly 1 mL by a rotary evaporator, which were subsequently cleaned up by the standard clean up method 3620B [25]. Extracts were briefly eluted with a 20 mL (7:3, v:v) mixture of n-hexane (95%, pesticide grade, Tedia) and DCM through a column packed with 5 g of anhydrous sodium sulphate and 8 g of florisil. The extracts were reduced to around 1 mL by a rotary evaporator post-clean up stage and allowed to further evaporate in the fume hood until the volume reached 200 μL.

GC–MS analysis was performed on a Hewlett Packard 6890 GC system equipped with a mass selective detector and a 30 m × 0.25 mm × 0.25 μm DB-5 capillary column (J & W Scientific Co. Ltd., USA). The Standard Method 8270 C [26] was adopted for the verification of the following 37 PCB congeners including 7 PCB indicators (PCB-28, -52, -101, -118, -138, -153 and -180). In addition, the concentrations of 22 PBDEs were determined using Shimadzu QP2010 GC/MS, following the methods described in [27], with a slight modification by Zheng [28]. The low molecular weight PBDE congeners (BDE-3 to -191) were measured with a DB-1 (30 m × 0.25 mm i.d. ×0.25 μm) column, whereas a short column (12 m) was used for high molecular weight BDE congeners (BDE-197, -196, -207, -206 and -209).

Table 1
Descriptions of different agricultural land uses and their respective number of sites under investigation.

Different types of agricultural land use	Number of sites	Site description
Organic farm	1	Vegetables are grown without the use of chemical fertilizers and pesticides in which nutrients are recycled within the cultivated area
Agricultural	1	Traditional farming system and the application of legal chemical fertilizers and pesticides are allowed
E-waste storage	1	Electronic waste storage sites with concrete flooring surrounded by concrete walls
Open burning site	2	Existing agricultural land using small areas to burn bulky woody furniture, household waste and wild grass
E-waste dismantling workshop	3	Breakdown of electronic components such as refrigerators, computers and printers on existing agricultural land
E-waste open burning site	1	Burning of electronic components such as refrigerators, computers, cables and printers on existing agricultural land

Table 2
Slope factors of PCBs and PBDEs via the exposure pathways of ingestion, dermal contact and inhalation [35].

PCBs and PBDEs	Slope factors for evaluating cancer risks		
	Ingestion SFO $(mg/kg/day)^{-1}$	Dermal contact SFO × GIBAS $(mg/kg/day)^{-1}$	Inhalation IUR $(mg/m^3)^{-1}$
Polychlorinated biphenyls (high risk)	2.00E+00	2.00E+00	5.70E−01
Decabromodiphenyl ether, 2,2′,3,3′,4,4′,5,5′,6,6′-(BDE-209)	7.00E−04	7.00E−04	–

SFO = oral slope factors, GIABS = gastrointestinal absorption factor, IUR = inhalation unit risk.

2.3. Quality control

A Standard Reference Material (SRM) 2585 (Organic Contaminants in House Dust) for PCBs was obtained from the National Institute of Standards and Technology (NIST, USA). The SRM and an analytical blank were included in every batch of extraction, where the mean SRM recoveries of PCBs ranged between 94 and 117%. The mean recoveries for the matrix spike of low molecular PBDEs were between 70 and 108% and ranged from BDE-3 to BDE-191, whereby the high molecular weight BDE-209 made up 51%. The detection limit, defined as a signal of three times the noise level, was 0.5 ng/g (dw) for PCBs, and 0.1 ng/g (dw) for PBDEs.

2.4. Risk characterization and estimation

Cancer risks via ingestion, dermal contact and inhalation of soil particles were estimated, based on the following Eqs. (1) and (2) [29] and Eq. (3) [30].

$$\text{Cancer risk}_{\text{ingest}} = \frac{C_{\text{soil}} \times \text{Ing}R \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times \text{CF} \times \text{SFO} \quad (1)$$

where Cancer $\text{risk}_{\text{ingest}}$ is cancer risk via ingestion of soil, C is soil concentration of the contaminant in soil (mg/kg), Ing$R$ is ingestion rate of soil (mg/day), EF is exposure frequency (days/year), ED is exposure duration (years), BW is average body weight (kg), AT is averaging time (days), CF is conversion factor (1×10^{-6} kg/mg), SFO is oral slope factor $(mg/kg/day)^{-1}$.

