



Presence of novel and legacy flame retardants and other pollutants in an e-waste site in China and associated risks

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ABSTRACT

Electrical and Electronic Equipment (EEE) residues and their management have been widely identified as potential sources of plasticizers and flame retardants to the environment, especially in non-formal e-waste facilities. This study evaluates the distribution, partitioning and environmental and human impact of organophosphate esters (OPEs), legacy polychlorinated biphenyls (PCBs), polybromodiphenyl ethers (PBDEs) and organochlorine pesticides (OCPs) in the e-waste recycling area of Baihe Tang village, in the Qingyuan county, Guangdong province, China. A plastic debris lump accumulated in a small pond during years was identified as the main source of pollution with \sum pollutants of 8400 $\mu\text{g/g dw}$, being OPEs the main contaminants detected, followed by PBDEs. This lump produced the contamination of water, sediments, soils and hen eggs in the surrounding area at high concentrations. Plastic-water and water-sediment partitioning coefficients explained the migration of OPEs to the water body and accumulation in sediments, with a strong dependence according to the K_{ow} . Triphenyl phosphate (TPhP), tricresyl phosphate (TCPs) and high chlorination degree PCBs produced a risk in soils and sediments, considering the lowest predicted no effect concentration, while the presence of PCBs and PBDEs in free range hen eggs exceeded the acceptable daily intake. OCPs were detected at low concentrations in all samples. The presence of organic contaminants in e-waste facilities worldwide is discussed to highlight the need for a strict control of EEE management to minimize environmental and human risks.

1. Introduction

Recycling activities are considered as an important way to minimize waste and residue generation from all kind of manufacturing products in the way towards circular economy and, ideally, aiming at zero waste so that all recycled matter can be used as a new raw material (Nandy et al., 2022). Recycling activities are especially important for Electrical and Electronic Equipment (EEE). EEE is defined as devices with either a battery or a power cord and can be classified in three major groups including (i) large household appliances (e.g. microwaves ovens, freezers or washing machines), (ii) information and telecommunication technology like computers and laptops and (iii) consumer equipment such as cell phones, TVs or video players among others (Perkins et al., 2014). After their operational lifetime they become electrical and electronic waste (e-waste). Worldwide production of e-waste was of 53.6

million metric tonnes (Mt) in 2019, equivalent of 7.6 kg per capita, and Asia generated the greatest volume (24.9 Mt), followed by the Americas (13.1 Mt) and Europe (12 Mt), while Africa and Oceania generated 2.9 Mt and 0.7 Mt, respectively (Forti et al., 2020; Shaikh et al., 2020).

Recycling of e-waste has economical potential due to the high market value of metals and rare earths contained in them. However, low market value materials, mostly plastics, are also generated and directly dumped to the environment (Li et al., 2020). Recycling is often performed in non-formal facilities generally in Asia, Africa and Latin America (Li et al., 2019a) which use basic handling and product recovery techniques, producing a high environmental impact (Awasthi et al., 2016) with serious human health implications (Cai et al., 2020). This is basically caused by the release of chemical pollutants linked to the plastics and electronic components, such as chlorinated biphenyls (PCBs) or polybrominated diphenyl ethers (PBDEs), recently replaced by

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organophosphate esters (OPEs) used both as plasticizers and flame retardants, polychlorinated dibenzodioxins and dibenzofurans (PCDD/F) or polycyclic aromatic hydrocarbons (PAHs) released from combustion processes, and lower market value metals such as Pb, Cr, Cd, Hg, Zn, Ni or Li among others (Shen et al., 2017). The presence of these pollutants is concerning from the environmental and human health point of view due to their accumulation and magnification potential and their toxic properties, which include endocrine disruption, neurotoxicity or developmental impairments among others (Bekele et al., 2021; Vagi et al., 2021). Therefore, it is important to determine the impact of such pollutants in areas affected by e-waste.

This study is focused in Baihe Tang, an e-waste recycling village located near Longtang town, Qingyuan County (Guangdong province, south China). Qingyuan is the third biggest e-waste recycling county in China after Guiyu and Taizhou, with more than 1300 informal and familiar small workshops in where around 1 million tons of e-waste have been recycled since the early 1990s (Han et al., 2019). Non-formal practices like open burning, acid washing, or manual mechanical dismantling of EEE have been undertaken during years and non-valuable residues, mostly plastics, have been dumped in a non-controlled way in the area (Zeng et al., 2016a). Particularly, a small pond of approximately 5000 m² and 2 m depth has received the biggest part of long-lasting dumped residues from several workshops bordering the pond (Liu et al., 2019). Material accumulated over the years has caused a big sedimentary deposition made of plastic debris and particulate matter, which has emerged as a sedimentary delta in the opposite side of the pond from where they were disposed occupying an area of 900 m² (around 20% of the pond surface). More recently, and due to recent laws and enforcements about e-waste recycling and disposal applied by central and local governments in China (Lu et al., 2014), several workshops in the surroundings and inside the village have ceased their activity, causing the abandonment and scattering of non-treated e-waste

such as CD lectors, phone batteries and chargers, capacitors, coils, circuit boards, chips, etc. In addition, the area holds other economic activities including duck and goose farming, tree growing for wood or small agricultural crops mostly dedicated to self-consumption or rural markets selling. Since it is already a highly polluted area, several studies have been carried out to investigate the accumulation of organohalogen pollutants in aquatic, amphibious and terrestrial organisms (Liu et al., 2018a), their biomagnification along the aquatic food webs (Wu et al., 2009), and human exposure (Zheng et al., 2015). However, there is a need to determine the specific sources and distribution of chemicals released during EEE recycling and abandonment considering the water-sediment-soil system and evaluation of the potential impacts both for the environment and humans.

Given the large amount of plastic and electronic debris present in the e-waste site in Baihe village, this study aims to evaluate the occurrence and impact of 14 OPEs, 11 PBDEs, 6 marker PCBs and 29 organochlorine pesticides (OCPs) in soils, sediments and free-range hen eggs. Additionally, we evaluated the presence and partitioning of the more soluble compounds, namely OPEs, in the artificial ponds affected by the e-waste pollution. We provide information on the environmental risks caused by these compounds and the human exposure through the consumption of hen eggs.

2. Materials and methods

2.1. Sample collection

The study area is the Baihe Tang village (50 m above sea level) which covers an area of approximately 73,000 m² and comprises 4 artificial ponds surrounded by agricultural areas (Fig. 1). The main economic activity is the dismantling of EEE. All samples were collected in two sampling campaigns during 2018–19.

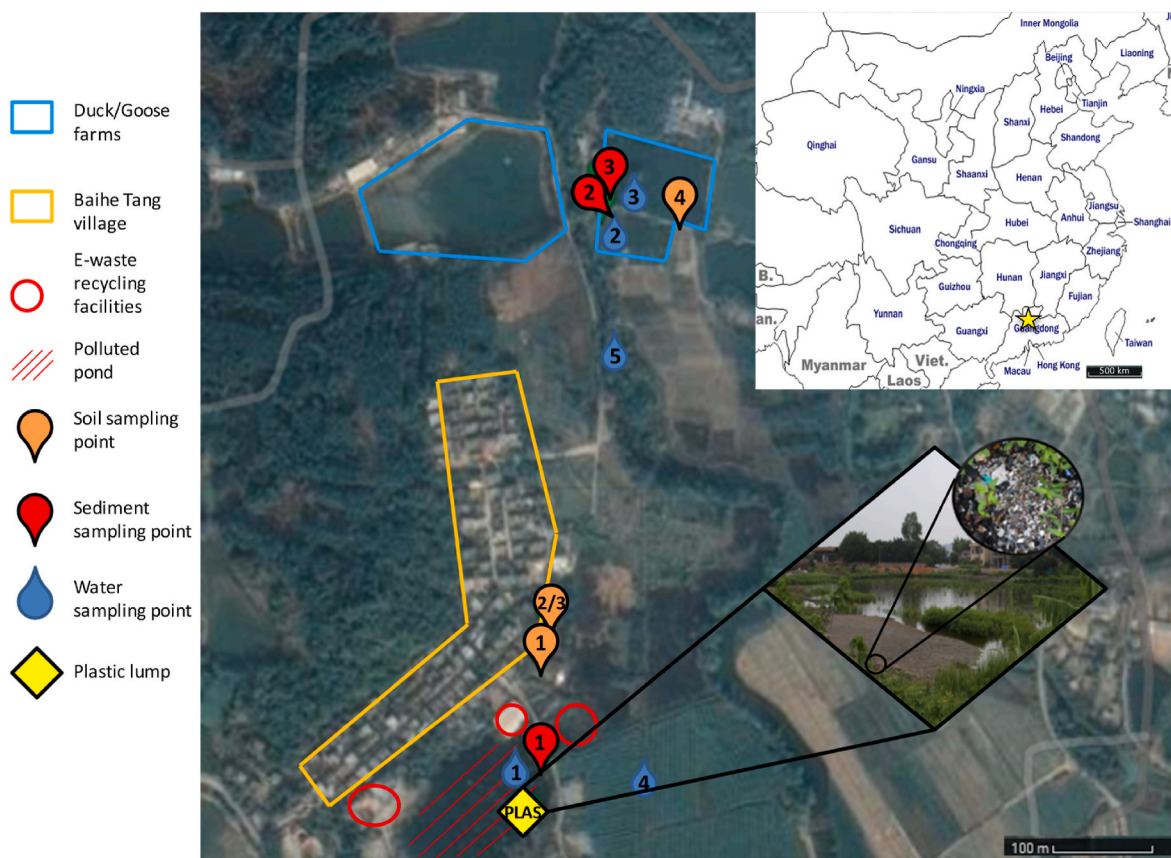


Fig. 1. Sampling points in Baihe Tang village (Qingyuan county, Guangdong province, China) and detail of the plastic lump.

Pond 1 was directly affected by e-waste debris (polluted Pond). In this pond, one sample consisting in a mixture of plastic debris mixed with particulate matter (PLAS sample) was collected directly at the “plastic lump delta”, and also a sediment sample (SED 1) and a water sample (WAT 1). Ponds 2, 3 and 4 were close to hen and duck farms (Fig. 1). In Pond 2, located at 578 m from pond 1, one sediment (SED 2) and a water sample (WAT 2) were collected. In pond 3, at 613 m from pond 1, a sediment was collected (SED 3) but at the time of sampling the pond was dry and no water was available. In pond 4, only water (WAT 3) was collected as there was a fence and sediment could not be reached. Four soil samples were taken around the ponds. SOIL 1 and 2 were natural soils, and SOIL 3 was a dredge, and they were collected between 174 m and 194 m away from the polluted pond. SOIL 4 was collected from a small corn crop close to Pond 2. Additional water samples were taken from an irrigation canal (WAT 4) and a nearby river (WAT 5). Table S1 of supporting information (SI) shows the GPS coordinates of collected samples and water content of soils and sediments. For sediment, 4 subsamples were collected with a stainless steel shovel and placed in a glass pot to make a pooled sample. The supernatant water was discarded *in situ*. For soils, 4 subsamples were collected from an approximately 1 m² with the hand shovel after removing the first layer (around 2 cm), pooled and placed in a glass pot. In between samples, the shovel was cleaned 3 times with water and ethanol. Soils and sediments were frozen at -70 °C and freeze-dried. Waters were grab sampled using a glass bottle tied to a long rope and thrown as close as possible to the centre of the water body and water was dosed in amber glass bottles. Samples were covered with aluminium foil, capped and kept refrigerated in a portable fridge and transported to the laboratory. Waters were kept at 2 °C until analysis which was performed in less than 15 days to avoid degradation of contaminants.

