Science of the Total Environment 649 (2019) 821–828

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Spatial distributions and transport implications of short- and medium-chain chlorinated paraffins in soils and sediments from an e-waste dismantling area in China

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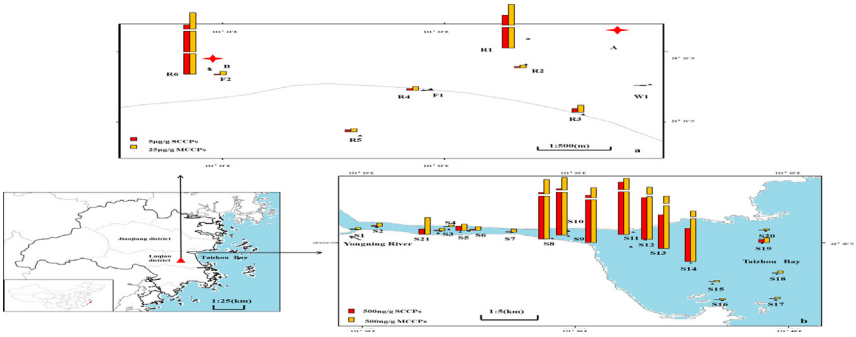
HIGHLIGHTS

- SCCPs and MCCPs in soils and sediments in an e-waste dismantling area were analyzed.
- The spatial distributions were closely related to e-waste pollution.
- Transportation via the air/water and deposition may be the main transport pathways.
- Ecological risks posed by CPs in soils and sediments were assessed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 9 July 2018
Received in revised form 21 August 2018
Accepted 25 August 2018
Available online 27 August 2018

Editor: Jay Gan

Keywords:
Short-chain chlorinated paraffin
Medium-chain chlorinated paraffin
E-waste dismantling area
Sediment
Soil

ABSTRACT

To investigate the spatial distributions, potential transport and ecological risks of chlorinated paraffins (CPs) in and around e-waste dismantling area, we collected soil samples within 5 km of the e-waste dismantling centers and sediment samples in the surrounding area from the lower reaches of Jiaojiang River. Short- and medium-chain chlorinated paraffins (SCCPs and MCCPs) were analyzed by two-dimensional gas chromatography coupled with electron-capture negative-ionization mass spectrometry (GC × GC-ECNI-MS). The SCCP and MCCP concentration ranges in soils were 68.5 to 2.20×10^5 ng/g dry weight (dw) and 507 to 4.40×10^6 ng/g dw, respectively. The ranges for the levels of SCCPs and MCCPs in sediments were 32.5–1.29×10^4 ng/g dw and 271–2.72×10^4 ng/g dw, respectively. No significant correlation was observed between total organic carbon (TOC) and CP concentrations ($P > 0.05$). The spatial distributions showed that the CP levels were closely related to e-waste pollution. Correspondence analysis revealed that shorter-chain and less chlorinated congeners were enriched in sediments from sites distant from e-waste pollution source, while longer-chain and higher chlorinated congeners were concentrated in soils and sediments collected near the pollution source, which indicated that complex environmental processes, such as transportation via atmosphere and/or water, and deposition, resulted in different CP profiles in different sampling locations and environment matrixes (e.g., soil and sediments).

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https://doi.org/10.1016/j.scitotenv.2018.08.355
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Principal component analysis (PCA) indicated that e-waste pollution could be the same source of SCCPs and MCCPs. The preliminary risk assessment indicated that CPs in soils within 1 km of e-waste dismantling centers at current levels posed a considerable risk to soil-dwelling organisms, and the sediment MCCPs in Jiaojiang estuary at present levels also posed a risk to sediment-dwelling organisms.

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1. Introduction

Chlorinated paraffins (CPs) or polychlorinated alkanes are extremely complex industrial mixtures that consist of thousands of homologs and isomers (Feo et al., 2009). According to the carbon chain length, CPs can be divided into short-chain CPs (SCCPs, $C_{10–13}$), medium-chain CPs (MCCPs, $C_{14–17}$) and long-chain CPs (LCCPs, $C_{18–30}$). CPs have been widely used as plasticizers and flame retardants, and as additives in lubricants, sealants, and cutting fluids (Bayen et al., 2006). Because of their mass production and widespread application, CPs are inevitably released into the environment during their production, storage, transportation and industrial use, and by release from manufactured products (Bayen et al., 2006). To date, CPs have been detected in many different environmental media such as air (Wang et al., 2013), water (Zeng et al., 2011b), soil (Wang et al., 2014), sediment (Marvin et al., 2003), and biota (Sun et al., 2017). There is growing concern about SCCPs worldwide because of their long-distance migration (Tomy et al., 1999), persistence (Iozza et al., 2008), bioaccumulation (Ma et al., 2014), and toxicity (Bezchlebova et al., 2007). The manufacture and use of SCCPs is prohibited or restricted in the European Union, United States, Japan, and Canada (van Mourik et al., 2016). Since May 2017, SCCPs have been included in the controlled list of Annex A of the Stockholm Convention on Persistent Organic Pollutants (POPs) (POPRC, 2017).