$$\text{Cancer risk}_{\text{dermal}} = \frac{C_{\text{soil}} \times \text{SA} \times \text{AF}_{\text{soil}} \times \text{ABS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times \text{CF} \times \text{SFO} \times \text{GIABS} \quad (2)$$

where Cancer $\text{risk}_{\text{dermal}}$ is cancer risk via dermal contact of soil, SA is surface area of the skin that contacts the soil (cm^2), AF_{soil} is skin adherence factor for soil (mg/cm^2), ABS is dermal absorption factor (chemical specific), GIABS is gastrointestinal absorption factor.

$$\text{Cancer risk}_{\text{inhale}} = \frac{C_{\text{soil}} \times \text{EF} \times \text{ED}}{\text{PEF} \times \text{AT}} \times \text{IUR} \quad (3)$$

where Cancer $\text{risk}_{\text{inhale}}$ is cancer risk via inhalation of soil particles, Inh$R$ is inhalation rate (m^3/day), IUR is inhalation unit risk $(mg/m^3)^{-1}$ = slope factor via inhalation, PEF is particle emission factor = 1.36×10^9 m^3/kg.

The PEF concerns the inhalation of pollutants adsorbed to respirable particles (PM_{10}) [31].

A conservative soil ingestion rate (Ing$R$) of 100 mg/day was recommended for adults [32], which was based on an exposure duration (ED) of 70 years (life time exposure period) and an assumed exposure frequency (EF) of 350 days/year. The EF took into account the working pattern of workers and farmers toiling the farmland all year round, in which the average time (AT) was calculated (excluding the 15 days of holiday) as (AT) = ED × 365 = 25,550 days. In this study, a body weight of 60 kg per adult worker was selected to accommodate the current local situation [33], in which the contact surface area of skin with soil (SA) was deemed as 3300 cm^2, while the skin adherence factor of soil (AF_{soil}) was 0.2 mg/cm^2 [29]. It was not recommended in the updated human health evaluation manual of 2009 for the ventilation rate and body weight to be applied in Eq. (3) [30], as "the amount of the chemical that reaches the target site of the chemical through the inhalation pathway is not the simple function of the ventilation rate and body weight". Therefore, the ventilation rate and body weight were excluded from Eq. (3).

The estimation of cancer risk via ingestion, dermal contact and soil inhalation was based on a human lifespan of 70 years, and a slope factor (SF) that can be defined as the human cancer risk per unit (mg/kg/day) dose, obtained from either animal bioassays or human data [29]. According to the Human Health Evaluation Manual [34], cancer risks can increase across different exposure pathways, but only when assessing the risks for the same individuals. Hence, the estimation of combined cancer risks through ingestion, dermal contact and inhalation may be applied in this study by adding the results of Eqs. (1)–(3) together. Furthermore, the total and bioaccessible concentrations of BDE-209 were used only to conduct the cancer risk assessment through ingestion and dermal contact of soils, as no IUR of BDE-209 was available from the US EPA [35]. In contrast, the total and bioaccessible PCB concentrations were utilized to conduct the cancer risk assessment of 3 different exposure pathways, namely ingestion, dermal contact and inhalation [35]. The SF for both BDE-209 and PCBs are listed in Table 2.

Lifetime cancer risks can be qualitatively described with the following of: very low when the estimated value is $\leq 10^{-6}$; low in the range of 10^{-6}< to <10^{-4}; moderate in the range of $10^{-4}\leq$ to <10^{-3}; high in the range of $10^{-3}\leq$ to <10^{-1}; and very high when the value is $\geq 10^{-1}$ [36].