Hen eggs were provided by local peasants who grow free range hens. Eggs were wrapped with a bubble plastic to prevent ruptures and transported to the laboratory in a portable fridge. Once in the laboratory, biometric assays were done before opening the eggs (Table S2 in SI). The 24 fresh eggs were crack-opened and homogenized in a total of 6 pooled samples corresponding to 3 pools of 3 eggs each for the samples collected in 2018 (samples HEN 1–3, 3 eggs/pool) and 3 more pools of 5 eggs each for the samples collected in 2019 (samples HEN 4–6, 5 eggs/pool). Pooled samples were frozen at -70 °C and freeze-dried.

2.2. Chemicals and reagents

Standard mixtures and/or individual solutions for OPEs, PBDEs, PCBs, and OCPs were purchased from AccuStandard (New Haven, CT, USA) at different concentrations (see section S1 in SI for studied compounds and details). Dicofol, endosulfan sulphate, hexachlorobutadiene (HCBu), pentachlorobenzene (PeCB) were purchased from Sigma-Aldrich (St.Louis, MO, USA). Three standard solutions, one for OPEs, one for PBDEs and one containing PCBs and OCPs were prepared at 1 µg/mL (0.1 µg/mL for PCBs) in toluene and used as mother solutions for the preparation of the calibration curves and quality control spikes. Surrogate standards were PCBs 65 and 209, TnPP-D21, TCPD-D18 and TPHP-D15 from AccuStandard, and ¹³C hexachlorobenzene and ¹³C 4,4'-DDE from Cambridge Isotope Laboratories (Andover, MA 01810, USA).

Three types of SPE cartridges were used for sample treatment: ENVI Florisil (500 mg, 3 mL, Supelco Bellefonte, PA, USA) for soils and sediments; Oasis HLB (200 mg, 6 cc, Waters, Milford, Massachusetts, USA) for OPEs extraction from water and eggs samples, and Hyper Sep Florisil cartridges (10 g, 75 mL, Thermo Scientific, Rockwood, TN, USA) for POPs in egg samples. Gas chromatography grade methanol, dichloromethane, acetonitrile and hexane were provided by Oceanpk (Sweden) and ethyl acetate by Honeywell (USA).

2.3. Sample extraction

2.3.1. Soils and sediments

Soils and sediments were fine grounded with a mortar and pestle and sieved to obtain the 100 µm fraction. One gram of sample was spiked with 0.1 µg of each surrogate standard and then Soxhlet extracted (48 h, 60 °C) with 200 mL of n-hexane:acetone (1:1). Copper pieces were added to the flask in order to remove elemental sulphur. Solvent was collected and evaporated in a rotary evaporator R-215 working at 450 bar (Buchi, Switzerland) at room temperature until 1 mL approximately before the clean-up step. ENVI florisil SPE cartridges were conditioned with 8 mL of ethyl acetate and 6 mL of n-hexane, then extracts were loaded and eluted by gravity with 5 mL of n-hexane followed by 5 mL of n-hexane:dichloromethane (DCM) (1:1) to obtain a first fraction containing non-polar compounds like PBDEs, PCBs and OCPs. A second fraction was obtained by elution with 8 mL of ethyl acetate and used for OPEs determination. Both fractions were solvent exchanged to 1 mL of n-hexane and stored at -20 °C until analysis. Pollutant concentrations in sediment and soil are expressed in µg/g dry weight (dw), and because the huge differences in concentration among pollutants, different significant digits are used to maintain the same concentration units.

Total Organic Carbon (TOC) was analyzed in an ELTRA CS-800 double dual range C/S detector (Haan, Germany) working at 2000 °C and 100 nm wavelength. Around 100 mg of sample were accurately weighed in porous ceramic capsules, then treated with HCl (aq) (35%) for 15 h at room temperature and heated at 80 °C for 1 h to dissolve carbonates, washed with double deionized H₂O until neutral pH and dried at 80 °C. Around 0.7 g of Fe (s) and 1.5 g of W (s) were added to each sample and each calibration standard. Calibration for TOC determination was performed in two levels with the certificate reference materials (i) soil standard AEB2178 (alpha resources LLC, Stevensville, MI Michigan, USA) with 3.07% C for medium organic content samples and (ii) steel chips calibration standard 92,400-4020 C215 (Eltra, Haan, Germany) with 0.462% of C for low content samples.

2.3.2. Waters

OPEs were the only compounds analyzed in water as the rest of compounds are not expected to be detected in this compartment due to their high hydrophobicity and adsorption to the organic matter of soils and sediments or accumulation in biota (Zhang et al., 2011). Water was filtered through cellulose paper (pore size 10 µm) to remove gross particulate matter and suspended solids like leaves or roots. 500 mL of water were spiked with 0.1 µg of OPEs surrogate standards. Extraction was performed with Oasis HLB cartridges (200 mg, 6 cc) which were conditioned with 4 mL ethyl acetate, 4 mL of methanol and 4 mL water. After preconcentration, cartridges were then dried and eluted with 8 mL of ethyl acetate. Approximately 1 g of anhydrous Na₂SO₄ was then added to the extract to eliminate remaining water and then the extract was filtered through 0.45 µm PTFE filters. Solvent was evaporated under a gentle N₂ stream to approximately 1 mL, transferred to a chromatographic vial, evaporated to dryness and reconstituted with 500 µL of n-hexane. Samples were then stored in the freezer (-20 °C) until instrumental analysis. OPEs concentrations in water are expressed as µg/L.

2.3.3. Eggs

Due to the high lipid content of eggs, the extraction for PBDEs, PCBs and OCPs detection was not compatible with the analysis of OPEs, and thus, two analytical protocols were undertaken. For OPEs, a previously optimized method for fish muscle based on freeze lipid precipitation as a key step for the clean-up process was used (Liu et al., 2018b). One gram of homogenized and lyophilized sample was spiked with 0.1 µg of surrogate standards and Soxhlet extracted with 200 mL of DCM at 50 °C during 24 h. The solvent was then evaporated to approximately 5 mL in a Buchi R-215 rotavapor, dosed in a test tube and filled with fresh solvent to 10 mL. One mL of the extract was used for lipid determination,

which was performed by gravimetric measurements after evaporating organic solvent at 60 °C for 24 h. The remaining extract was evaporated to dryness under N₂ stream, reconstituted with 8 mL of MeOH and frozen at -20 °C overnight. Then, the MeOH supernatant was decanted to separate it from the frozen lipids. The freeze lipid precipitation process was repeated with another portion of 8 mL of MeOH. Both MeOH extracts were transferred to a 500 mL flask previously filled with 300 mL water. For the clean-up process, Oasis HLB cartridges (200 mg, 6 cc) were conditioned with 4 mL ethyl acetate followed by 4 mL MeOH. Sample was preconcentrated at a flow of 15 mL/min, dried under vacuum for 30 min and eluted with 8 mL ethyl acetate. If some lipids remained (turbid liquid), Z-sep C18 sorbent (Supel QuE Z-sep, Supelco) was used to remove them before filtering through 0.45 µm PTFE syringe filters. If some water was also present, the same procedure was done with Na₂SO₄ instead of Z-sep C18 sorbent. The extract was evaporated until dryness and reconstituted in 500 µL of n-hexane.

PBDEs, PCBs and OCPs were extracted based on previously published studies (Zapata et al., 2018). Briefly, 1 g of homogenized and lyophilized sample was spiked with 0.1 µg of surrogate standards, vortexed with 25 mL n-hexane:DCM (1:1) and ultrasound extracted for 10 min and this procedure was repeated 3 times. The extract was further purified with Hyper Sep florisil SPE cartridges (10 g) using 35 mL of n-hexane:DCM (1:1) elution solvents by gravity, evaporated until near dryness under N₂ stream and reconstituted in 500 µL n-hexane. Pollutant concentrations in hen eggs are expressed as µg/g wet weight (ww). Mean water and lipid contents were 72.7 ± 1% and 3.3 ± 0.6%, respectively (Table S2 of SI).

2.4. Instrumental analysis

OPEs, PCBs and OCPs were analyzed in an Agilent 7890 A GC system coupled with an Agilent 7000 A GC/MS Triple Quadrupole detector. An Agilent J&W DB-5MS column (30 m length, 0.25 mm internal diameter, 0.25 µm film thickness) was used. Two instrumental methods, one for the analysis of OPEs adapted from Poma et al. (2018) and one for the analysis of PCBs and OCPs (Zapata et al., 2018) were used. Oven, injector and ion source temperatures, together with retention times, selected transitions and collision energies are shown in the SI, Section S2, Table S3 for OPEs and S4 for OCPs and PCBs.

PBDEs were analyzed in an Agilent 7890 A GC system coupled to an Agilent 5975C mass spectrometer working in negative chemical ionization. A short DB-5MS column (15 m length, 0.25 mm internal diameter, 0.25 µm film thickness, Agilent J&W) was used to enable BDE 209 determination. Details about operating conditions can be found in section S2 of the SI. Monitored ions for PBDEs are indicated in Table S4.

2.5. Quality assurance and quality control

Samples were processed in 12 position batches. Each batch included one matrix blank (precleaned soil and sediment, commercial hen egg or double deionized water), one procedural blank (no matrix), one spiked Quality Control (QC) and 9 samples. For QC, a blank soil sample was spiked with native compounds at a concentration of 0.1 µg/g dw (n = 5), commercial hen eggs were spiked at 0.1 µg/g ww (n = 5) and distilled water was spiked 0.2 µg/L (n = 5).

All glass material was baked at 450 °C to avoid external contamination. However, OPEs were detected in blank samples due to their presence in filters, plastics or laboratory equipment used for their analysis. Quantification of OPEs was done at 3 times the blank contribution. Mean contribution for each compound detected in blanks, namely TEP, TiPP, TnBP, TCEP, TCPP, TPhP, EHDPP and TEHP, was subtracted from calculated samples concentration. Recovery, blank contribution and Method Detection Limits (MDL) calculated as 3 times the signal to noise ratio or considering the blank contribution in the case of OPEs for each sample matrix are compiled in Tables S3 and S4 of the SI.

