



Potential health risk for residents around a typical e-waste recycling zone via inhalation of size-fractionated particle-bound heavy metals



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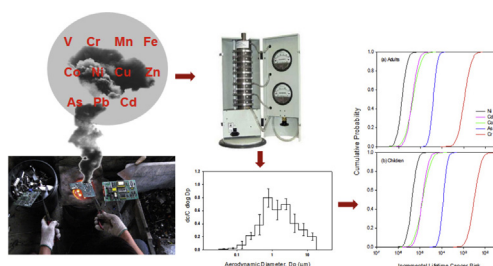
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HIGHLIGHTS

- Anthropogenic metals were predominantly enriched in fine particles.
- Crustal elements tended to accumulate in coarse particles.
- Health risk for residents through inhalation of particle-bound heavy metals was high.

GRAPHICAL ABSTRACT



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ABSTRACT

Health risk of residents dwelling around e-waste recycling zones has been a global concern, but has not been adequately examined. The present study was intended to evaluate the potential health risk of residents through inhalation exposure to size-fractionated particle-bound heavy metals in a typical e-waste recycling zone, South China. Anthropogenic metals (Zn, Se, Pb, Sb, As, and Cd) were predominantly enriched in fine particles ($D_p < 1.8 \mu\text{m}$), whereas the crustal elements (Ti, Fe, and Co) tended to accumulate in coarse particles ($D_p > 1.8 \mu\text{m}$). Although the daily inhalation intakes of the target metals were significantly lower than those through food consumption and ingestion of house dust, the hazard quotients of total metals for adults (95% CI: 1.0–5.5) and children (95% CI: 3.0–17) were greater than 1. Moreover, the incremental lifetime cancer risks of five carcinogenic metals (Cr, Co, Ni, As, and Cd) for adults and children were 1.3×10^{-3} (95% CI: 4.1×10^{-4} – 3.0×10^{-3}) and 3.9×10^{-3} (95% CI: 1.3×10^{-3} – 8.6×10^{-3}), respectively, substantially higher than the acceptable cancer risk range of 10^{-6} – 10^{-4} . All these findings suggested that health risks were high for local residents dwelling around the e-waste recycling zone through inhalation exposure to particle-bound heavy metals, for both adults and children.

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1. Introduction

E-waste recycling has been a global issue as the lifespan of electrical and electronic products has become increasingly shorter in recent decades [1,2]. Due to the pursuit of economic growth and thirsty for raw materials, developing countries, such as India,

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Pakistan, and China, have intentionally or unintentionally been encouraging e-waste dismantling and recycling activities. For example, the amount of copper from e-waste recycling in Qingyuan, one of the significant e-waste recycling zones, accounted for approximately 25% of total reclaimed copper production in China in 2013 [3]. On the other hand, largely primitive recycling techniques employed may have accelerated the release of toxic pollutants into the surrounding environments, subjecting local workers and residents to health hazards [4–9]. For instance, Xu et al. [10] found that preschool children in an e-waste recycling area in Guiyu of China had higher Pb blood levels and lower hepatitis B surface antibody titers than those in a reference region.

Dietary intake, dermal contact and inhalation are three main pathways for human exposure to toxic pollutants. Dietary intake can be substantially reduced by mainly consuming foods not produced locally. For example, samples of rice produced within an e-waste recycling region (Taizhou, China) contained significantly higher concentrations of heavy metals than those from reference areas, and thereby local residents mainly purchased food items transported from other places to reduce the exposure risk [4,11]. In addition, wearing clean clothes can considerably decrease the uptake of pollutants through dermal contact. By comparison, inhalation exposure is perhaps the hardest to alleviate. Our previous study demonstrated that the inhalation intake rate of particle-bound polybrominated diphenyl ethers in the e-waste recycling zone was similar to those through food consumption in the urban regions [12]. Although the health risk for residents was low from inhalation exposure to particle-bound brominated and organophosphate flame retardants [12,13], the amounts of heavy metals released from e-waste recycling activities were 1–2 orders of magnitude greater than those of organic pollutants [1], thereby human health effects induced from heavy metals must be further evaluated.

