



# PCBs and PCDD/Fs in soil from informal e-waste recycling sites and open dumpsites in India: Levels, congener profiles and health risk assessment

Paromita Chakraborty<sup>a,b,\*</sup>, Sakthivel Selvaraj<sup>a</sup>, Masafumi Nakamura<sup>c</sup>, Balasubramanian Prithiviraj<sup>a</sup>, Alessandra Cincinelli<sup>d</sup>, John J. Bang<sup>e</sup>

<sup>a</sup> SRM Research Institute, SRM University, Kattankulathur, Tamil Nadu 603203, India

<sup>b</sup> Department of Civil Engineering, SRM University, Kattankulathur, Tamil Nadu 603203, India

<sup>c</sup> Hiyoshi Corporation, Kitanosho 908, Omihachiman, Shiga 523-0806, Japan

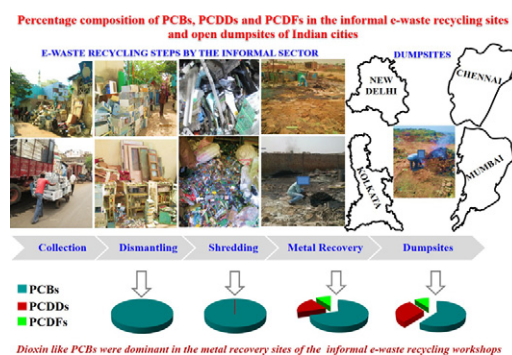
<sup>d</sup> Department of Chemistry "Ugo Schiff", Via della Lastruccia, 3, 50019 Sesto Fiorentino, Florence, Italy

<sup>e</sup> Department of Environmental, Earth and Geospatial Science, NCCU, Durham, NC, USA

## HIGHLIGHTS

- Over 60%  $\Sigma$ PCDD/Fs and 90%  $\Sigma_{26}$ PCBs were found in the soil of informal e-waste recycling sites.
- Metal recovery process using acid bath resulted in maximum TEQ for PCDD/Fs in New Delhi.
- Nearly three fourth of dioxin like PCBs were observed in the e-waste metal recovery sites.
- Combustion of PVC materials can be reasoned for high PCB-126 in e-waste sites and dumpsites.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Growth of informal electronic waste (e-waste) recycling sector is an emerging problem for India. The presence of halogenated compounds in e-wastes may result in the formation of persistent organic pollutants like polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) during recycling processes. We therefore investigated PCBs and PCDD/Fs in surface soils explicitly from the informal e-waste recycling sites and nearby open dumpsites of major metropolitan cities from four corners of India, viz., New Delhi (North), Kolkata (East), Mumbai (West) and Chennai (South). In the informal e-waste recycling sites, the range of  $\Sigma_{26}$ PCBs (0.4–488 ng/g) and  $\Sigma$ PCDD/Fs (1.0–10.6 ng/g) were higher than  $\Sigma_{26}$ PCBs (0.3–21 ng/g) and  $\Sigma$ PCDD/Fs (0.15–7.3 ng/g) from open dumpsites. In the e-waste sites,  $\Sigma$ PCDDs were found with increasing trend from  $\Sigma$ TetraCDD to OctaCDD, whereas  $\Sigma$ PCDFs showed a reverse trend. The dominance of PCDF congeners and maximum toxicity equivalents (TEQ) for both PCDDs (17 pg TEQ/g) and PCDFs (82 pg TEQ/g) at Mandoli in New Delhi has been related to intensive precious metal recovery process using acid bath. Among dumpsites, highest TEQ for PCDD/Fs was observed at Kodangaiyur dumpsite of Chennai (CN<sub>DS-02</sub>, 45 pg TEQ/g). Positive Matrix Factorization (PMF) model identified distinct congener pattern based on the functional activities, such as e-waste dismantling, shredding, precious metal recovery and open burning in dumpsites. E-waste metal recovery factor was loaded with 86–91% of PCB-77, -105, -114, -118 and 30% of PCB-126, possibly associated with the burning of wires during the copper extraction process. Almost 70% of the  $\Sigma_{26}$ PCB concentrations was comprised of the dioxin-like PCB congeners with a maximum concentration of 437 ng/g at New Moore market in Chennai, followed