2.6. Solid-liquid partitioning ratios

In order to assess the migration of OPEs from the plastic lump to the water and the accumulation from the water to the sediment, partitioning ratios (*K*) were calculated between PLAS and WAT1 and between WAT1 and SED1, respectively, following equations (1) and (2):

$$K_{WAT1/PLAS} = \frac{C_{i,WAT1} \left(\frac{\mu\text{g}}{\text{L}} \right)}{C_{i,PLAS} \left(\frac{\mu\text{g}}{\text{kg}} \right)} \quad (1)$$

$$K_{SED1/WAT1} = \frac{C_{i,SED1} \left(\frac{\mu\text{g}}{\text{kg}} \right)}{C_{i,WAT1} \left(\frac{\mu\text{g}}{\text{L}} \right)} \quad (2)$$

where *C*_{*i*,PLAS} is the measured concentration of pollutant *i* in sample PLAS, *C*_{*i*,WAT1} is the measured concentration of pollutant *i* in sample WAT1 from and *C*_{*i*,SED1} is the measured concentration of pollutant *i* in sample SED1, both from Pond 1.

2.7. Environmental risk assessment and estimated daily intake (EDI)

To evaluate the impact of pollutants in soil/sediments and water, the risk quotients (RQ) were calculated. RQ are the ratio between the measured environmental concentration (MEC) and a toxicity reference value (equation (3)) (Fan et al., 2021).

$$RQ = \frac{MEC}{PNEC} \quad (3)$$

The lowest predicted no effect concentration (PNEC) from the NORMAN ecotoxicology database (2021) for each pollutant was used to represent the worst-case scenario (Table S5 of the SI). RQ values lower than 1 represents no environmental risk, while 1.0 = RQ < 10 indicates a small potential for adverse effects; 10 = RQ < 100 indicates significant potential for adverse effects; RQ ≥ 100 indicates that potential adverse effects should be expected. In order to assess the risk associated to chemical mixtures, the concentration addition (CA) model was used to calculate a total RQ for each family of pollutants, assuming the same modes of toxic action for substances of the same family (Backhaus and Faust, 2012).

To estimate the exposure of humans from the ingestion of polluted eggs, estimated daily intakes (EDIs) were calculated using equation (4):

$$EDI = \frac{C_i \times CR}{BW} \quad (4)$$

where *C*_{*i*} is the amount of each pollutant in the eggs expressed in µg/g ww, CR (consumption rate) is the amount of egg consumed daily per person, obtained from a consumption survey in Guangdong province (14 g per person and day in rural areas) (Ma et al., 2005) and BW is the average body weight (63 kg for adults (Zhou et al., 2012) and 14.65 kg for children (Huang et al., 2018)). EDIs have been calculated as mean values of the 6 pooled egg samples. Calculated EDI values were compared with reference doses (RfD) or acceptable daily intakes (ADIs), depending on the available data, to obtain the margin of exposure (MOE) as shown in equation (5):

$$MOE = \frac{RfD}{EDI} \sim \frac{ADI}{EDI} \quad (5)$$

MOE provide information about the risks associated with pollution exposure due to ingestion. MOE values higher than 2.5 represent no health concern; the lower MOE value, the higher risk for human health.

3. Results and discussion

3.1. Contamination of sediments from the Baihe e-waste site

Table 1 shows the concentration of each studied OPEs, PBDEs and

Table 1

Concentration of pollutants ($\mu\text{g/g dw}$) in the plastic lump (PLAS), sediments and soils from the e-waste site. Distance to the main source and total organic carbon (TOC) content in percentage are also indicated. na = not analyzed; nd = not detected.

	PLAS	SED 1	SED 2	SED 3	SOIL 1	SOIL 2	SOIL 3	SOIL 4
Distance to source (m)	source	0	578	613	174	197	197	614
%TOC	na	8.7	1.9	1.1	6.9	0.2	5.8	0.9
TEP	0.89	0.14	nd	nd	nd	nd	0.08	0.08
TiPP	0.87	0.14	nd	0.01	nd	0.11	0.01	0.09
TnPP	0.07	0.10	0.003	nd	0.01	0.01	nd	0.01
TnBP	7.17	0.47	0.43	0.06	0.12	0.11	0.47	0.18
TCEP	10.9	2.03	0.16	0.18	1.31	0.06	1.34	0.34
TCPP	3450	10.1	0.24	0.45	1.19	0.08	6.24	22.7
TDCP	5.79	0.32	0.02	0.02	0.21	0.004	0.82	0.14
TPhP	4100	40.8	0.19	1.30	2.55	0.01	8.52	42.7
EHDPP	9.12	0.08	0.01	0.01	0.02	0.01	0.02	0.25
TEHP	1.15	0.27	0.01	0.01	nd	0.01	0.05	0.03
TCP1	184	33.2	0.05	0.85	0.41	nd	0.48	4.52
TCP2	108	14.2	0.03	0.38	0.27	0.02	0.30	2.58
TCP3	70.3	6.54	0.03	0.15	0.18	0.06	0.21	1.66
TCP4	56.7	3.23	0.02	0.06	nd	nd	0.15	1.30
$\sum\text{OPEs}$	8005	112	1.2	3.5	6.3	0.5	19	76
BDE 28	17.6	0.59	0.003	nd	0.05	0.01	0.05	nd
BDE 47	63.7	2.35	0.03	nd	0.22	0.01	0.21	nd
BDE 66	25.4	0.71	0.005	nd	0.09	0.01	0.09	nd
BDE 100	37.7	1.82	0.005	nd	0.03	0.005	0.06	nd
BDE 99	52.5	2.44	0.03	nd	0.32	0.01	0.59	nd
BDE 85	4.86	0.11	nd	nd	0.01	0.01	0.02	nd
BDE 154	3.50	0.13	0.005	nd	0.03	0.001	0.06	nd
BDE 153	15.6	0.37	0.01	nd	0.08	n.d.	0.12	nd
BDE 138	1.76	0.08	0.003	nd	0.01	nd	0.01	nd
BDE 183	10.2	0.63	0.02	nd	0.10	nd	0.07	nd
BDE 209	190	29.8	18.1	0.05	2.72	0.03	3.10	0.16
$\sum\text{PBDEs}$	423	39	18	0.05	3.6	0.08	4.4	0.16
PCB 28	3.10	3.34	0.33	0.10	2.25	0.02	5.82	0.03
PCB 52	0.97	1.26	0.15	0.01	0.75	0.003	0.78	nd
PCB 101	0.32	0.95	0.22	0.03	0.68	0.01	2.29	nd
PCB 153	0.37	5.64	0.17	0.02	0.46	nd	24.2	0.01
PCB 138	0.27	5.56	0.14	0.02	0.59	nd	22.4	0.01
PCB 180	1.33	3.37	0.08	nd	0.18	nd	19.8	0.01
$\sum\text{PCBs}$	6.36	20	1.0	0.2	4.9	0.03	75	0.06
HCBu	nd	nd	nd	nd	nd	0.02	nd	nd
PeCB	nd	0.60	nd	0.05	0.03	nd	0.14	nd
$\alpha\text{-HCH}$	0.001	nd	0.0006	0.001	0.0006	0.0009	nd	0.0007
HCB	0.017	0.24	0.006	0.02	0.009	0.0005	0.12	0.0009
$\beta\text{-HCH}$	nd	nd	nd	0.001	nd	nd	nd	nd
$\delta\text{-HCH}$	nd	nd	0.0003	nd	nd	nd	nd	0.0004
$\gamma\text{-HCH}$	nd	nd	nd	nd	nd	nd	nd	nd
$\alpha\text{-endosulfan}$	nd	nd	nd	nd	nd	nd	0.007	nd
Transchlordan	0.008	nd	0.004	nd	nd	0.0005	nd	nd
Dieldrin	0.915	nd	nd	nd	nd	nd	nd	nd
4,4'-DDE	0.005	0.005	nd	nd	0.006	nd	nd	nd
$\beta\text{-endosulfan}$	nd	0.06	nd	nd	nd	nd	0.16	nd
4,4'-DDD	0.005	nd	0.0008	nd	0.002	nd	nd	nd
Chlordecone	nd	nd	nd	nd	nd	nd	nd	nd
4,4'-DDT	nd	nd	nd	nd	0.01	nd	nd	nd
Metoxychlor	nd	nd	nd	nd	nd	nd	nd	nd
Mirex	0.011	0.06	nd	nd	nd	nd	nd	nd
$\sum\text{OCPs}$	0.05	0.97	0.01	0.07	0.06	0.02	0.43	0.003
$\sum\text{pollutants}$	8400	172	20	3.8	14.9	0.6	99	77

In general 3 significant digits are provided, although for some compounds and due to the low concentrations 3 or 4 decimals are provided to maintain the $\mu\text{g/g}$ units, for better comparison among high and low concentrations. Total concentrations rounded to significant digits.

PCBs in the plastic lump, sediments and soils. No correlation was found between the concentration of contaminants and distance from the polluted pond or with the TOC. The plastic lump (PLAS) had the highest levels among samples, with a $\sum\text{pollutants}$ of $8400 \mu\text{g/g dw}$, where OPEs accounted for 95% of $\sum\text{pollutants}$, followed by PBDEs (5%), and PCBs and OCPs accounted for less than 0.1%. $\sum\text{OPEs}$ were detected at $8005 \mu\text{g/g dw}$, with a profile dominated by TPhP and TCPP with concentrations of $4100 \mu\text{g/g dw}$ and $3450 \mu\text{g/g dw}$, respectively, and represented 51% and 43% of $\sum\text{OPEs}$, followed by TCP isomers ($420 \mu\text{g/g dw}$ for the sum of the 4 TCP isomers, 5% of $\sum\text{OPEs}$). TPhP is a plastic additive also used as flame retardant for the manufacture of electrical, electronic and optical equipment, machineries and vehicles. TCPP is mainly used as plastic additive for rubber products and polymers, adhesives and

sealants, coating products, laboratory chemicals and leather treatment products, and in formulation of mixtures and/or re-packaging (ECHA, 2022). TCPs are used as additive in lubricants, hydraulic fluids, or engine oils, and as plasticizers and flame retardant (de Ree et al., 2014). TCEP, EHDPP, TnBP and TDCP were detected at concentrations from 5.79 to $10.9 \mu\text{g/g dw}$ and were also associated to their use as flame retardants or plasticizers present in EEE. $\sum\text{PBDEs}$ were detected in PLAS sample at $423 \mu\text{g/g dw}$, and BDE 209 was the main congener detected (45% of $\sum\text{PBDEs}$), followed by BDE 47, 99 and 100. The high levels of OPEs compared to PBDEs may point to their more recent use related with newer EEE devices dumped and/or stored there. PLAS also contained $6.36 \mu\text{g/g dw}$ of $\sum\text{PCBs}$, with a profile dominated by PCB 28 and 180. Although OCPs are not directly associated to EEE, dieldrin was

detected in PLAS at the highest concentration (0.915 $\mu\text{g/g dw}$), followed by hexachlorobenzene (HCB), mirex, transchlordane and DDTs at 0.005–0.017 $\mu\text{g/g dw}$. The concentration of other OCPs are indicated in Table 1.