2.5. In vitro digestion model to extract bioaccessible PBDEs and PCBs

The cancer risks from total BDE-209 were classified as very low amongst the 6 types of land use and so no analysis of bioaccessible PBDEs was completed. The bioaccessible PCBs were established by the physiologically based extraction test described in Ruby et al. [37], with a slight modification, which involved simulating the conditions of both the human stomach and intestine [38]. The gastric solution used in this study consisted of a mixture of 17.55 g of NaCl, 1.0 g of citrate, 1.0 g of malate, 0.85 mL of lactic acid, 1.0 mL of acetic acid, and 2.5 g of pepsin (P7000, Sigma Chemical Co.) into 2 L of deionised water, which was adjusted to pH 1.5 with 12 M HCl. Then, 1 g of soil was added into a 50 mL plastic centrifuge tube followed by 30 mL of gastric solution. This mixture was then shaken in a shaking incubator (SHEL LAB 1575 R) above 55 rpm for 1 h at 37 °C and subjected to simulated intestinal conditions by adjusting the pH to 7.0 with 1 M NaOH. Meanwhile, 0.06 g of porcine bile extract (B8631, Sigma Chemical Co.) and 0.018 g of porcine pancreatin (P1500, Sigma Chemical Co.) were added to each tube. These samples were also shaken with the same shaking incubator above 55 rpm for 4 h at 37 °C, during the intestinal condition simulation. Next, the samples were centrifuged at 3300 rpm for 10 min at 37 °C and filtered with an Advantec 5C filter paper. Finally, the filtrate was diluted and topped up to 35 mL with deionised water.

Bioaccessible PCBs were subsequently extracted by using the liquid–liquid extraction method. The 35 mL filtrate was mixed with 35 mL of DCM and acetone (v:v 1:1) for 2 min in a separatory funnel and left to stand for half an hour for extraction to occur. The organic sample extracts were then collected and excess anhydrous sodium sulphate was used to absorb any water left in the extracts. The sample extracts were concentrated to 1 mL by a rotary evaporator and the same cleanup and analytical procedures were applied as for the total PCBs (Section 2.2).

2.6. Selection of soil samples for the In vitro digestion model, bioaccessible PCB recoveries and risk assessment of bioaccessible PAHs

The minimum, median and maximum cancer risks of PCBs via the ingestion pathway for each land use type were chosen for determining their bioaccessible PCB concentrations: 3 (minimum, median and maximum) × 6 types of land use = 18 soil samples, in which each sample was performed in duplicates. The SRM 2585 (Organic Contaminants in House Dust) and an analytical blank were also included in every batch of extraction, and the mean bioaccessible PCB recoveries of SRM 2585 found to range from 0.71 to 29.7%. The cancer risk assessment of bioaccessible PCBs was estimated by the same method in Section 2.4.

3. Results and discussion

The two PBDE congeners namely, BDE-47 and BDE-99, attract the most public concern due to their toxicity and persistence, with reports implying that they can disturb the activities of thyroid hormones and neurobehavioral development [40,41]. Amongst the 6 sampling sites, EW (OBS) contained the highest concentration of BDE-47 (2287 μg/kg) and BDE-99 (1410 μg/kg) (Table 3). However, when the Guiyu study was compared with this study [39], it was deduced that the former site contained higher total PBDEs of 63,300 μg/kg, but lower BDE-47 of 15.4 μg/kg and BDE-99 of 24 μg/kg, than the latter site of 32,337 μg/kg, 2287 μg/kg and 1410 μg/kg, respectively (Table 3). The commercial products of PBDEs include BDE-99 (also known as penta-BDE), octa-BDE and deca–BDE. The soil of EW (OBS) and OBS contained relatively high concentrations of BDE-47 and BDE-99 than Guiyu, which may be attributed to the e-waste soil samples containing mostly BDE-99 in Hong Kong and also the degradation of BDE-99 to BDE-47 (also known as tetra-BDE). However, since only 5 samples were taken for each studied land use type, a greater number of soil samples from the EW (OBS) and OBS have to be taken for analysis in order to prove this conjecture. In addition, the US EPA imposed guidelines on PBDEs for residential (BDE-47 = 7800 μg/kg; BDE-99 = 7800 μg/kg) and industrial soils (BDE-47 = 100,000 μg/kg; BDE-99 = 100,000 μg/kg) [35], where the concentrations in the samples of the current study were below these standards. However, it is important to point out that these standards are not specific for agricultural soils but it is envisaged that more stringent guidelines should be adopted for these types of soils.