\* Corresponding author at: SRM Research Institute, SRM University, Kattankulathur, Tamil Nadu 603203, India.  
E-mail address: [paromita.c@res.srmuniv.ac.in](mailto:paromita.c@res.srmuniv.ac.in) (P. Chakraborty).

by Wire Lane (102 ng/g), in Mumbai. We speculate that PCB-126 might have resulted from combustion of plastic materials in e-waste stream and dumped waste.

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## 1. Introduction

Electronic and electrical waste (e-waste) recycling sector is considered to be a profitable business but is of serious concern for the environmental and health impacts in developing countries, such as China, Ghana, Brazil and India where environmental laws are still not so stringent (Garlapati, 2016). India generates nearly 350,000 tons of e-waste every year due to large and fast consumption rates of electrical and electronic (EE) products and imports about 50,000 tons of used information technology (IT) devices every year from overseas (Brigden et al., 2008). Recent studies (Liu et al., 2008; Ma et al., 2008; Shen et al., 2009; Wang et al., 2011; Hu et al., 2013) have evidenced that e-waste contains a large range of toxic compounds, including polychlorinated biphenyls (PCBs), brominated flame retardants and toxic metals. Such toxicants and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) can be released during the e-waste recycling processes. They have been also found in workplace environments, and in the surrounding soil and watercourses (Chan et al., 2007).

In India, the majority of the domestic and/or imported e-waste reach the informal recyclers through the scrap dealers for extraction of precious metals and poor people are engaged in the process at a minimum wage (Toxics Link, 2004; GOI, 2011). Leftovers from such recyclers end up in the dumping grounds or landfills. In India, 95% of the e-waste is treated and processed by informal e-waste recycling sector, which consists of unregulated and unregistered, low-technology units where both adults (male and female) and children carry out dangerous procedures without any safety tools (Toxics Link, 2007).

In 2016, the Ministry of Environment, Forest and Climate Change (MoEFCC) released the updated E-waste (Management) Rules, which came in supersession of the E-waste in India (GOI, 2016). Now producers can have Producer Responsibility Organisation (PRO) in which they ensure the collection and disposal of e-waste in a sustainable manner. New rules, released in 2016, are more powerful than older one due to awareness about e-waste among government and masses.

Limited studies have been conducted in India to evaluate the environmental and health impact of recycling electronic waste by the informal sector. Chakraborty et al. (2013) ascribed atmospheric emission of PCBs from source regions like informal e-waste recycling and open burning in the municipal dumpsites. Such source types were prime factors for the presence of high PCB concentrations in surface soil and evidenced re-emission from the hotspots (Chakraborty et al., 2016a). Recently, Chakraborty et al. (2016b) determined PCDD/Fs and dioxin-like PCBs in surface soil from informal e-waste recycling sites by using chemically activated luciferase expression (CALUX), which is a very suitable screening method to generate bioassay toxic equivalents for dioxins and dioxins like compounds. Very high PCB levels have been observed in surface dust collected from the informal e-waste recycling workshops engaged in metal recovery process compared with the industrial sites of Chennai city (Chakraborty et al., 2016c).

With the exponential growth of both domestically generated and imported e-waste, informal e-waste recycling in developing countries is an important environmental and health issue. Therefore, in this study, we investigated PCBs and PCDD/Fs in surface soil samples from informal e-waste recycling sites and nearby open dumpsites of the major metropolitan cities from four corners of India, viz., New Delhi, Kolkata, Mumbai and Chennai, generating >50% of the domestic e-waste in India (Chatterjee, 2011). Congener specific PCB concentrations obtained from these four cities were subjected to source-receptor positive matrix factorization (PMF) model to individuate the distribution pattern of PCB congeners associated with the e-waste recycling processes in the informal sectors. Taking into consideration that soils act both as contamination and sink source and show fundamental role in contaminating and

transferring of contaminants to water bodies, plants and organisms, toxic equivalency (TEQ) values were also estimated and discussed.