SED 1 close to the plastic lump had \sum pollutants of 172 $\mu\text{g/g dw}$, 49 times lower than PLAS. OPEs (65% of \sum pollutants) were again detected at the highest concentrations (0.08–40.8 $\mu\text{g/g dw}$) and were 72 times lower than in PLAS, while a decrease of 10 times was observed for PBDEs (23% of \sum pollutants, 0.08–29.8 $\mu\text{g/g dw}$) with respect to PLAS sample. The higher decrease for OPEs compared to PBDEs was attributed to their higher water degradability (Cristale et al., 2017a). PCBs (12% of \sum pollutants, 0.95–5.64 $\mu\text{g/g dw}$) were higher in SED 1 than in PLAS, and OCPs (1% of \sum pollutants, nd – 0.60 $\mu\text{g/g dw}$) were at similar concentrations. Although concentrations differed (Table 1), a similar profile was observed among pollutants in PLAS and SED 1, dominated by TPhP, TCPs and TCPP, BDEs 209 and 47 and all PCB congeners. Considering logarithmic transformed individual concentrations of OPEs and PBDEs, a good correlation was observed among PLAS and SED 1 ($R^2 = 0.76$, $p < 0.001$) (Fig. 2), and this indicates that the plastic lump is most probably the source of pollution of the whole pond. On the contrary, PCBs and OCPs did not correlate among PLAS and SED 1, and this indicates that these legacy pesticides originated from other past activities. Specifically, SED 1 contained 0.60 $\mu\text{g/g dw}$ of PeCB, which was not detected in PLAS, and 0.24 $\mu\text{g/g dw}$ HCB. Both these compounds are classified as unintentionally produced by the Stockholm Convention (2009) and used as additives in PCB mixtures, rubbers, electronic equipment or as flame retardants.

SED 2 and 3 were located in ponds 2 and 3, at 600 m approximately from the polluted pond, and had \sum pollutants of 20 and 3.8 $\mu\text{g/g dw}$, respectively. SED 2 was dominated by PBDEs, which accounted for 89% of \sum pollutants with BDE 209 as the main congener detected. SED 3 was dominated by OPEs, which accounted for 92% of \sum pollutants with TPhP, TCP, TCPP and TCEP as the main OPEs. In these two sediments, PCBs ranged from 0.01 to 0.33 $\mu\text{g/g dw}$ and OCPs ranged from n. d. to 0.05 $\mu\text{g/g dw}$, with PeCB and HCB, again, as the main OCPs detected.

3.2. Water-sediment partitioning of OPEs

The concentration of OPEs in water samples are compiled in Table 2. WAT 1, from the polluted pond, had higher concentration (\sum OPEs 1.62 $\mu\text{g/L}$) than the rest of water samples (\sum OPEs 0.36–0.76 $\mu\text{g/L}$) due to the influence of the plastic lump. Main compounds detected in waters were TPhP, TCPP, TEP, TiPP, and the rest of compounds (TnBP, TCEP,

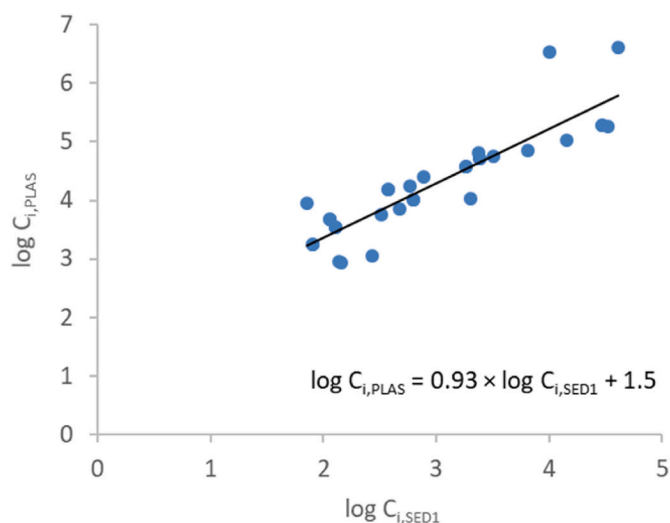


Fig. 2. Correlation between individual OPEs and PBDEs log-transformed concentrations in samples PLAS and SED1 ($R^2 = 0.78$, $p = 2E-11$).

Table 2

Concentration of OPEs ($\mu\text{g/L}$) in water samples from the e-waste site and distance to the main source (m). \sum RQ considering the concentration addition model also provided. nd = not detected. Different number of decimals provided as the concentrations largely differed but are always expressed as $\mu\text{g/L}$.

COMPOUNDS	WAT 1	WAT 2	WAT 3	WAT 4	WAT 5
Distance to the source (m)	0.0	569	613	165	430
TEP	0.22	0.10	0.06	0.04	0.08
TiPP	0.39	0.01	0.01	0.03	0.04
TnPP	nd	nd	nd	nd	nd
TnBP	0.03	0.09	0.04	0.04	0.02
TCEP	0.02	0.04	0.03	0.05	0.03
TCPP	0.34	0.48	0.15	0.15	0.11
TDCP	0.003	0.01	0.001	0.02	0.002
TPhP	0.52	nd	nd	nd	0.17
EHDPP	0.001	0.001	0.001	0.001	0.001
TEHP	nd	0.02	nd	0.02	0.02
TCP1	0.04	nd	0.02	0.007	nd
TCP2	0.02	nd	0.02	0.003	0.002
TCP3	0.02	nd	0.02	nd	nd
TCP4	0.01	0.02	0.02	nd	0.03
\sum OPEs	1.62	0.76	0.39	0.36	0.50
\sum RQ	4.8	1.4	2.8	1.1	2.0

EHDPP, TEHP, TDCP and TCPs) were detected at much lower concentration. TnPP was not detected in any water sample.

In Pond 1, OPEs contained in the plastic debris produced during the non-formal recycling processes leached to water, and a water-sediment partitioning was observed according to the polarity and degradability of contaminants. TPhP was the main OPE in the plastic lump, and also in sediment and in water of Pond 1, and this indicates that plasticizers leaching from the plastic lump solubilized in water and accumulated in sediment. Other OPEs detected in WAT 1 at high concentrations were TiPP, TCPP and TEP (0.22–0.39 $\mu\text{g/L}$) and correspond to OPEs with the highest solubility and lowest K_{ow} , indicating that they were easily solubilized in water. A negative correlation between $\log K_{WAT1/PLAS}$ and $\log K_{ow}$ was found to be statistically significant ($R^2 = 0.54$, $p = 0.004$) (Fig. 3A), suggesting that the migration from plastic to water decreases when the hydrophobicity of the compound increases. On the other hand, a positive correlation was found between $\log K_{SED1/WAT1}$ and $\log K_{ow}$ ($R^2 = 0.61$, $p = 0.002$) (Fig. 3B), which indicate that an increase in the hydrophobicity of the compound implies a higher accumulation in the sediment. In contrast, waters collected from Pond 2 (WAT2) had a similar OPEs profile than the sediment from this pond, but no significant correlation was found between them, probably due to the fact that Pond 2 was not directly affected by a plastic lump and that concentrations in water and sediment were much lower.

3.3. Presence of POPs and OPEs in soils

\sum pollutants in soils ranged from 0.60 to 98.8 $\mu\text{g/g dw}$, with SOIL 3 > SOIL 4 \gg SOIL 1 \gg SOIL 2 (Table 1). These concentrations reflect the impact of the EEE dismantling activities in the soil quality. Although there was no correlation among pollutants with TOC, SOIL 2, the natural soil with a red/brown colour characteristic from the iron oxides present in high clay content soils, had low carbon content (0.2% TOC) and the lowest concentrations of pollutant. Contrarily, SOIL 3, the dredge with a brown-dark colour containing 5.8% TOC was the most contaminated soil. Pollution profiles in these 4 soil samples were very different. SOIL 1 contained OPEs > PCBs > PBDEs, SOIL 2 contained OPEs > PBDEs > PCBs, SOIL 3 was dominated by PCBs (76% of \sum pollutants) especially highly chlorinated ones and SOIL 4, the agricultural soil, was dominated by OPEs. In all cases OCPs were detected at low concentrations.

\sum OPEs ranged from 0.5 to 76 $\mu\text{g/g dw}$. Except for SOIL 2, OPEs were dominated by TPhP and TCPP, ranging from 2.55 to 42.7 $\mu\text{g/g dw}$ and 1.19 and 22.7, respectively, and contributed from 27 to 56% of \sum OPEs, followed by TCEP, and TDCP. \sum PBDEs in SOIL 1 and 3 were of 3.6 and 4.4 $\mu\text{g/g dw}$ respectively, and were always dominated by BDE 209,

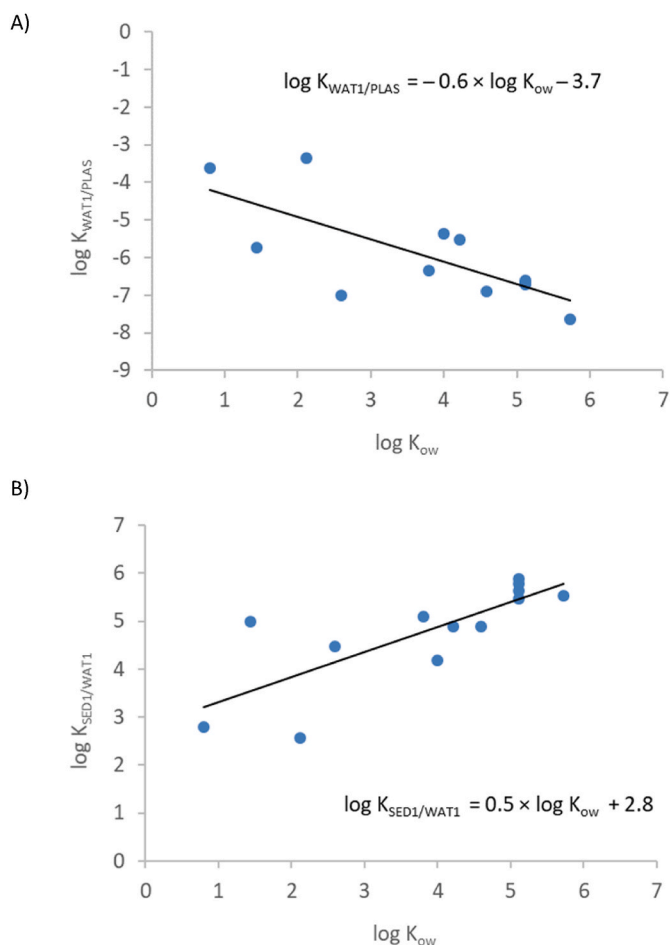


Fig. 3. Correlations for individual OPEs A) between the log-transformed $K_{WAT1/PLAS}$ ratio and $\log K_{OW}$ ($R^2 = 0.54$, $p = 0.004$) and B) between the log-transformed $K_{SED1/WAT1}$ ratio and $\log K_{OW}$ ($R^2 = 0.61$, $p = 0.002$); $\log K_{OW}$ values were obtained from the Estimation Program Interface (EPI) suite (US EPA, 2012).

which accounted for 74 and 71% of Σ PBDEs, followed by BDE 99 and 47. Much lower concentrations were detected in SOIL 2 (low TOC), that contained all studied PBDE congeners, and in SOIL 4 (agricultural soil), where BDE 209 was the only congener detected. Σ PCBs ranged from 0.03 $\mu\text{g/g dw}$ in SOIL 2 to 75 $\mu\text{g/g dw}$ in SOIL 3, which showed a profile dominated by PCBs 153, 138 and 180, and 28 in a minor level. Previous studies in the area suggest that the high levels of PCBs in this site are associated to PCB-containing devices dismantled in the past (Zheng et al., 2015). Specifically, SOIL 3 which was a dredge might reflect this past contamination. Pesticides were sporadically detected in soils with Σ OCPs between 0.003 and 0.43 $\mu\text{g/g dw}$, being PeCB, HCB, and β -endosulfan the main compounds detected. OCPs heptachlor and its epoxides, oxychlordane, cis-chlordane, aldrin, isodrin, dieldrin, endrin, 2,4'-DDE, 2,4'-DDD, chlordane, endosulfan sulphate, dicofol, metoxychlor and mirex were never detected in soils.