Electronic waste dismantling workshop (1061 μg/kg) contained the highest concentration of total PCBs (Table 4), which far exceeded the background surface soil from other places including Hong Kong (2.45 μg/kg) [42], China (0.515 μg/kg) [43], North America (196 μg/kg) [44] and other parts of the world (5.41 μg/kg) [45], as well as, the Canadian soil quality guideline for environmental health (500 μg/kg) [46]. Electronic waste dismantling workshop also demonstrated the most elevated total of the 6 PCB indicators and 7 PCB indicators. The total 6 PCB indicators (57.7 μg/kg) found in EW (DW)

Table 3
Summary of PBDEs concentrations in this and other studies, and the regional screening level (RSL) tables (μg/kg) taken from the US EPA [35].

Types of soil	Country/region	Sample no.	Penta	Octa	BDE-47	BDE-99	BDE-153	BDE-209	Total BDEs	Reference
Organic farm (OF)	HK, China	5	2.28	2.18	1.62	0	0	0	23.5	This study
Agricultural (A)	HK, China	5	3.02	7.07	1.77	0.714	0.902	2.24	27.5	This study
E-waste storage EW (S)	HK, China	5	14.6	0	5.67	4.99	2.06	0	50.5	This study
Open burning site (OBS)	HK, China	5	28.6	4453	12.9	9.3	14.8	55.2	28,111	This study
E-waste dismantling workshop EW (DW)	HK, China	5	375	787	384	236	117	1542	6875	This study
E-waste open burning site EW (OBS)	HK, China	5	1993	5731	2287	1410	799	5806	32,337	This study
Within e-waste storage site	HK, China		7.5–190	–	–	–	–	–	–	EPD [62]
Outside e-waste storage site	HK, China		16–89	–	–	–	–	–	–	EPD [62]
Pak Heung e-waste storage site	HK, China	1	607	–	274	478	–	4610	32,746	Greenpeace China [63]
Fanling e-waste storage site	HK, China	2	18.9–28	–	9.57–16	15.2–22.2	–	148–162	274–306	Greenpeace China [63]
Ash 1 (charred plastic parts)	Guiyu, China	1	2080	–	899	758	–	–	9224	Luksemburg et al. [64]
Ash 2 (mixture of ash, mud, sand)	Guiyu, China	1	2100	–	1350	1270	–	–	8291	Luksemburg et al. [64]
Ash 3 (mixture of ash, mud, sand)	Guiyu, China	1	2090	–	372	521	–	–	12,094	Luksemburg et al. [64]
Combusted residue	Guiyu, China	3	64.4	–	15.4	24.0	–	48,633	63,300	Leung et al. [39]
Regional screening level										
Resident soil	US EPA		120,000	180,000	7800	7800	16,000	430,000	–	US EPA [35]
Industrial soil	US EPA		120,0000	180,0000	100,000	100,000	200,000	250,0000	–	US EPA [35]

Table 4
Summary of PCB concentrations in this and other studies, and the soil quality standards (μg/kg) of VROM (2000) and CCME (1991) [46,47].

Types of soil	Country/region	Sample no.	Total 6 indicator PCBs	Total 7 indicator PCBs	Total PCBs	Reference
Organic farm	HK, China	5	1.11 (0.945–1.22)	1.27 (1.08–1.38)	3.25 (2.92–3.38)	This study
Agricultural	HK, China	5	1.42 (0.926–2.76)	1.22 (0.887–1.48)	5.96 (2.67–16.5)	This study
E-waste storage	HK, China	5	3.09 (1.25–6.68)	3.69 (1.54–8.41)	9.53 (3.5–22.8)	This study
Open burning site	HK, China	5	2.16 (N.D.-8.80)	22.3 (11.3–36.1)	37.7 (19.5–64.2)	This study
E-waste dismantling workshop	HK, China	5	57.7 (2.56–404)	64.3 (448–3.05)	1061 (7.13–14,542)	This study
E-waste open burning stie	HK, China	5	41.5 (2.51–110)	49.5 (4.55–127)	144 (2.51–110)	This study
Hong Kong surface soil	HK, China	7	2.42 (0.04–9.87)	2.45 (0.04–9.87)	2.45 (0.04–9.87)	Zhang et al. [42]
Chinese surface soil	China	52	0.0945 (N.D.–0.799)	0.110 (N.D.–1.06)	0.515 (0.138–1.84)	Ren et al. [43]
Global background surface soil	Global	191	2.47 (0.00900–51.2)	2.91 (0.00950–57.91)	5.41 (0.0260–96.6)	Meijer et al. [45]
	North America	18	173 (5.2–2839)	185 (6.9–3042)	196 (7.9–3136)	Wilcke and Amelung [44]
Soil quality standards						
Dutch Target Value	Netherlands		20			VROM [47]
Dutch Intervention Value	Netherlands			1000		VROM [47]
SQE_E	Canada				500	CCME [46]

