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# High levels of antimony in dust from e-waste recycling in southeastern China

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## ABSTRACT

Environmental contamination due to uncontrolled e-waste recycling is an emerging global issue. Antimony (Sb) is a toxic element used in semiconductor components and flame retardants for circuit board within electronic equipment. When e-waste is recycled, Sb is released and contaminates the surrounding environment; however, few studies have characterized the extent of this problem. In this study, we investigated Sb and arsenic (As) distributions in indoor dust from 13 e-waste recycling villages in Guiyu, Guangdong Province, southeastern China. Results revealed significantly elevated concentrations of Sb (6.1–232 mg/kg) in dust within all villages, which were 3.9–147 times higher than those from the non e-waste sites, indicating e-waste recycling was an important source of Sb pollution. On the contrary, As concentrations (5.4–17.7 mg/kg) in e-waste dusts were similar to reference values from the control sites. Therefore, dusts emitted from e-waste recycling may be characterized by high Sb/As ratios, which may help identify the contamination due to the e-waste recycling activities.

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# 1. Introduction

Environmental contamination by e-waste recycling is an emerging global issue (Robinson, 2009). In 2006, the world's production of ewaste was estimated at 20-50 million tonnes per year (UNEP, 2006) and it is growing at around 4% per year (Focus, 2005). It is estimated that 50-80% of the e-waste from US, Europe, and other areas of the world is legally or illegally imported to Asia each year, and 90% of which is destined for China (Chen et al., 2009). E-waste contains large quantities of hazardous chemicals, which may be released during uncontrolled e-waste recycling processes, threatening the ecosystem and the health of local residents. In previous reports, all environmental compartments, including air, water, soil and organisms, surrounding ewaste processing sites, were severely contaminated by toxic chemicals, such as cadmium (Cd), copper (Cu), lead (Pb), zinc (Zn), polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins (PCDDs) and furans (PCDFs), and polybrominated diphenyls ethers (PBDEs) (Li et al., 2007; Wong et al., 2007; Leung et al., 2008; Chen et al., 2009; Robinson, 2009; Luo et al., 2011). Many studies reported elevated levels of heavy metals and persistent organic pollutants in e-waste workers and in children residing nearby (Bi et al., 2007; Huo et al., 2007).

Antimony (Sb) is an emerging toxic contaminant with unknown biological function. Sb and its compounds are classified as priority

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pollutants by the European Union and the Environmental Protection Agency of the United States (EU, 1976; US-EPA, 1979) and are found on the list of banned hazardous compounds specified in the Basel Convention (2005). Antimony is used in semiconductor components and flame retardants within electronic equipments. So, Sb may be an important pollutant released from e-waste recycling. But to the best of our knowledge, only a few studies reported Sb contamination due to e-waste recycling activities (e.g., Ngoc Ha et al., 2009). In this study, we collected dust samples from e-waste workshops for the purpose of determining Sb contamination status in the dust emitted during e-waste recycling. Since arsenic (As) has similar geochemical behavior to that of Sb (Wilson et al., 2010), characterization of As was also included.

## 2. Materials and methods

#### 2.1. Site description

Guiyu town in Shantou, Guangdong province, China, is 52 km<sup>2</sup> and the local population is 150,000 (Li et al., 2011). To make a living, farmers in the area initiated e-waste recycling in the mid 1990s. At present there are 500 recycling workshops registered with the local Industry and Commerce Administration. It is estimated that the annual recycling capacity of the town is 100,000 tonnes (Li et al., 2011). Approximately 60– 80% of families in the town have engaged in e-waste recycling operations, which were conducted by small scale family-run workshops, with approximately 100,000 migrant workers employed in processing e-waste (Huo et al., 2007). Two kinds of family-run workshops were

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included in this study. One is engaged with circuit boards from computers and other large appliances, which are dismantled with a hammer and melted over honeycombed coal blocks, releasing valuable electronic components, such as diodes, resistors, and microchips. The other is plastic processing, including plastic scraps sorting and grinding (Huo et al., 2007).