3.4. Presence of POPs and OPEs in hen eggs

In e-waste sites, contamination of soils affects hens picking plastics and other residues while they get fed (Wang et al., 2014; Zeng et al., 2018). In this study we used free range hen eggs as an indicator of human exposure through ingestion, which raises concerns on human health (Li et al., 2020). Σ pollutants ranged from 0.40 to 5 $\mu\text{g/g ww}$ (Table S6 of the SI for individual concentrations, Table 3 for mean concentrations). PCBs were the most abundant compounds, with mean

$\Sigma_{6\text{marker}}\text{PCB}$ of 0.37 $\mu\text{g/g ww}$, followed by PBDEs (mean $\Sigma_{11}\text{PBDE}$ of 2.95 $\mu\text{g/g ww}$). The PCB profile was dominated by PCB 138 and PCB 153, followed by PCB 28; BDE 209 was the main PBDEs detected although HEN1 had high levels of BDE 99 and 154. TEP, TnBP and TCPP were the main OPEs detected and the rest were below the MDL as these compounds can be metabolized (Wang et al., 2021). Mean Σ OCPs was of 0.98 $\mu\text{g/g ww}$ (Table S6). Despite OCPs were detected at low concentrations (Table 3), some pesticides might be associated to agricultural activities while others such as HCHs, HCBu, PeCB and HCB are associated to EEE dismantling due to their use as flame retardants, additives and/or plasticizers. The rest of POPs (γ -HCH, heptachlor, aldrin, isodrin, dieldrin, endrin, heptachlor epoxides, oxychlordane, cis or trans isomers of chlordane, 2,4'-DDE, 2,4'-DDD, α - or β -isomers of endosulfan, metoxychlor, endosulfan sulphate, and PBDE 138) were never detected in studied hen eggs.

Considering the median concentration in eggs and daily egg ingestion, EDIs for each contaminant ranged from 0.0001 to 0.51 $\mu\text{g/kg body weight (bw)}$ per day in adults and from 0.0004 to 2.18 $\mu\text{g/kg bw}$ per day in children (Table 3). Regarding MOEs, values for the 3 detected OPEs in hen eggs and OCPs (individually or grouped by families) were much higher than 2.5, which implies no human health concern. On the other hand, MOE values for Σ marker PCBs were of 0.10 and 0.03 for adults and children, respectively, and BDE 99 had a MOE of 1.4 and 0.3 in adults and children, respectively, and Σ pentaBDEs of 0.9 in children, and indicates that consumption of free range hen eggs can be hazardous. As comparison, hen eggs from Qingyuan e-waste sites collected in 2010 contained PCBs 10 to 100 times higher than those from the control site, and estimated daily intake ranged from 0.07 to 0.32 $\mu\text{g/kg bw}$ per day (Zheng et al., 2012). In eggs collected in 2013 and 2016 also in Qingyuan, PCBs daily intake was above the minimal risk levels for adults in 70% of the samples and for children in all the samples (Zeng et al., 2018). Those same samples were also evaluated for PBDEs, and EDIs were lower than the EPA reference values but BDE 47 and BDE 99 showed a MOE lower than 2.5, which implies potential health risks for humans (Huang et al., 2018). Therefore, EEE dismantling activities in Bahie Tang village and in other e-waste sites are a cause of concern for human health as eggs constitute a basic and nutritious food.

3.5. Risk assessment and environmental impacts of the Baihe e-waste site

Pollutants detected in water, soil and sediments were present at toxicologically relevant concentrations which can have implications for the environment and human health. Table 4 indicates RQs of each individual contaminant for PLAS sample and for sediments and soils. Waters in general showed a low Σ RQ (Table 2) but PLAS, some sediments and soils had a very high risk in the order $\text{PLAS} \gg \text{SOIL 3} > \text{SED 1} > \text{SOIL 4}$. The rest of the samples had a much lower risk (Table 4).

Considering the PNEC values of individual compounds, the Σ RQ of PLAS sample was very high (160,000) and this plastic debris formed by the long-term accumulation of plastic waste associated to EEE should be considered as a toxic residue. Compounds contributing to the toxicity are indicated in Table 4 and are mainly OPEs and BDEs 47, 99 and 183. Transchlordane and dieldrin, despite detected at 0.008 and 0.915 $\mu\text{g/g dw}$ (Table 1), posed a risk due to the very low PNEC value. Other OCPs, PBDEs or PCBs produced a moderate risk.

Σ RQs in SED 1 from the polluted pond was 5 times lower than for the PLAS sample, and the main contributor was β -endosulfan, that despite being detected at a low concentration has a very high toxicity. OPEs highly contributed to the total RQ, together with PeCB and PCBs 180 and 153. Other compounds are indicated in Table 4. The overall high RQ detected in SED 1 reflect the leaching of contaminants from the plastic debris and accumulation to sediments at concentrations that cause an environmental impact. SED 2 and 3 had a much lower risk than SED 1, and mainly caused by TCPs and transchlordane in SED 2 and TCPs and PeCB in SED 3.

Regarding natural soils, Σ RQ were of 414 for SOIL 1 (moderate risk)

Table 3

Mean concentration of pollutants in pooled hen eggs in $\mu\text{g/g ww}$ ($n = 6$), reference doses (RfD) or acceptable daily intakes (ADI) obtained from different references. Estimated daily intakes (EDI) and margin of exposure (MOE) calculated from RfD or ADI due to eggs consumption for adults and children. ΣTCPs includes 4 TCP isomers; $\Sigma\text{6m-PCBs}$ include congeners 28, 52, 101, 138, 153 and 180; ΣHCHs includes α , β , γ and δ isomers; ΣDDTs includes 2,4'-DDT, 2,4'-DDE, 2,4'-DDD, 4,4'-DDT, 4,4'-DDE and 4,4'-DDD. Three decimal figures except for some OCPs where 4 decimals are included as levels are very low but still significant so that the same units ($\mu\text{g/g ww}$ or $\mu\text{g/kg bw per day}$) are provided for all compounds.

Compound	Mean ($n = 6$) ($\mu\text{g/g ww}$)	RfD ($\mu\text{g/kg bw per day}$)	ADI ($\mu\text{g/kg bw per day}$)	Reference	EDI adults ($\mu\text{g/kg bw per day}$)	MOEs adults	EDI children ($\mu\text{g/kg bw per day}$)	MOEs children
TEP	0.03	125	na	Li et al., (2019b)	0.007	17,045	0.03	3964
TnBP	0.03	24	na		0.007	3484	0.03	810
TCPP	0.02	80	na		0.004	21,170	0.016	4924
BDE 47	0.002	0.1	na	US EPA (2017)	0.004	225	0.002	52
BDE 99	0.33	0.1	na		0.07	1.4	0.32	0.3
$\Sigma\text{penta BDEs}$	2.27	2	na		0.51	3.9	2.18	0.9
BDE 153	0.05	0.2	na		0.01	19	0.05	4.4
BDE 209	0.08	7	na	Huang et al., (2018)	0.02	404	0.07	94
$\Sigma\text{6m-PCBs}$	0.37	0.01	na	Arnich et al., (2009)	0.083	0.1	0.35	0.03
HCBu	0.001	na	0.2	Zhang et al., (2019)	0.0002	900	0.001	209
PeCB	0.002	na	na	–	0.0004	na	0.002	na
HCB	0.001	na	0.6	Mahmoud et al., (2016)	0.0002	2700	0.001	628
Chlordecone	0.0004	na	na	–	0.0001	na	0.0004	na
Mirex	0.02	na	na	–	0.004	na	0.02	na
ΣHCHs	0.07	na	5	Mahmoud et al., (2016)	0.02	331	0.06	77
ΣDDTs	0.004	na	10	Arisekar et al., 2022 (SI)	0.001	11,250	0.004	2616
Transchlordanane	0.003	na	na	–	0.001	13,548	0.003	3150

nd = not detected; na = not available.

and 31 for SOIL 2 (low risk), mainly attributed to OPEs (Table 4). In contrast, a very high risk was observed for SOIL 3, the dredge, caused by legacy PCB 180, PCB 153, endosulfans and PeCB which might reflect past contamination of the area. Some OPEs were also detected at concentrations causing a risk (Table 4). This dredge should also be considered hazardous according to concentrations detected and PNEC values. SOIL 4, the agricultural soil, had a RQ of 2450 caused mainly by the high concentrations of OPEs, which were the highest among soils. The applications of sewage sludge for agricultural purposes and irrigation with contaminated water are two main sources of OPEs in soils and the highest risk correspond to those compounds with the highest sorption capacity, as TPHP and TCP (Cristale et al., 2017b). Implications of soil toxicity on crops are to be further studied.

For waters, ΣRQs of OPEs were between 1.1 and 4.8 (Table 2), indicating a low risk. The highest RQ was found for the polluted pond (WAT1), mainly attributed to TCP 1 and TCP 2 with RQs >1 . The other compounds did not pose a risk ($\text{RQ} \ll 1$). WAT 2 and 3 from pond 2 and 4 had RQs of 1.4 and 2.8, respectively, and attributed mainly to TCP4, although the individual RQ were always <1 . WAT 4 from the canal showed the lowest risk, while WAT 5 corresponding to river water had a RQ of 2, attributed mainly to TCP4 that had a risk of 1.08, and all other individual OPEs had values <1 . Thus, the presence of OPEs in water produces a low risk and only compounds that could be of concern are TCPs, which despite being present at concentrations between 0.002 and 0.04 $\mu\text{g/L}$, their PNEC values are among the lowest for OPEs. Even in WAT 1, compounds detected at the highest concentrations, e.g. TPHP, TCPP and TiPP did not pose an environmental risk.