Note: In this study: Total of 6 PCB indicators (PCB 28, 52, 101, 138, 153 (+157) and 180) and 7 PCB indicators (PCB 28, 52, 101, 118, 138, 153 (+157) and 180). Other studies: Total of 6 PCB indicators (PCB 28, 52, 101, 138, 153 and 180) and 7 PCB indicators (PCB 28, 52, 101, 118, 138, 153 and 180). SQE_E = soil quality guideline for environmental health and N.D. = not detected.

Table 5
Total cancer risks from exposure via ingestion and dermal contact of soils in humans from different types of agricultural land use, based on the total BDE-209 concentrations at the 5th, median and 95th centiles.

Sampling site	Total cancer risks from exposure via ingestion and dermal contact		
	5th centile	Median	95th centile
Organic farm	N.D.	N.D.	N.D.
Agricultural	N.D.	2.08E−05	2.08E−05
E-waste storage	N.D.	6.62E−05	3.68E−04
Open burning site	N.D.	N.D.	N.D.
E-waste dismantling workshop	N.D.	2.98E−03	5.60E−03
E-waste open burning site	N.D.	9.25E−03	1.70E−02

Note: N.D. means not detected and the values of cancer risk are in the unit of 10^{-6}.

were more than twice the Dutch Target Value (20 μg/kg) [47], indicating its potential health risk. Nonetheless, EW (DW) contained a lower total 7 PCB indicator concentration (64.3 μg/kg) than the Dutch Intervention Value (1000 μg/kg) [47]. The demolition of electronic components during e-waste dismantling activities causes the release of PCBs into the soils, which may have resulted in the detection of high PCB concentrations in EW (DW). In another e-waste dissembling site in Zhejiang, China, cable coating was revealed to contain the highest concentration of total PCBs (680 μg/kg dw) [48]. This was attributed to the addition of PCBs into polyvinyl chloride in order to improve the insulating ability of high-voltage cables [49]. Consequently, the dismantling of cables most probably will lead to high PCB concentrations in the surrounding environment.

On the other hand, significantly higher soil organic matter (SOM) was noticed in the soils of OBS, EW (DW) and EW (OBS) (6.11, 8.63 and 6.30%, respectively) (Table S1 – supplementary data). In addition, there were positive correlations between SOM and total PCBs ($r = 0.357$; $p < 0.05$) and PBDEs ($r = 0.318$; $p < 0.05$) (Table S2 – supplementary data). SOM may be able to bind the PCBs and PBDEs once they had been deposited into the soils [50,51]. The strong sorption may render the PBDEs and PCBs more resistant to degradation and leaching from soils [50,51].

Table S3 shows the supplementary data for the cancer risks via ingestion and dermal contact of soils on humans, based on the total BDE-209 concentration. The carcinogenic risks for PBDEs by combining the above exposure pathways are displayed in Table 5. Rodent experiments have illustrated that the intake of BDE-209 in contaminated food may cause liver tumors [52], and in the present study only BDE-209 was considered for cancer risk assessment, as there were no SF for other PBDEs. Furthermore, there was no SF for BDE-209 via the exposure pathway of inhalation (Table 2) due to the scarcity of inhalation toxicity factors needed to obtain IUR $(mg/m^3)^{-1}$ [53]. Therefore, the cancer risks of BDE-209 via inhalation were not taken into account for this study. The results show that the cancer risk values of the soils from all the 6 sites were below 1 in a million people at the 5th, 50th and 95th centiles (Table 5), which means that the cancer risks imposed by BDE-209 are very low.