## 2.2. Sampling procedure

In 2010, 29 indoor dust samples were collected from 13 villages in Guiyu (Table 1). In addition, five dusts were sampled from a control area, and were likely free from the e-waste contamination. Dust samples were collected inside from the floor using a brush and plastic spatula, stored in polyethylene bags, then transported to the laboratory. All samples were air-dried at room temperature, and passed through a 2-mm sieve to remove rocks, plants, hair and other impurities. The homogenized dust samples were ground to a fine powder texture with an agate mortar prior to chemical analyses.

# 2.3. Sample analysis

About 0.2 g of prepared dust sample were digested with 10 ml of aqua regia (3:1, HCl:HNO<sub>3</sub>). Antimony and As concentrations of the digest solutions were determined by a hydride generation-atomic fluorescence spectrometer system (HG-AFS). QA/QC included reagent blanks, analytical duplicates, and analysis of the standard reference material (SRM) (GBW-07406). The recovery rates for Sb and As in the SRM were between 75 and 105%.

# 3. Results and discussion

The background Sb value in soil from Guangdong province is 0.54 mg/kg (CNEMC, 1990). Dust samples collected from non e-waste sites contained 0.66–2.45 mg/kg of Sb, which were higher than the background value. Antimony concentrations in the indoor dust from

#### Table 1

Dust sa	mples	description	and	the Sb	and	As	concentra	tions
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Sampling village	Sample no.	Type of the workshop	Sb (mg/kg)	As (mg/kg)		
Beilin	1	Circuit board baking	232	5.4		
	2	Circuit board baking	209	10.4		
Xindun	3	Plastic processing	31.6	10.3		
	4	Circuit board baking	12.2	8.9		
	5	Circuit board baking	88.6	10.7		
	6	Circuit board baking	18.9	13.3		
Nanan	7	Circuit board baking	40.6	17.7		
	8	Circuit board baking	81.9	16.3		
Simei	9	None <sup>a</sup>	9.2	12.8		
	10	Circuit board baking	16.7	14.6		
Huamei	11	Circuit board baking	57.0	13.3		
	12	Circuit board baking	41.5	14.3		
	13	Plastic processing	15.8	13.3		
Xianma	14	Plastic processing	11.2	9.0		
	15	Plastic processing	68.6	14.7		
Hougang	16	Plastic processing	45.6	13.5		
	17	Plastic processing	48.4	14.2		
Xianpeng	18	Plastic processing	30.0	12.2		
	19	Plastic processing	12.7	16.3		
Longgang	20	None	6.5	11.6		
	21	Plastic processing	16.7	14.3		
Nanyang	22	None	8.4	10.8		
	23	None	6.1	10.4		
Houwang	24	Plastic processing	24.8	11.6		
	25	None	8.1	10.9		
Yujiao	26	Plastic processing	36.2	8.7		
	27	Circuit board baking	27.7	13.1		
Shangpeng	28	Plastic processing	32.1	9.2		
-	29	Plastic processing	35.1	13.7		
Reference samples from control sites $(N=5)$ 0.66–2.45 9.2–11.6						

<sup>a</sup> Means the house has not been used as a workshop for e-waste recycling.