3.6. Comparison with other e-waste sites

E-waste recycling areas like Baihe village are so called hotspots of environmental pollution derived from the non-formal methods used to recover high market value materials from all types of EEE. Even with the policies and laws implemented since 2010 to ban e-waste disposal in small family workshops and the efforts made by the Qingyuan county government to relocate those potentially hazardous activities to modern

industrial facilities, high amounts of pollutants associated with e-waste are still present and distributed among environmental compartments. OPEs and PBDEs were the main pollutants detected in the Baihe e-waste, and similar findings are reported in other e-waste sites.

OPEs in sediments from Liangjiang River (Guiyu, Guangdong, China) had $\Sigma_{13}\text{OPFRs}$ between 0.22 and 14 $\mu\text{g/g dw}$ near the non-formal and uncontrolled e-waste dismantling area and from 0.032 to 0.42 $\mu\text{g/g dw}$ close to a flame retardants production site in Xiaoqing River (NW China, Shandong Province, China) (Chen et al., 2021), indicating that production can be less harmful than recycling activities. Another study in Guiyu reported $\Sigma_8\text{OPEs}$ from 6 to 2000 $\mu\text{g/g dw}$ in e-waste influenced sediments (Li et al., 2019a). OPEs in an e-waste site in northern Vietnam contained $\Sigma_{10}\text{OPEs}$ from 0.006 to 0.125 $\mu\text{g/g dw}$ in river sediments, and from 0.018 to 3.43 $\mu\text{g/g dw}$ in soils (Matsukami et al., 2015), indicating also that the area was impacted by these chemicals. Soils from a multi-waste recycling area in Tianjin (north-east China) showed OPEs concentration between 0.12 and 2.1 $\mu\text{g/g}$ (Wang et al., 2018b), and $\Sigma_7\text{OPEs}$ between 0.046 and 1.69 $\mu\text{g/g dw}$ were reported in an e-waste area in Pakistan, near Karachi city (Iqbal et al., 2017). In water, our results are comparable with those ones reported for waters from Taihu lake ($\Sigma_{11}\text{OPEs}$ between 0.16 and 1.53 $\mu\text{g/L}$) highly influenced by an OPEs manufacturing area (Wang et al., 2018a).

PBDEs are also ubiquitous in sediments and soils from e-waste recycling areas in China. $\Sigma_7\text{PBDEs}$ (without BDE 209) ranged from 0.005 to 0.03 $\mu\text{g/g dw}$, and BDE 209 was the main contributor ranging from 0.19 to 43 $\mu\text{g/g dw}$ in an e-waste site near Shenzhen city, Guangdong province, with no concentration gradient along the depth of the sediment (Zhou et al., 2019). In an abandoned e-waste recycling area in Guiyu, $\Sigma_{16}\text{PBDEs}$ ranged from 5 to 115 $\mu\text{g/g dw}$ in river sediments and from 0.04 to 7.2 $\mu\text{g/g dw}$ in soils (Wu et al., 2019). In a plastic recycling area in Hebei province, $\Sigma_{21}\text{PBDEs}$ ranging from 0.018 to 9.8 $\mu\text{g/g dw}$ were detected in sediments and between 0.001 and 5.5 $\mu\text{g/g dw}$ in soils (Tang et al., 2014). In crops soils affected by e-waste in Guangdong province, $\Sigma_9\text{PBDEs}$ ranged from 0.002 to 0.02 $\mu\text{g/g dw}$ and BDE 209 from 0.025 to 0.10 $\mu\text{g/g dw}$ as the dominating congener (Zou et al., 2007). Higher levels were detected in soils from another e-waste

Table 4

Risk quotients (RQs) calculated for each pollutant in soil and sediment samples. In red RQ ≥ 100, in orange 10 = RQ < 100, and in green RQ < 10, all in a degraded colour scale.

	PLAS	SED1	SED2	SED3	SOIL1	SOIL2	SOIL3	SOIL4
TPhP	64000	630	3.0	20	40	0.2	130	660
TCP2	32000	4200	8.3	105	80	4.4	89	760
T CPP	29000	84	2.0	3.8	16	0.7	52	190
TCP1	11600	2800	4.4	73	35	0.0	41	390
TCP4	8100	460	3.4	9.4	0.0	0.0	21	190
TCP3	4300	400	1.8	9.1	11	3.6	13	100
EHDPP	4000	32	3.0	2.2	8.7	2.3	7.7	110
TCEP	1100	220	18	20	141	6.2	145	37
BDE 47	500	18	0.2	0.0	1.7	0.1	1.6	0.0
BDE 99	370	17	0.2	0.0	2.3	0.1	4.2	0.0
TDCP	280	16	1.0	1.2	11	0.2	40	6.8
BDE 183	240	15	0.4	0.0	2.4	0.0	1.7	0.0
TiPP	74	12	0.0	0.9	0.0	9.4	1.0	8.0
Transchlordane	56	0.0	28	0.0	0.0	3.3	0.0	0.0
BDE 209	54	8.4	5.1	0.0	0.8	0.0	0.9	0.0
TEHP	47	11	0.4	0.3	0.0	0.3	1.9	1.1
BDE 66	41	1.3	0.0	0.0	0.1	0.0	0.1	0.0
Dieldrin	38	0.0	0.0	0.0	0.0	0.0	0.0	0.0
BDE 28	27	0.9	0.0	0.0	0.1	0.0	0.1	0.0
BDE 100	27	1.3	0.0	0.0	0.0	0.0	0.0	0.0
BDE 138	26	1.2	0.0	0.0	0.2	0.0	0.2	0.0
BDE 153	25	0.6	0.0	0.0	0.1	0.0	0.2	0.0
PCB 180	23	59	1.4	0.0	3.1	0.0	345	0.1
BDE 85	19	0.4	0.0	0.0	0.0	0.0	0.1	0.0
PCB 28	13	14	1.4	0.4	9.4	0.1	24	0.1
TnBP	7.4	0.5	0.4	0.1	0.1	0.1	0.5	0.2
PCB 52	7.0	9.1	1.1	0.1	5.4	0.0	5.6	0.0
BDE 154	5.7	0.2	0.0	0.0	0.0	0.0	0.1	0.0
PCB 153	4.3	66	1.4	0.3	5.4	0.0	280	0.1
PCB 101	3.9	11	2.7	0.3	8.2	0.1	28	0.0
TnPP	2.7	4.5	0.1	0.0	0.4	0.2	0.0	0.5
PCB 138	1.9	38	0.9	0.1	4.1	0.0	150	0.0
HCB	1.3	17	0.4	1.6	0.6	0.0	9.0	0.1
PeCB	0.0	550	0.0	48	28	0.0	130	0.0
α-endosulfan	0.0	0.0	0.0	0.0	0.0	0.0	2300	0.0
β-endosulfan	0.0	19300	0.0	0.0	0.0	0.0	49000	0.0
SUM	160000	29000	89	296	414	31	52500	2450

RQ rounded to significant digits.

recycling area, where Σ₄₁PBDEs were up to 13 µg/g dw and BDE 209 was again the dominant congener with levels ranging from 0.005 to 12 µg/g dw (Wang et al., 2014).

Legacy pollutants like PCBs have been also detected in different e-waste sites. Soils from Guiyu contained Σ₃₅PCBs between 0.0006 and 0.004 µg/g dw, which are much lower than the present study (Luo et al., 2020). Σ₂₆PCBs in 4 e-waste sites in India ranged from 0.0004 to 0.48 µg/g dw (Chakraborty et al., 2018). Lower levels at the pg/g dw were found for Σ₂₂PCBs in soils from a plastic recycling area in east China (Qin et al., 2022). In Zhejiang province, Σ₁₉PCBs ranged from 11.7 to 2.61 µg/g dw in an abandoned e-waste recycling facility and from 0.0035 to 0.051 µg/g dw in a newly built e-waste facility, suggesting that controlled EEE dismantling factories and proper management of residues can reduce PCBs emissions (Liu et al., 2020).

The presence of flame retardants or other pollutants in food produced near or in the surroundings of non-formal e-waste recycling

facilities or workshops can constitute a human risk considering that the diet of local peasants is based on local produced food. In this study we used hen eggs to evaluate the exposure through diet and the levels encountered are compared to other e-waste areas. Hen eggs close from the e-waste disassembling site and close to a municipal waste incinerator area from Zhejiang province had median Σ_{6marker}PCBs of 0.001 and 0.0002 µg/g ww, respectively (Shen et al., 2017). In an e-waste site in Taizhou, Zhejiang province, Σ_{indicator}PCBs in home produced hen eggs were of 0.004 µg/g ww (Xing et al., 2010). Σ₂₇PCBs at 0.035 µg/g ww and Σ₁₂PBDEs at 0.0004 µg/g ww (BDE 209 not analyzed) were found in eggs from Taizhou e-waste site (Zhao et al., 2009), and in Wenling Σ₇PBDEs mean concentration were of 0.038 µg/g ww (Qin et al., 2011), both in Zhejiang province. Chicken and goose home-produced eggs from Guiyu, one of the biggest e-waste areas in China, contained median Σ₁₇PBDEs values between 0.011 and 0.214 µg/g ww (0.4 and 7.7 µg/g lw), respectively (Zeng et al., 2016b). In chicken eggs grown near a

municipal dumpsite in Nabuja city, Nigeria, mean Σ_7 PBDEs ranged from 0.026 to 0.029 $\mu\text{g/g}$ ww but the study did not include BDE 209 (Oloruntoba et al., 2019). Therefore, considering the concentrations detected and that hen eggs constitute a nutritious and essential diet, special attention should be given to control the concentration of contaminants in areas affected by e-waste residues.

4. Conclusions

Baihe village is a well-documented e-waste hotspot where non-formal EEE recycling activities produce an impact to the environment and human health. In this paper, OPEs, followed by PBDEs and, in a lesser extent PCBs and OCPs, were detected in soils and sediments from the most polluted pond in the village but also in the nearby surrounding areas. The plastic lump which has been accumulated during years of illegal dumping of plastic residues from the e-waste recycling activities was identified as the main pollution source. Considering the concentrations detected in the plastic lump and the PNEC values, this plastic lump should be considered as a hazardous waste. Leaching of OPEs to water and accumulation in sediment and soils produced moderate to high risks for most contaminants, according to PNEC values. In addition, consumption of hen eggs reared in family farms can represent a risk especially for children due to the high concentrations of PCBs and PBDEs. Overall, this study highlights the need for the long-term monitoring of sites affected by EEE dismantling activities to protect both the environment and human health.