At the 95th centile, soils from OF and EW (S) did not show any cancer risks as BDE-209 was not detected. While in the case of the other four land use types the cancer risk trend was as follows: EW (OBS) > EW (DW) > OBS > OF in both ingestion and dermal contact pathways. For both pathways, even the highest cancer risk for PBDEs of EW (OBS) (0.0102×10^{-6} for ingestion; 0.00676×10^{-6} for dermal contact), calculated to be 3 times greater than the other types of land use, and still grouped under the category of 'very low'. The higher PBDE carcinogenic risk of the soil from EW (OBS) may be due to the burning activities of e-waste, as PBDEs are commonly applied to circuit boards and as coatings of flame retardants [3]. Open burning of the PBDE containing

Table 6
Total cancer risks from exposure via ingestion, dermal contact and inhalation of soils in humans from different types of agricultural land use based on the total PCBs concentrations at the 5th, median and 95th centiles.

Sampling sites	Total cancer risks from exposure via ingestion, dermal contact and inhalation		
	5th centile	Median	95th centile
Organic farm	0.018	0.0198	0.0222
Agricultural	0.0164	0.0242	0.102
E-waste storage	0.0215	0.0418	0.145
Open burning site	0.120	0.229	0.389
E-waste dismantling workshop	0.0438	0.107	**40**
E-waste open burning site	0.0445	0.183	**2.1**

Note: Cancer risks are in bold and the values of cancer risk are in the unit of 10^{-6}.

e-waste may thus be the major cause for the release of PBDEs and its associated elevated carcinogenic risk. Positive correlations were observed between the concentrations of PBDEs in fish (tilapia and bug head) and sediment from Guiyu [54]. Since PBDEs can be bioaccumulated and biomagnified in the ecosystem [55], they may be accumulated in human bodies and cause cancer after sustained ingestion of contaminated fish. It has been clearly shown in Guiyu that PBDE contamination can affect humans and cause adverse health effects via various pathways, such as ingestion of contaminated fish, hence, PBDE pollution must not be overlooked in Hong Kong.

The carcinogenic risks for PCBs were estimated via a tiered approach, which depended on different levels of human SF for environmental PCBs, through various exposure pathways. In this study, the PCB cancer risks were estimated using tiers of high risk and persistence (a relatively high SF), based on the criteria of ingestion, dermal contact and inhalation of soil being within this tier [56]. The estimated cancer risks for PCBs are given in Table 6 with the supplementary data included in Table S4. All 6 types of land use exhibited very low carcinogenic risks at the 5th and 50th centiles, while EW (DW) and EW (OBS) showed low cancer risks through ingestion and dermal contact at the 95th centile. Higher cancer risks were found in EW (DW) (ingestion: 20.8×10^{-6}; dermal contact: 19.2×10^{-6}; inhalation: 0.000873×10^{-6}) and EW (OBS) (ingestion: 1.09×10^{-6}; dermal contact: 1.01×10^{-6}; inhalation: 4.59×10^{-11}) when compared to the other types of land uses, which may be a consequence from the release of PCBs from e-waste. PCBs are commonly detected at e-waste recycling sites since they are used in the manufacturing of capacitors and transformers [1] and the dismantling and burning activities promote its leakage into surrounding soils. For the exposure pathways, the trend of decreasing cancer risks was as follows: ingestion > dermal contact > inhalation. Inhalation presented the lowest cancer risks (EW (DW): 0.000873×10^{-6}; EW (OBS): 4.59×10^{-11}) because PCBs possess a low volatility [56].

Ingestion is a significant exposure pathway for PCBs usually due to involuntary consumption of soil. As mentioned above, cancer risks for PCBs through ingestion (EW (DW): 20.8×10^{-6}; EW (OBS): 1.09×10^{-6}) were relatively high when compared to dermal contact and inhalation. Consequently, an *in vitro* system that simulated the human digestive system, was adopted in the current study in order to investigate the amount of bioaccessible carcinogenic PCBs in the soil samples. According to Table 7, it can be observed that the cancer risk for bioaccessible PCBs from EW (DW) was significantly higher than the other types of soil (mean = $1.71 \times 10^{-6} \pm 2.96 \times 10^{-6}$), implying that the bioaccessible PCBs concentrations in EW (DW) may pose potential cancer risk in humans.