the e-waste recycling sites were highly elevated, ranging from 6.1 to 232 mg/kg with a mean value of 44 mg/kg (Table 1), which were 3.9-147 times higher than the levels in the control sites, and 11-430 times higher than the local soil background value. The highest concentration was found in the village of Beilin where the e-waste recycling activities were most concentrated. Leung et al. (2008) also reported extremely high concentrations of other metals (Pb 110,000 mg/kg, Cu 8360 mg/kg, Zn 4420 mg/kg, and Ni 1500 mg/kg) in dust from the same village. Relatively low Sb concentrations (6.1-9.2 mg/kg) were found in several homes (Table 1) where e-waste workshops were not located, and the Sb likely originated from the outside. The mean dust Sb concentration (75 mg/kg) due to circuit board baking was significantly higher (P=0.003) than that of the plastic processing (31 mg/ kg), suggesting that the former activities would release more Sb. Ngoc Ha et al. (2009) reported similar high concentrations of Sb (2.9-180 mg/kg) in soils collected from the e-waste recycling sites in Bangalore, India, reflecting that Sb contamination due to e-waste recycling might be a common phenomenon. The authors also reported higher levels of Sb in hair of e-waste workers compared with a reference population (Ngoc Ha et al., 2009), indicating elevated Sb levels will increase the human body burden for residents in the area. Therefore, adverse health effects posed by e-waste Sb require more attention (Ngoc Ha et al., 2009).

Arsenic has similar chemical and physical properties and geochemical behaviors to that of Sb, and hence usually coexists with Sb in many environments (Wilson et al., 2010). However, in contrast to Sb levels, As concentrations in dust from the e-waste sites ranged from 5.4 to 17.7 mg/kg, which were not higher than the reference As values (9.2–11.6 mg/kg) from the control sites, suggesting that ewaste recycling was not a pollution source of As. In this region, dust emitted from e-waste recycling had high ratios of Sb/As (0.6–43 in this study), which might differ from those of other pollution sources. Data regarding the Sb and As concentrations in natural and anthropogenic matrices were compared (Fig. 1). Traffic emission, coal



**Fig. 1.** Sb and As concentrations in different natural and anthropogenic matrices. Data sources: Dust from e-waste site and control site (this study), soil from Sb mining and smelting site (Fu et al., 2011), Dust from Zn/Pb smelting site (this study, unpublished data), Dust from Zn smelting site (this study, unpublished data), Coal fly ash (Song et al., 2003; Wang et al., 2003; Leng et al., 2005), Urban road dust (Han et al., 2008), and local background soil (CNEMC, 1990).

combustion, Zn–Pb smelting, and Sb mining and smelting are the most important Sb pollution sources. When compared with these anthropogenic sources, it's clear that samples from e-waste recycling sites have distinctly high Sb/As ratios and hence could be recognizable amidst the numerous anthropogenic matrices (Fig. 1). Therefore, the high Sb/As ratios may help identify the e-waste contamination. However, for a better understanding of the e-waste contamination, other information should be integrated, such as Ag, Cu and Sn concentrations (Robinson, 2009), since some samples, especially those from Sb mining and smelting areas, may have high Sb/As ratios (Fu et al., 2011; Wu et al., 2011) (Fig. 1).

Based on our findings, it is likely that large quantities of Sb have been emitted into the air during e-waste recycling activities in the past dozen years. Since Sb has been recognized as a global pollutant which can undergo long-range atmospheric transport and contaminate remote areas (Cloy et al., 2005; Krachler et al., 2005; Shotyk et al., 2005), Sb emissions from the e-waste recycling not only pose a threat to the local environment, but also may contribute to the global cycle of this element in the atmosphere. The Arctic snow and ice data show that enrichments of Sb in Arctic air have increased 50% during the past three decades (Krachler et al., 2005). Krachler et al. (2005) argued that most of this Sb is produced in Asia, primarily from Sb sulfides, as well as a by-product of Pb and Cu smelting. The results of the present study suggest that e-waste recycling in Asia may be one of the reasons for this increasing Sb enrichment; however, the extent of the contribution needs to be further investigated.

#### 4. Conclusion

In the present study, we reported for the first time the Sb and As concentrations in dust from an e-waste recycling area of Guiyu, southeastern China. Analytical results showed significantly elevated Sb levels in the dust, while dust As concentrations remained unchanged compared with the reference values. According to these data, high Sb/As ratios may help identify the contamination due to the e-waste recycling activities. Our findings demonstrated that e-waste recycling activity is one of the major sources of Sb pollution. The adverse effects of the e-waste Sb on the ecosystem and human health require attention and should be further studied.

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