



Concentrations and occupational exposure assessment of polybrominated diphenyl ethers in modern Chinese e-waste dismantling workshops

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HIGHLIGHTS

- PBDE emissions from modern Chinese e-waste recycling processes were investigated.
- PBDE concentrations in air and dust varied among e-waste recycling workshops.
- The atmospheric concentrations of PBDEs in office area were quite low.
- Fly ash samples were firstly reported with high PBDEs concentration.

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ABSTRACT

In this work, the concentrations of polybrominated diphenyl ethers (PBDEs) were determined in air, dust and fly ash samples from three legal waste electrical and electronic equipment dismantling plants with strict pollution controls. The risks posed by PBDEs to workers at the plants were assessed. The atmospheric concentrations of PBDEs in the different e-waste recycling workshops were $0.58\text{--}2.89 \times 10^3 \text{ ng/m}^3$, and predominantly distributed in the particle phase (90.7%–99.9%). The concentrations of the PBDEs in the floor dust and fly ash samples from bag-type dust collectors in different workshops were 2.39–125 $\mu\text{g/g}$, 5.84–128 $\mu\text{g/g}$, respectively. The contributions of BDE-209 in air, floor dust and fly ash samples were 84.0%–97.9%, 11.2%–95.3% and 74.0%–94.9%, respectively, indicating that deca-BDE commercial formulations were their major sources. Daily exposure to PBDEs was also lower than has been found for workers in other recycling workshops. Human exposures to BDE-47, BDE-99, BDE-153, and BDE-209 were all below the levels considered to pose appreciable risks. Dust ingestion was the main exposure route for manual recyclers, and inhalation was the main exposure route for waste transportation workers. The results of this study indicate that PBDEs emissions and risks are lower in modern, legal e-waste recycling facilities with effective pollution controls. However, the effectiveness of the pollution controls need to be further researched in plastic crushing areas.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs), one of the most widely used flame retardants, are added to plastics and textiles as well as electrical and electronic equipment (Frank et al., 2001; Besis et al., 2016). Large amounts of PBDEs have been produced and used

around the world, particularly in China (Meng et al., 2007). Tetra- to octa-bromodiphenyl ethers and deca-bromodiphenyl ether commercial formulations have been added to Annex A of the Stockholm Convention on Persistent Organic Pollutants (UNEP, 2017).

Numerous waste electrical and electronic equipment (WEEE) have been recycled to recover valuable materials via manual dismantling, burning, nitric acid treatment, mercury amalgamation, and cyanide salt leaching of waste printed circuit boards (Fu et al., 2008; Labunska et al., 2015; Matsukami et al., 2015). However, there are serious concerns about the environmental pollution caused by the release of PBDEs from discarded WEEEs or from those that are recycled (Qu et al., 2007; Chan et al., 2013; Jiang et al.,

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2014). High PBDEs concentrations have been found in air, soil, water, sediment, and animal and plant tissues (including breast milk) in the Guiyu and Taizhou areas of China, where there are many illegal WEEE recycling workshops (Zheng et al., 2008; Tang et al., 2010), which has drawn attention from around the world.

Increasing concern about PBDEs emissions has led many countries to seek to decrease the amounts of toxic chemicals used in electronic products, to establish appropriate WEEE treatment and recycling processes, to decrease the amount of WEEE produced, and to recover valuable resources from WEEE. The Chinese government has established regulations (especially a WEEE processing fund) to promote and normalize appropriate WEEE recycling processes. These regulations require recycling sites to be located in designated industrial zones and to use approved recycling equipment and pollution control facilities (such as dust removal facilities and negative pressure operating tables). In 2016, a total of 7.94×10^7 WEEE units were dismantled at 109 legal enterprises monitored by local environmental protection bureaus (Ministry of Environmental Protection of People's Republic of China, 2017).

However, in a survey, we found that the environmental impact assessments performed by these recycling plants were deficient, as only the environmental risks posed by total suspended particulate (TSP) emissions were taken into account. It is well-known that the plastics in WEEE can contain PBDEs and that these PBDEs can be easily released as dust that is produced when WEEE are processed. It is therefore inappropriate to evaluate the pollution risks and occupational exposure risk of WEEE recycling plants only in terms of TSP emissions. It is important to assess the risks posed by PBDEs emissions from WEEE recycling plants using centralized and normalized processes with strict pollution control measures.

The objective of this study is to determine the PBDEs concentrations and congener profiles in air, floor dust (FD), and fly ash from bag-type dust collectors (BD) in different modern e-waste recycling workshops, and to estimate the daily PBDE intake of workers through inhalation and dust ingestion.