Recently, environmental problems caused by electronic waste (e-waste) dismantling have arisen around the world. The distributions, environmental behaviors and risk assessment of POPs, such as polybrominated diphenyl ethers (PBDEs) (Leung et al., 2007), have been investigated in e-waste dismantling areas. CPs are used as plasticizers and flame retardants in electrical devices and can be released to the ambient environment during improper dismantling processes (Fiedler, 2010). Because of their high K_{oc} values, CPs mainly distribute into soil and sediment phases when released into the environment (Fisk et al., 1998). Previous studies have shown that SCCPs are present in soils (Chen et al., 2014b), surface particulates (Zeng et al., 2016), paddy seeds, and snails (Yuan et al., 2017) in the e-waste dismantling area from Taizhou. Chen et al. observed that the concentrations of CPs in sediments from e-waste recycling ponds were higher than those in other regions in the Pearl River Delta (PRD), South China (Chen et al., 2011). What's more, bioaccumulations of CPs have been found in aquatic and terrestrial organisms (Luo et al., 2015; Sun et al., 2017), which indicated that e-waste dismantling could be an important source of CPs. However, few studies have investigated transport pathways of CPs released by e-waste processing in the e-waste dismantling area. Once released, CPs can undergo long-range transportation from high to low industry activities areas (Chen et al., 2011; Wang et al., 2013), and may pose potential risks to ecosystems and human health in the surrounding environment. Moreover, information on CPs, especially MCCPs, in e-waste dismantling areas is still scare. To the best of our knowledge, no information on the occurrences of MCCPs in soils from e-waste dismantling areas has been available before now. Although MCCPs show lower toxicity than SCCPs (Wei et al., 2016; Wyatt et al., 1993), they exhibit bioaccumulation (Houde et al., 2008) and even higher levels than SCCPs (Chen et al., 2011; Wang et al., 2013). Both SCCPs and MCCPs in e-waste dismantling regions deserve more attention.

Taizhou, located in the southeast of Zhejiang Province, is one of the two largest e-waste recycling areas in China (Fu et al., 2011), and up to 2 million tons of e-waste was dismantled annually (Han et al., 2009). The Jiaojiang River is the largest waterbody in Taizhou. Pollutants, including CPs, can be moved from the e-waste dismantling area to the Jiaojiang River by atmospheric or waterborne transport (Zhou et al., 2012). Because they are hydrophobic, CPs typically combine with particulate matter and then accumulate into sediments (Feo et al., 2009). In addition, ports where large quantities of e-waste are loaded and unloaded are located near the river estuary and could be potential sources of CP pollution. Therefore, a comprehensive investigation into the distribution and transportation of CPs was considered necessary to evaluate the impact of CPs released by e-waste processing on e-waste dismantling areas and the surrounding environment.

In this study, we used two-dimensional gas chromatography coupled with electron-capture negative-ionization mass spectrometry (GC × GC-ECNI-MS) to analyze SCCPs and MCCPs in soils and sediments from an e-waste dismantling area in China. The objectives of this study were to study the contamination status and spatial distributions of SCCPs and MCCPs in e-waste dismantling area and the surrounding area. From these data, the transport pathways of CPs were then further explored and the influencing factors related to CP congener patterns were revealed. Finally, preliminary ecological risks posed by CPs were assessed.

2. Materials and methods

2.1. Sampling

Fengjiang (A) and Baifeng'ao (B) are typical e-waste dismantling centers in Taizhou (Chen et al., 2014b). The e-waste dismantling area covers a radius of approximate 20 km from Fengjiang center (Yuan et al., 2017). Jiaojiang River is about within 20–30 km from the center. Nine surface soil samples were collected within 5 km of these two centers using a stainless steel scoop in June 2017 (Fig. S1a). All soil samples were classified as farmland soil (F1 and F2), woodland soil (W1), or roadside soil (R1–R6) according to land use. For each sampling site, five soil subsamples were taken from the same area (at a depth of 0–20 cm) and then mixed together to form one composite sample. At the same time, 21 surface sediment samples were collected from the lower reaches of the Jiaojiang River in Taizhou (Fig. S1b) using a grab sampler. The geographical location of the ports near Jiaojiang estuary limits the collection of soil samples. All samples were air-dried at room temperature. After removing stones and residual roots, the samples were sieved through a 60-mesh sieve and then sealed in amber glass bottles and conserved at −20 °C until required for analysis. The soil and sediment samples were analyzed for their total organic carbon (TOC) contents (Table S1).

2.2. Chemicals and materials

Three standard SCCP mixtures (with chlorine contents of 51.5%, 55.5%, and 63.0%) and three standard MCCP mixtures (with chlorine contents of 42.0%, 52.0%, and 57.0), all at 100 ng/μL in cyclohexane, were obtained from Dr. Ehrenstorfer (Augsburg, Germany). SCCP solutions with chlorine contents of 53.5% and 59.2% were prepared by 1:1 (v/v) mixing of the 51.5% + 55.5% and 55.5% + 63% standard solutions, respectively. In the same way, MCCP solutions with chlorine contents of 47% and 54.5% were obtained by mixing the 42% + 52% and 52% + 57% standard solutions, respectively. $^{13}C_{10}$-*trans*-Chlordane (Cambridge

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