Credit authors statement

Pablo Zapata-Corella: Conceptualization, Investigation, Formal analysis, Writing – original draft, Writing – review & editing. **Zi-He Ren:** Conceptualization, Investigation. **Yin-E Liu:** Conceptualization, Investigation. **Silvia Lacorte:** Conceptualization, Investigation, Validation, Writing – review & editing, Supervision, Funding acquisition. **Anna Rigol:** Conceptualization, Investigation, Validation, Writing – review & editing, Supervision. **Xiao-Jun Luo:** Conceptualization, Investigation, Validation, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envres.2022.114768>.

References

- Arisekar, U., Shakila, R.J., Shalini, R., Jeyasekaran, G., Padmavathy, P., 2022. Effect of household culinary processes on organochlorine pesticide residues (OCPs) in the seafood (Penaeus vannamei) and its associated human health risk assessment: our vision and future scope. *Chemosphere* 297, 134075. <https://doi.org/10.1016/j.chemosphere.2022.134075>.
- Arnich, N., Tard, A., Leblanc, J.C., Le Bizec, B., Narbonne, J.F., Maximilien, R., 2009. Dietary intake of non-dioxin-like PCBs (NDL-PCBs) in France, impact of maximum levels in some foodstuffs. *Regul. Toxicol. Pharmacol.: RTP (Regul. Toxicol. Pharmacol.)* 54, 287–293. <https://doi.org/10.1016/j.yrtph.2009.05.010>.
- Awasthi, A.K., Zeng, X., Li, J., 2016. Environmental pollution of electronic waste recycling in India: a critical review. *Environ. Pollut.* 211, 259–270. <https://doi.org/10.1016/j.envpol.2015.11.027>.
- Backhaus, T., Faust, M., 2012. Predictive environmental risk assessment of chemical mixtures: a conceptual framework. *Environ. Sci. Technol.* 46, 2564–2573. <https://doi.org/10.1021/es2034125>.
- Bekele, T.G., Zhao, H., Yang, J., Chegen, R.G., Chen, J., Mekonen, S., Qadeer, A., 2021. A review of environmental occurrence, analysis, bioaccumulation, and toxicity of organophosphate esters. *Environ. Sci. Pollut. Res. Int.* 28, 49507–49528. <https://doi.org/10.1007/s11356-021-15861-8>.
- Cai, K., Song, Q., Yuan, W., Ruan, J., Duan, H., Li, Y., Li, J., 2020. Human exposure to PBDEs in e-waste areas: a review. *Environ. Pollut.* 267, 115634. <https://doi.org/10.1016/j.envpol.2020.115634>.
- Chakraborty, P., Selvaraj, S., Nakamura, M., Prithiviraj, B., Cincinelli, A., Bang, J.J., 2018. PCBs and PCDD/Fs in soil from informal e-waste recycling sites and open dumpsites in India: levels, congener profiles and health risk assessment. *Sci. Total Environ.* 621, 930–938. <https://doi.org/10.1016/j.scitotenv.2017.11.083>.
- Chen, P., Ma, S., Yang, Y., Qi, Z., Wang, Y., Li, G., Tang, J., Yu, Y., 2021. Organophosphate flame retardants, tetrabromobisphenol A, and their transformation products in sediment of e-waste dismantling areas and the flame-retardant production base. *Ecotoxicol. Environ. Saf.* 225, 112717. <https://doi.org/10.1016/j.ecoenv.2021.112717>.
- Stockholm Convention, 2009. COP4 final. *Earth Negotiation Bulletin* 15, 174, 11th May.
- Cristale, J., Dantas, R.F., De Luca, A., Sans, C., Esplugas, S., Lacorte, S., 2017a. Role of oxygen and DOM in sunlight induced photodegradation of organophosphorous flame retardants in river water. *J. Hazard Mater.* 323, 242–249. <https://doi.org/10.1016/j.jhazmat.2016.05.019>.
- Cristale, J., Álvarez-Martín, A., Rodríguez-Cruz, S., Sánchez-Martin, M.J., Lacorte, S., 2017b. Sorption and desorption of organophosphate esters with different hydrophobicity by soils. *Environ. Sci. Pollut. Res.* 24, 27870–27878. <https://doi.org/10.1007/s11356-017-0360-0>.
- de Ree, H., van den Berg, M., Brand, T., Mulder, G.J., Simons, R., van Zanten, B.V., Westerink, R.H.S., 2014. Health risk assessment of exposure to TriCresyl Phosphates (TCPs) in air-craft: a commentary. *Neurotoxicology* 45, 209–215. <https://doi.org/10.1016/j.neuro.2014.08.011>.
- ECHA, 2022. European Chemicals Agency. <https://echa.europa.eu/substance-information/-/substanceinfo/100.033.766>. accessed April 2022.
- Fan, Q., Zou, X., Gao, J., Cheng, Y., Wang, C., Feng, Z., Ding, Y., Zhang, C., 2021. Assessing ecological risk of organophosphate esters released from sediment with both of total content and desorbable content. *Sci. Total Environ.* 772, 144907. <https://doi.org/10.1016/j.scitotenv.2020.144907>.
- Forti, V., Balde, C.P., Kuehr, R., Bel, G., 2020. In: The Global E-Waste Monitor 2020: Quantities, Flows and the Circular Economy Potential. United Nations University (UNU)/United Nations Institute for Training and Research, Bonn/Geneva/Rotterdam (UNITAR) - co-hosted SCYCLE Programme, International Telecommunication Union (ITU) & International Solid Waste Association (ISWA). <https://ewastemonitor.info/gem-2020/>. Accessed October 2022.
- Han, Y., Tang, Z., Sun, J., Xing, X., Zhang, M., Cheng, J., 2019. Heavy metals in soil contaminated through E-waste processing activities in a recycling area: implications for risk management. *Process Saf. Environ. Protect.* 125, 189–196. <https://doi.org/10.1016/j.psep.2019.03.020>.
- Huang, C.C., Zeng, Y.H., Luo, X.J., Tang, B., Liu, Y.E., Ren, Z.H., Mai, B.X., 2018. Level changes and human dietary exposure assessment of halogenated flame retardant levels in free-range chicken eggs: a case study of a former e-waste recycling site, South China. *Sci. Total Environ.* 634, 509–515. <https://doi.org/10.1016/j.scitotenv.2018.03.386>.
- Iqbal, M., Syed, J.H., Breivik, K., Chaudhry, M.J.I., Li, J., Zhang, G., Malik, R.N., 2017. E-waste driven pollution in Pakistan: the first evidence of environmental and human exposure to flame retardants (FRs) in Karachi city. *Environ. Sci. Technol.* 51, 13895–13905. <https://doi.org/10.1021/acs.est.7b03159>.
- Li, W., Achal, V., 2020. Environmental and health impacts due to e-waste disposal in China - a review. *Sci. Total Environ.* 737, 139745. <https://doi.org/10.1016/j.scitotenv.2020.139745>.
- Li, H., La Guardia, M.J., Liu, H., Hale, R.C., Mainor, T.M., Harvey, E., Sheng, G., Fu, J., Peng, P., 2019a. Brominated and organophosphate flame retardants along a sediment transect encompassing the Guiyu, China e-waste recycling zone. *Sci. Total Environ.* 646, 58–67. <https://doi.org/10.1016/j.scitotenv.2018.07.276>.
- Li, J., Zhao, L., Letcher, R.J., Zhang, Y., Jian, K., Zhang, J., Su, G., 2019b. A review on organophosphate Ester (OPE) flame retardants and plasticizers in foodstuffs: levels, distribution, human dietary exposure, and future directions. *Environ. Int.* 127, 35–51. <https://doi.org/10.1016/j.envint.2019.03.009>.
- Liu, Y., Luo, X.J., Huang, L.Q., Tao, L., Zeng, Y.H., Mai, B.X., 2018a. Halogenated organic pollutants in aquatic, amphibious, and terrestrial organisms from an e-waste site: habitat-dependent accumulation and maternal transfer in watersnake. *Environ. Pollut.* 241, 1063–1070. <https://doi.org/10.1016/j.envpol.2018.06.038>.

