



## Distribution of heavy metal pollution in sediments from an acid leaching site of e-waste



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### HIGHLIGHTS

- Distribution of heavy metal in sediments from an acid leaching site was studied.
- In addition to common pollutants, these sediments were polluted with Sn and Sb.
- Sn and Sb may exert environmental risks because of their high total concentrations.
- Pollution level of sediments decreased in the order of Sb > Sn > Cu > Cd > Pb > Zn > Ni.
- The dominant source of heavy metal in sediments was e-waste recycling activities.

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### ABSTRACT

The spatial distribution, bioavailability, potential risks and emission sources of 12 heavy metals in sediments from an acid leaching site of e-waste were investigated. The results showed that the sediments from the acid leaching site were significantly contaminated with Cu, Zn, Cd, Sn, Sb and Pb, especially in the middle sediments (30–50 cm), with average concentrations of 4820, 1260, 10.7, 2660, 5690 and 2570 mg/kg, respectively. Cu, Cd and Pb were mainly present in the non-residual fractions, suggesting that the sediments from the acid leaching site may exert considerable risks. Mn, Ni, Zn, Sn and Sb were predominantly associated with the residual fraction. Despite their low reactivity and bioavailability, uncommon pollutants, such as Sn and Sb, may exert environmental risks due to their extremely elevated total concentrations. All of these results indicate that there is an urgent need to control the sources of heavy metal emission and to remediate contaminated sediments.

**Capture abstract:** In addition to Ni, Cu, Zn, Cd and Pb, the sediments from an acid leaching site in Guiyu were heavily polluted with uncommon heavy metal pollutants, such as Sn and Sb.

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### 1. Introduction

Electronic waste (e-waste) comes from discarded electrical and electronic equipment, including computers, televisions, air conditioners, washing machines and their components, such as printed circuit boards (Tang et al., 2010; Wong et al., 2007a). A large number of electronic products are abandoned due to their short lifespan and the expansion of the global market. Approximately 20–50 million tons of e-waste is generated annually in the world, one third of this waste is from the US and one quarter of this waste is from the EU (UNEP, 2005). Furthermore, this global rate of e-waste production is increasing by 4% per year (Peng et al., 2009). In China, 1.1 million tons of e-waste was generated in 2003 and the production of e-waste has been

increasing at the rate of 5%–10% per year (Wang, 2008). E-waste is a complex mixture of metals, plastics, glasses and ceramics (M.H. Wong et al., 2007). If disposed of improperly, e-waste can become a source of metal and organic contaminants, and may impose great threats to local ecosystems and human health (Wong et al., 2007b). However, driven by profits, unregulated recycling methods/technologies, such as melting plastic to reduce waste volume, open-burning of wires and cables to recycle Cu, and acid leaching of waste printed circuit boards to recover precious metals (e.g. Au, Ag, Pt and Pd), are usually used (Fu et al., 2008; Wong et al., 2007b; Zheng et al., 2008). These primitive methods for processing e-waste have led to the release of numerous toxic metals (e.g., Cu, Zn, Cd and Pb) and persistent organic pollutants (e.g., polycyclic aromatic hydrocarbons and polybrominated diphenyl ethers) into local aquatic and terrestrial ecosystems (Fink et al., 2000; Wang et al., 2011).

Heavy metal contamination in sediments is a significant environmental problem because of the toxicity, non-degradation and easy

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bio-accumulation of heavy metals (Bozkurt et al., 2000). Common heavy metal pollutants in the sediments include Cr, Ni, Cu, Zn, Cd and Pb (Monikh et al., 2013; Nithya et al., 2011). Heavy metals are easily accumulated on fine-grained particles of sediments (Mendiguchía et al., 2006; Zhang et al., 2012). These metals that are fixed to sediments may be returned to water bodies via chemical and biological processes and subsequently transported to downstream rivers (Liu et al., 2009). Thus, sediment is the most important sink for heavy metals and may act as a carrier and source of heavy metals in estuary systems (Luo et al., 2008; Yang et al., 2012). The acid leaching sites are always located in fields adjacent to rivers. The acid waste and wastewater generated during acid leaching processes are discharged into nearby streams, while solid wastes are deposited on-site with no or very few pollution control measures. Therefore, a comprehensive evaluation of heavy metal contamination in sediments from acid leaching sites is important for the environmental management and pollution control of rivers.

Several studies have reported some common heavy metal pollutants from surface soil/sediments (<40 cm) in e-waste recycling sites. Previous soil/sediment pollution investigation of heavy metals in e-waste processing areas had included open-burning sites (Luo et al., 2008), pond areas (Luo et al., 2011), paddy fields (Zhang and Min, 2009), deserted areas, etc. (Luo et al., 2011). It has been reported that the top-soil in the e-waste incineration sites of Longtang were significantly polluted with Cu (11,100 mg/kg), Zn (3690 mg/kg), Cd (17.1 mg/kg) and Pb (4500 mg/kg; Luo et al., 2011). Furthermore, the paddy soils in the Luqiao district, which is the city center of Taizhou, were heavily polluted with Cd (6.4 mg/kg), and weakly polluted with Cu (256 mg/kg) and Zn (210 mg/kg) (Zhang and Min, 2009). However, available information on the status of heavy metal contaminations in deep sediments from e-waste processing sites, especially in acid leaching sites, is limited. In addition, uncommon potential heavy metal contaminants, such as Be, Sn and Sb in sediments from e-waste recycling areas have also rarely been reported. It has been reported that Be is widely used in printed circuit boards and may be a human carcinogen (Peng et al., 2009). Sn may be released from solders within electronic equipment (Li et al., 2011), and Sb is used in semiconductor components and flame retardants within electronic devices (Bi et al., 2011). It should be noted that SnCl<sub>2</sub> may induce tumor generation in the thyroid gland (Ferancová et al., 2007). Sb and its compounds are listed as priority pollutants by the Environmental Protection Agency of the United States (US-EPA, 1979). Therefore, these uncommon pollutants may be the important pollutants released from uncontrolled e-waste processing activities.