2. Materials and methods

2.1. Study area

Three legal WEEE recycling plants (known here as plants A, B, and C) that benefit from the Chinese WEEE fund were selected based on the amount of e-waste processed at all 109 legal WEEE recycling facilities in China (Ministry of Environmental Protection of People's Republic of China, 2017). Plant A is in an industrial area in the center of China, and had been recycling e-waste for only two years when the study was performed. Approximately 2.0×10^6 e-waste units (including refrigerators, washing machines, and televisions (TVs)) are dismantled at plant A each year using the most up-to-date automatic and manual dismantling techniques. Plants B and C are located in a village and small town in Southwest China, respectively. Plants B and C dismantle approximately 5.0×10^5 and 2.0×10^5 e-waste units, respectively, each year (corresponding to the middle and lower ends, respectively, of the scale of Chinese WEEE disassembly plants) (Ministry of Environmental Protection of People's Republic of China, 2017). The e-waste recycling processes used in these plants had been implemented for 3 years when the study was performed. The dismantling lines in all of the plants contained dust removal facilities and negative pressure operating tables.

2.2. Sampling

Air samples (287–899 m³) were collected from the plants in May and June 2015 using Echo Hi-Vol high-flow air samplers (TCR

Tecora, Fontenay sous Bois, France). The sampler was equipped with a glass fiber filter (GFF) for sampling particle-bound PBDEs, followed by a glass cartridge containing polyurethane foam (PUF; 6.3 cm diameter, 7.6 cm length) for sampling gaseous chemicals. Each GFF was weighed before and after sampling to determine the TSP concentration. The air samplers were placed 1 m away from the dismantling lines and were collected from different workshops only when WEEE were being dismantled. The details of the location of the air samples from plants A, B, and C are presented in Table 1. An FD sample from each workshop was collected using a broom in each workshop. Fly ash samples from bag-type dust collectors (labeled BD) were also collected at the end of the sampling period. Each sample was wrapped in aluminum foil and transported to the laboratory. The FD and BD samples were screened using 80 meshes sieves (0.180 mm) to remove any debris. All of the samples were stored at -20°C until analysis.

2.3. Sample extraction and purification

The methods used to extract and purify the samples have been described previously (Nie et al., 2015; Tang et al., 2016). Briefly, a sample was spiked with surrogate standards (¹³C-labeled polychlorinated biphenyl #141, 3.84 ng) to monitor the recoveries. Then they were extracted for 24 h using 200 mL of a 1:1 v/v mixture of acetone and hexane in the Soxhlet Apparatus. The extract was concentrated and exchanged into hexane before fractionation by passing it through a multilayer silica/alumina column. The PBDEs were eluted and a clean extract was evaporated prior to concentration to incipient dryness by a gentle nitrogen stream. Finally, 200 μL of the 19.2 ng/mL internal standard (¹³C-labeled polychlorinated biphenyl #208) was added before the extract was analyzed by gas chromatography mass spectrometry.

2.4. Instrumental analysis

A total of 21 PBDE congeners (BDE-28, BDE-47, BDE-66, BDE-85, BDE-99, BDE-100, BDE-138, BDE-153, BDE-154, BDE-183, BDE-190, BDE-196, BDE-197, BDE-201, BDE-202, BDE-203, BDE-205, BDE-206, BDE-207, BDE-208, and BDE-209) were analyzed in this study (as presented in the supplemental material). The concentrations of PBDEs in the sample extracts were determined using an Agilent 6890N gas chromatograph with an Agilent 5973 inert mass selective detector (MSD) (Agilent Technologies, Santa Clara, CA, USA), which was used in electron capture negative chemical ionization mode. Separation was achieved using a J&W DB-5 MS capillary column (15 m long, 0.25 mm i.d., 0.11 μm film thickness; Agilent Technologies). The column and instrument parameters used were selected based on recent studies (Nie et al., 2015; Tang et al., 2016). The carrier gas was helium, and it which was supplied at a constant flow rate of 1.8 mL/min for a linear velocity of 70 cm/s. The oven temperature started at 90 °C and was held for 1 min before increasing at 20 °C/min to 340 °C, which was held for 2 min. The injector, ion source, and quadrupole temperatures were 280, 230, and 150 °C, respectively. The reagent gas was high purity methane. The mass selective detector was operated in selected ion monitoring mode.

2.5. Quality assurance and quality control

A procedural blank (10.0 g of acetone-washed diatomite) and a spiked blank (5.00 ng of each lighter congeners and 10.0 ng of each of heavier congeners) were run with each batch of 10 samples to assess any potential sample contamination and to perform a recovery analysis. The analyte concentrations in the procedural blanks were lower than the detection limits, with the exception of

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