The major exposure pathway of PBDEs and PCBs to workers or farmers in OF, A, EW (S) and EW (DW) was discovered to be dermal contact. But the pathway in OBS and EW (OBS) in contrast was via inhalation, as the combustion activities in these land use types tend to generate ultra fine particles less than $PM_{0.1}$, which can penetrate deeply into the lungs and cause adverse health effects [57]. The estimation of health risks via inhalation should be based on pollutants adsorbed onto respirable particles of soils (less than PM_{10}) [31]. Only the inhaled soil particles with a size of less than PM_{10} can be deposited in the upper part of the respiratory tract or penetrate deeply into the lungs [58,59]. Fine soil particles (less than PM_{10}) with organic pollutants (such as PAHs) and inorganic pollutants (such as Cu, Cd and Zn), may be able to cause oxidative stress and inflammation after penetrating into the lungs [60,61]. This study used soil particles with a diameter of less than 2 mm to estimate cancer health risks in humans via the exposure route of inhalation, implying that not all soil particles were able to penetrate into the lungs. In addition, the concentrations of pollutants in soil particles with a diameter of less than 2 mm should be lower than the particles smaller than PM_{10}. Consequently, the human health risks based on pollutant concentrations would more than likely be underestimated. Furthermore, the absent IUR of BDE-209 causes this evaluation of cancer risks to be underestimated. There is a need to derive pollutant toxicity values based on the inhalation pathway from experimental data in order to fill the gap of risk assessment via this means especially in the OBS and EW (OBS).

Evaluating the health risks by using bioavailable pollutant concentrations is commonly regarded as the most accurate way, because only the bioavailable portion of the contaminants will ultimately reach our bloodstream and exert adverse effects on our body [20]. However, this method usually brings along ethical concerns due to the involvement of animal experiments, therefore, assessing bioaccessible fractions of pollutants may be a suitable alternative in portraying the reality [20]. In this study, bioaccessible fractions of PCBs were used to estimate the health risks via ingestion using an *in vitro* digestion model. No bioaccessible fractions of pollutants were used in the cancer risk estimations of the other two studied pathways of the present

Table 7
Cancer risks via ingestion of soils in humans from different types of agricultural land uses based on bioaccessible PCB toxic equivalent concentrations at minimum, medium, maximum and mean.

Sampling sites	Cancer risks via ingestion			
	Min	Median	Max	Mean
Organic farm	N.D.	N.D.	N.D.	N.D.
Agricultural	N.D.	N.D.	0.0152	0.00507 ± 0.00879
E-waste storage	N.D.	N.D.	0.00359	0.00120 ± 0.00207
Open burning site	N.D.	0.0203	0.0365	0.0189 ± 0.0183
E-waste dismantling workshop	N.D.	0.00136	**5.13**	**1.71 ± 2.96**
E-waste open burning site	N.D.	0.00611	0.139	0.0483 ± 0.0784

Note: cancer risk are in bold, N.D. means not detected and the values of cancer risk are in the unit of 10^{-6}.

study, consequently, the cancer risks from PCBs may be underestimated.

4. Conclusion

Inhalation of soil particles is the major exposure pathway of PBDEs and PCBs to humans from OBS and EW (OBS). Whereas, the major exposure pathway of other land use types including (OF, A. EW (S) and EW (DW)) is via dermal contact of soils. Soils from EW (DW) and EW (OBS) were of the greatest concern in terms of threatening human health as they contained the highest concentrations of PCBs and PBDEs, resulting in relatively high cancer risks amongst the 6 types of land use. The burning and dismantling activities in e-waste sites may still potentially pose cancer risks to humans. Although the cancer risks of PBDEs via the exposure pathways of ingestion and dermal contact of soils in EW (DW) and EW (OBS) were still very low, these two pathways were not the major exposure pathways in EW (OBS). Hence, regular monitoring is required as these pollutants may be continuously deposited on soils and eventually accumulated to hazardous levels.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jhazmat.2011.08.010.

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