The aims of this study are (1) to further investigate the distribution of 12 heavy metals (Be, V, Cr, Mn, Co, Ni, Cu, Zn, Cd, Sn, Sb and Pb) in sediments from an acid leaching site related with the disposal of e-waste; (2) to obtain more available information on the bioavailability of heavy metals in these sediments; (3) to evaluate the potential risks of heavy metals in these sediments to human health and their surrounding environment; and (4) to explore the relationships among different heavy metals and to identify emission sources of heavy metals in these sediments. It is expected that our findings will provide scientific evidence for preventing heavy metal pollution in sediments from e-waste processing sites.

## 2. Materials and methods

### 2.1. Reagents

All of the chemicals used in our experiments were of analytical reagent grade or better and were also examined for possible heavy metal pollution. Milli-Q water with a resistance of 18.25 MΩ · cm at 25 °C was prepared using a water purification system. The 500 mg/L Sn (GSB G 62042-90, China) and 500 mg/L Sb (GSB G 62043-90, China) stock solution were obtained from the China Iron and Steel Research Institute. A multi-element standard stock solution (cat# CLMS-2AN, USA) containing 10 mg/L Be, V, Cr, Mn, Co, Ni, Cu, Zn, Cd and Pb

was purchased from SPEX CertiPrep. All plastic tubes and bottles were strictly cleaned, soaked in a 10% (v/v) diluted nitric acid solution for at least 24 h and rinsed 3 times with Milli-Q water prior to the experiments.

### 2.2. Study area and sample collection

Guiyu (116.40°E, 23.33°N) is the largest e-waste recycling center in China. The average annual rainfall is 1386 mm. E-waste processing activities have been ongoing in Guiyu since 1995. There are 3207 dismantling and recycling workshops (including companies and individual households) in this town according to the estimations from the Statistical Bureau of the Chaoyang district. Moreover, most e-waste dismantling processes are conducted in family-run workshops where primitive and traditional recycling methods (e.g., acid leaching and open-burning of e-waste) are always used. The acid waste, wastewater and solid wastes containing multiple heavy metals and organic compounds might severely contaminate local rivers.

Sediment sampling was performed in May of 2013. The locations of the sampling sites are shown in Fig. 1. A total of 15 sediment cores were collected using a professional heavy metal sampler equipped with a polyvinyl chloride (PVC) liner. To comprehensively assess heavy metal pollution of sediments at various depths, each sediment core was divided into three parts: surface sediment (0–20 cm, SS), middle sediment (30–50 cm, MS) and deep sediment (60–80 cm, DS). All 45 subsamples from the 15 sediment cores were placed in polyethylene bags (Ziploc) and stored at 4 °C prior to pretreatment.

### 2.3. Pretreatment and analysis

All sediment samples were dried for 48 h in an 80 °C oven, finely powdered using an agate mortar and sieved to pass through a 100-mesh nylon sieve, thoroughly mixed and placed into glass bottles. The pH (water/solid = 2.5:1) of the sediments was measured using a Leichi pH meter (pHS-3C, China). The C, H, N and S contents of all sediments were analyzed using an elemental analyzer (Vario EL-III, Germany). After removing inorganic carbon with 5% (v/v) HCl, the total organic carbon (TOC) in the sediments was measured using a CM250 TOC analyzer (USA).

Approximately 0.2 g of each sample was digested with a mixture of concentrated acids (HF/HNO<sub>3</sub>/HCl = 5:5:2) (Li et al., 2011) using a hot plate (ML-2-4, Beijing, China) at 200 °C for the determination of 12 heavy metals. The digestion liquids were filtered through 0.45 μm micro-porous membranes and diluted to 10 mL. The total concentrations of heavy metals were determined using an Agilent 7700X inductively coupled plasma-mass spectrometer (ICP-MS). The instrument was calibrated with 0.05 mg/L internal standards (<sup>4</sup>Li, <sup>42</sup>Sc, <sup>72</sup>Ge, <sup>103</sup>Rh, <sup>115</sup>In and <sup>219</sup>Bi). The detection limits for Be, V, Cr, Mn, Co, Ni, Cu, Zn, Cd, Sn, Sb and Pb were 0.001, 0.006, 0.078, 0.041, 0.012, 0.008, 0.081, 0.112, 0.010, 0.005, 0.011 and 0.042 μg/L, respectively. In addition to reagent blanks and replicates, a Standard Reference Materials Estuarine Sediment (SRM 1646a) was used for evaluating the reliability of the analytical methods. The percentages of recoveries of the 12 heavy metals from SRM 1646a were reasonably good (84.2%–120.5%).

Chemical speciation of heavy metals in an average sediment sample, which was prepared by mixing an equal amount of each of the 45 subsamples, was conducted based on the modified BCR three-step sequential extraction procedure (Baig et al., 2009; Pueyo et al., 2008). The extraction procedure used in this study classified heavy metals into four chemical fractions: acid-soluble fraction, reducible fraction, oxidizable fraction and residual fraction. The residual fraction was digested using the same procedure used for analyzing the total heavy metal concentration in the sediment described above. Triplicates and procedure blanks were conducted for quality assurance and quality control purposes. The sum of four fractions of Be, V, Cr, Mn, Co, Ni, Cu, Zn, Cd, Sn,

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