## 2. Materials and methods

### 2.1. Sampling sites

Surface soil samples (0–20 cm depth) were collected during May 2014, from informal e-waste recycling sites and nearby open municipal dumpsites of four major Indian cities viz., New Delhi, Kolkata, Mumbai and Chennai located in northern, eastern, western and southern India, respectively. From Kolkata, only dumpsite samples were collected since electronic scrap recycling takes place mostly in the suburban sites of Howrah district and it was not possible to get access to e-waste recycling workshops. Sampling sites are indicated in Fig. 1, while average air temperature, latitude, longitude and percentage of total organic carbon (TOC%) are reported in supplementary information (Table S1). Based on a previous survey on the stages involved in informal e-waste processing (Chakraborty et al., 2016d), e-waste sampling sites were classified as: e-waste dismantling (EWD), e-waste shredding or grinding (EWS) and e-waste metal recovery (EWR). Dumpsite samples (DS) were collected from nearby open dumping grounds. Control sites (CS) were selected nearly 20–30 km away from the e-waste areas, in natural reserves characterized by minimum human activity. At each location, duplicate composite samples were collected each consisting of five sub-samples collected from an area of about  $10 \times 10 \text{ m}^2$ . Samples were homogenized to obtain about 1000 g of a representative sample, twigs and plastic materials were removed, then soil samples were grounded with a mortar and pestle, sieved through a 1 mm mesh/sieve and stored at  $-20^\circ\text{C}$  until extraction.

### 2.2. Soil sample extraction and clean-up

Soil samples were extracted by MARS 6 One Touch Microwave Accelerated Reaction System (CEM Corporation, Matthews, USA), using the USEPA 3546 method (USEPA, 2007). Before analysis, the extraction cylinders (Teflon vessels fitted with PTFE-lined caps) were rinsed with *n*-hexane and acetone. 20 g of soil sample was placed into the extraction vessels and extracted with a mixture of 20 mL of acetone/*n*-hexane (1:1 v/v). More details on microwave operating conditions are given in Supporting Information. Prior to extraction, all samples and blanks were spiked with surrogate standards,  $^{13}\text{C}_{12}$ -labeled coplanar PCBs, PCDD/Fs and PCB-209 to monitor the analytical recovery efficiencies.

The extracted volume was reduced to 1 mL using a rotary evaporator. Then, samples were cleaned-up and fractionated on a multilayered silica gel column containing from bottom: 0.9 g of anhydrous sodium sulfate, 3 g of silica gel impregnated by potassium hydroxide, 0.9 g of silica, 4.5 g of silica gel impregnated with 44% sulfuric acid, 6 g of silica gel impregnated with 22% sulfuric acid, 0.9 g of silica, 3 g of silica gel impregnated with 10% silver nitrate, 0.9 g of silica and, finally, 6 g of sodium sulfate on the top. The sample extracts were loaded in the above-mentioned multilayer column and subsequently eluted with 150 mL of *n*-hexane, followed by an active carbon column to separate dl-PCBs fraction using 40 mL of DCM/*n*-hexane (3:1 v/v) mixture. The dioxin/furan fraction was eluted afterward with 60 mL of toluene by the reverse carbon column for complete separation of PCDD/Fs absorbed in the top layer. The concentrated extract was kept under a gentle stream of pure Nitrogen using Turbo Vap (Caliper, USA) and reduced to 1.0 mL. Similarly extracted duplicate soil sample from each site was cleaned-up using the procedure given elsewhere (Chakraborty et al., 2016a) to analyse both dioxin like and non-dioxin like PCB congeners by GC-MS.

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