- Liu, Y.E., Huang, L.Q., Luo, X.J., Tan, X.X., Huang, C.C., Corella, P.Z., Mai, B.X., 2018b. Determination of organophosphorus flame retardants in fish by freezing-lipid precipitation, solid-phase extraction and gas chromatography-mass spectrometry. *J. Chromatogr., A* 1532, 68–73. <https://doi.org/10.1016/j.chroma.2017.12.001>.
- Liu, Y.E., Luo, X.J., Zapata Corella, P., Zeng, Y.H., Mai, B.X., 2019. Organophosphorus flame retardants in a typical freshwater food web: bioaccumulation factors, tissue distribution, and trophic transfer. *Environ. Pollut.* 255, 113286 <https://doi.org/10.1016/j.envpol.2019.113286>.
- Liu, C., Wei, B.K., Bao, J.S., Wang, Y., Hu, J.C., Tang, Y.E., Chen, T., Jin, J., 2020. Polychlorinated biphenyls in the soil-crop-atmosphere system in e-waste dismantling areas in Taizhou: concentrations, congener profiles, uptake, and translocation. *Environ. Pollut.* 257, 113622 <https://doi.org/10.1016/j.envpol.2019.113622>.
- Lu, C., Zhang, L., Zhong, Y., Ren, W., Tobias, M., Mu, Z., Ma, Z., Geng, Y., Xue, B., 2014. An overview of e-waste management in China. *J. Mater. Cycles Waste Manag.* 17, 1–12. <https://doi.org/10.1007/s10163-014-0256-8>.
- Luo, C., Hu, B., Wang, S., Wang, Y., Zhao, Z., Wang, Y., Li, J., Zhang, G., 2020. Distribution and chiral signatures of polychlorinated biphenyls (PCBs) in soils and vegetables around an e-waste recycling site. *J. Agric. Food Chem.* 68, 10542–10549. <https://doi.org/10.1021/acs.jafc.0c00479>.
- Ma, W.J., Deng, F., Xu, Y.J., Xu, H.F., 2005. The study on dietary intake and nutritional status of residents in Guangdong, 2002. *Guangdong J. Health Epidemic Prev.* 31 (1), 1–5.
- Mahmoud, A.F., Ikenaka, Y., Yohannes, Y.B., Darwish, W.S., Eldaly, E.A., Morshdy, A.E., Nakayama, S.M., Mizukawa, H., Ishizuka, M., 2016. Distribution and health risk assessment of organochlorine pesticides (OCPs) residue in edible cattle tissues from northeastern part of Egypt: high accumulation level of OCPs in tongue. *Chemosphere* 144, 1365–1371. <https://doi.org/10.1016/j.chemosphere.2015.10.016>.
- Matsukami, H., Tse, N.M., Suzuki, G., Someya, M., Tuyen, L.H., Viet, P.H., Takahashi, S., Tanabe, S., Takigami, H., 2015. Flame retardant emission from e-waste recycling operation in northern Vietnam: environmental occurrence of emerging organophosphorus esters used as alternatives for PBDEs. *Sci. Total Environ.* 514, 492–499. <https://doi.org/10.1016/j.scitotenv.2015.02.008>.
- Nandy, S., Fortunato, E., Martins, R., 2022. Green economy and waste management: an inevitable plan for materials science. *Prog. Nat. Sci.: Mater. Int.* 32, 1–9. <https://doi.org/10.1016/j.pnsc.2022.01.001>.
- NORMAN ecotoxicology database, 2021. Network of Reference Laboratories, Research Centres and Related Organisations for Monitoring of Emerging Environmental Substances. <https://www.norman-network.com/nds/ecotox/lowestPnscIndex.php>. accessed november 2021.
- Oloruntoba, K., Sindiku, O., Osibanjo, O., Balan, S., Weber, R., 2019. Polybrominated diphenyl ethers (PBDEs) in chicken eggs and cow milk around municipal dumpsites in Abuja, Nigeria. *Ecotoxicol. Environ. Saf.* 179, 282–289. <https://doi.org/10.1016/j.ecoenv.2019.04.045>.
- Perkins, D.N., Brune Drisse, M.N., Nxele, T., Sly, P.D., 2014. E-waste: a global hazard. *Annals of global health* 80, 286–295. <https://doi.org/10.1016/j.aogh.2014.10.001>.
- Poma, G., Sales, C., Bruylant, B., Christia, C., Goscinny, S., Van Loco, J., Covaci, A., 2018. Occurrence of organophosphorus flame retardants and plasticizers (PFRs) in Belgian foodstuffs and estimation of the dietary exposure of the adult population. *Environ. Sci. Technol.* 52, 2331–2338. <https://doi.org/10.1021/acs.est.7b06395>.
- Qin, X., Qin, Z., Li, Y., Zhao, Y., Xia, X., Yan, S., Tian, M., Zhao, X., Xu, X., Yang, Y., 2011. Polybrominated diphenyl ethers in chicken tissues and eggs from an electronic waste recycling area in southeast China. *J. Environ. Sci.* 23, 133–138. [https://doi.org/10.1016/s1001-0742\(10\)60384-1](https://doi.org/10.1016/s1001-0742(10)60384-1).
- Qin, Y., Liu, Y., Wang, J., Lu, Y., Xu, Z., 2022. Emission of PAHs, PCBs, PBDEs and heavy metals in air, water and soil around a waste plastic recycling factory in an industrial park, Eastern China. *Chemosphere* 294, 133734. <https://doi.org/10.1016/j.chemosphere.2022.133734>.
- Shaikh, S., Thomas, K., Zuhair, S., 2020. An exploratory study of e-waste creation and disposal: upstream considerations. *Resour. Conserv. Recycl.* 155, 104662 <https://doi.org/10.1016/j.resconrec.2019.104662>.
- Shen, H., Guan, R., Ding, G., Chen, Q., Lou, X., Chen, Z., Zhang, L., Xing, M., Han, J., Wu, Y., 2017. Polychlorinated dibenzo-p-dioxins/furans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in Zhejiang foods (2006-2015): market basket and polluted areas. *Sci. Total Environ.* 574, 120–127. <https://doi.org/10.1016/j.scitotenv.2016.09.038>.
- Tang, Z., Huang, Q., Cheng, J., Yang, Y., Yang, J., Guo, W., Nie, Z., Zeng, N., Jin, L., 2014. Polybrominated diphenyl ethers in soils, sediments, and human hair in a plastic waste recycling area: a neglected heavily polluted area. *Environ. Sci. Technol.* 48, 1508–1516. <https://doi.org/10.1021/es404905u>.
- US EPA, 2012. Estimation Programs Interface Suite™ for Microsoft® Windows, 4.1. United States Environmental Protection Agency, Washington, DC, USA.
- US EPA, 2017. Technical Fact Sheet - Polybrominated Diphenyl Ethers (PBDEs). EPA 505-F-17-015. <https://www.epa.gov/fedfac/technical-fact-sheet-polybrominated-diphenyl-ethers-pbdes-and-polybrominated-biphenyls-pbbs>. accessed October 2022.
- Vagi, M.C., Petsas, A.S., Kostopoulou, M.N., 2021. Potential effects of persistent organic contaminants on marine biota: a review on recent research. *Water* 13, 2488. <https://doi.org/10.3390/w13182488>.
- Wang, S., Zhang, S., Huang, H., Niu, Z., Han, W., 2014. Characterization of polybrominated diphenyl ethers (PBDEs) and hydroxylated and methoxylated PBDEs in soils and plants from an e-waste area, China. *Environ. Pollut.* 184, 405–413. <https://doi.org/10.1016/j.envpol.2013.09.021>.
- Wang, X., Zhu, L., Zhong, W., Yang, L., 2018a. Partition and source identification of organophosphate esters in the water and sediment of Taihu Lake, China. *J. Hazard Mater.* 360, 43–50.
- Wang, Y., Sun, H., Zhu, H., Yao, Y., Chen, H., Ren, C., Wu, F., Kannan, K., 2018b. Occurrence and distribution of organophosphate flame retardants (OPFRs) in soil and outdoor settled dust from a multi-waste recycling area in China. *Sci. Total Environ.* 625, 1056–1064. <https://doi.org/10.1016/j.scitotenv.2018.01.013>.
- Wang, X., Zhu, Q., Liao, C., Jiang, G., 2021. Human internal exposure to organophosphate esters: a short review of urinary monitoring on the basis of biological metabolism research. *J. Hazard Mater.* 418, 126279 <https://doi.org/10.1016/j.jhazmat.2021.126279>.
- Wu, J.P., Luo, X.J., Zhang, Y., Yu, M., Chen, S.J., Mai, B.X., Yang, Z.Y., 2009. Biomagnification of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls in a highly contaminated freshwater food web from South China. *Environ. Pollut.* 157, 904–909. <https://doi.org/10.1016/j.envpol.2008.11.001>.
- Wu, Q., Du, Y., Huang, Z., Gu, J., Leung, J.Y.S., Mai, B., Xiao, T., Liu, W., Fu, J., 2019. Vertical profile of soil/sediment pollution and microbial community change by e-waste recycling operation. *Sci. Total Environ.* 669, 1001–1010. <https://doi.org/10.1016/j.scitotenv.2019.03.178>.
- Xing, G.H., Wu, S.C., Wong, M.H., 2010. Dietary exposure to PCBs based on food consumption survey and food basket analysis at Taizhou, China - the world's major site for recycling transformers. *Chemosphere* 81, 1239–1244. <https://doi.org/10.1016/j.chemosphere.2010.09.032>.
- Zapata, P., Ballesteros-Cano, R., Colomer, P., Bertolero, A., Viana, P., Lacorte, S., Santos, F.J., 2018. Presence and impact of Stockholm Convention POPs in gull eggs from Spanish and Portuguese natural and national parks. *Sci. Total Environ.* 633, 704–715. <https://doi.org/10.1016/j.scitotenv.2018.03.081>.
- Zeng, Y.H., Tang, B., Luo, X.J., Zheng, X.B., Peng, P.A., Mai, B.X., 2016a. Organohalogen pollutants in surface particulates from workshop floors of four major e-waste recycling sites in China and implications for emission lists. *Sci. Total Environ.* 569–570, 982–989. <https://doi.org/10.1016/j.scitotenv.2016.06.053>.
- Zeng, Y.H., Luo, X.J., Tang, B., Mai, B.X., 2016b. Habitat- and species-dependent accumulation of organohalogen pollutants in home-produced eggs from an electronic waste recycling site in South China: levels, profiles, and human dietary exposure. *Environ. Pollut.* 216, 64–70. <https://doi.org/10.1016/j.envpol.2016.05.039>.
- Zeng, Y., Huang, C., Luo, X., Liu, Y., Ren, Z., Mai, B., 2018. Polychlorinated biphenyls and chlorinated paraffins in home-produced eggs from an e-waste polluted area in South China: occurrence and human dietary exposure. *Environ. Int.* 116, 52–59. <https://doi.org/10.1016/j.envint.2018.04.006>.
- Zhang, N., Yang, Y., Tao, S., Liu, Y., Shi, K.L., 2011. Sequestration of organochlorine pesticides in soils of distinct organic carbon content. *Environ. Pollut.* 159, 700–705. <https://doi.org/10.1016/j.envpol.2010.12.011>.
- Zhang, H., Shen, Y., Liu, W., He, Z., Fu, J., Cai, Z., Jiang, G., 2019. A review of sources, environmental occurrences and human exposure risks of hexachlorobutadiene and its association with some other chlorinated organics. *Environ. Pollut.* 253, 831–840. <https://doi.org/10.1016/j.envpol.2019.07.090>.
- Zhao, G., Zhou, H., Wang, D., Zha, J., Xu, Y., Rao, K., Ma, M., Huang, S., Wang, Z., 2009. PBBs, PBDEs, and PCBs in foods collected from e-waste disassembly sites and daily intake by local residents. *Sci. Total Environ.* 407, 2565–2575. <https://doi.org/10.1016/j.scitotenv.2008.11.062>.
- Zheng, X.B., Wu, J.P., Luo, X.J., Zeng, Y.H., She, Y.Z., Mai, B.X., 2012. Halogenated flame retardants in home-produced eggs from an electronic waste recycling region in South China: levels, composition profiles, and human dietary exposure assessment. *Environ. Int.* 45, 122–128. <https://doi.org/10.1016/j.envint.2012.04.006>.
- Zheng, X., Xu, F., Chen, K., Zeng, Y., Luo, X., Chen, S., Mai, B., Covaci, A., 2015. Flame retardants and organochlorines in indoor dust from several e-waste recycling sites in South China: composition variations and implications for human exposure. *Environ. Int.* 78, 1–7. <https://doi.org/10.1016/j.envint.2015.02.006>.
- Zhou, P., Zhao, Y., Li, J., Wu, G., Zhang, L., Liu, Q., Fan, S., Yang, X., Li, X., Wu, Y., 2012. Dietary exposure to persistent organochlorine pesticides in 2007 Chinese total diet study. *Environ. Int.* 42, 152–159. <https://doi.org/10.1016/j.envint.2011.05.018>.
- Zhou, H., Tam, N.F.Y., Cheung, S.G., Wei, P., Li, S., Wu, Q., 2019. Contamination of polybrominated diphenyl ethers (PBDEs) in watershed sediments and plants adjacent to e-waste sites. *J. Hazard Mater.* 379, 120788 <https://doi.org/10.1016/j.jhazmat.2019.120788>.
- Zou, M.Y., Ran, Y., Gong, J., Mai, B.X., Zeng, E.Y., 2007. Polybrominated diphenyl ethers in watershed soils of the Pearl River Delta, China: occurrence, inventory, and fate. *Environ. Sci. Technol.* 41, 8262–8267. <https://doi.org/10.1021/es071956d>.