Most research efforts appear to have focused on health risk assessment based on total suspended particles, which may overestimate health risk from inhalation exposure [12,14,15]. Therefore, the size dependence of inhalation exposure should be considered. In the present study, size-segregated suspended particle samples were collected and analyzed for heavy metals. A typical e-waste recycling zone, i.e., Qingyuan, in Guangdong Province of South China was selected as the sampling site. The size distributions of heavy metals were characterized at three different heights (1.5, 5, and 20 m), and used to estimate the deposition fluxes in the human respiratory system with a model developed by the International Commission on Radiological Protection (ICRP). In addition, the non-carcinogenic effect and incremental lifetime cancer risk to local residents (both adults and children specific) through inhalation exposure were evaluated and compared with those from dietary intake.

2. Methods and Materials

2.1. Sample collection

Field sampling was conducted on November 11–15, 2012 with a Micro-Orifice Uniform Deposit Impactor (MOUDI) on the first (1.5 m) and sixth (20 m) floors of a residential building and the second floor (5 m) of another building within an e-waste recycling zone in Qingyuan. The e-waste dismantling workshops were dispersed around the sampling site. A total of 165 samples were collected on 47-mm diameter polytetrafluoroethylene (PTFE) membrane filters (Pall Corporation, Ann Arbor, MI, USA) drawn at a flow rate of 30 L min⁻¹ and fractioned into 11 sizes, i.e., >18, 10–18, 5.6–10, 3.2–5.6, 1.8–3.2, 1.0–1.8, 0.56–1.0, 0.32–0.56, 0.18–0.32, 0.10–0.18, and 0.056–0.10 μm. Daily sampling time was 12 h

from 07:30 to 19:30. Size-segregated particles were classified as coarse ($D_p > 1.8 \mu\text{m}$), accumulation mode ($0.1 \mu\text{m} < D_p < 1.8 \mu\text{m}$), and ultrafine ($D_p < 0.10 \mu\text{m}$) particles, and the sum of accumulation mode and ultrafine particles is defined as fine particles.

2.2. Sample extraction

The loaded filters were cut into pieces and placed in PTFE digestion vessels, and digested with an acid mixture of 5-mL nitric acid (Optima A467-500, Fisher Scientific, Hampton, NH, USA) and 1-mL hydrofluoric acid (Optima A463-500, Fisher Scientific, Hampton, NH, USA) in an XT-9912 microwave digestion instrument (XTrust Instruments, Shanghai, China). After cooling down, the digestion vessels were heated at 120 °C in a sample heating device (XT-9816-II, XTrust Instruments, Shanghai, China) until the digestion solutions were evaporated down to a few microliters. Finally, the digestion solutions were transferred to 10-mL centrifuge tubes and diluted with 4% nitric acid to 7 mL before being analyzed with an Agilent inductively coupled plasma mass spectrometer (Santa Clara, CA, USA).

2.3. Quality assurance and quality control

Procedural blanks ($n=23$) and standard reference materials ($n=2$; No. SRM2783; National Institute of Standard and Technology, Gaithersburg, MD, USA) were digested in parallel with field samples. The average concentration of 15 heavy metals in blank samples was 34 ng m⁻³, and Fe, Zn, and Ti collectively accounted for 93% of the total concentration. The average recoveries of V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Pb, and Ti from the standard reference materials were 101%, 85%, 89%, 110%, 91%, 98%, 93%, 107%, 106%, 92%, and 97%, respectively. The measured concentrations of heavy metals in field samples were corrected by procedure blanks analyzed in the same batch, but not corrected by the recoveries of target metals from the standard reference materials. The lowest calibration concentration of a target metal divided by the actual sampling volume (21.6 m³) was defined as the reporting limit, which was calculated as 0.16 ng m⁻³ for target metals. In addition, if a concentration was below the reporting limit, half of the reporting limit was used to estimate deposition flux and health risk.

2.4. Calculation of deposition fraction

The simplified equations from the ICRP [16] were used to calculate deposition fractions and fluxes of size-fractionated particles in the human respiratory tract. The fractions of inhaled particles deposited in three main parts of the human respiratory tract, i.e., head airways (HA, including nose, mouth, pharynx, and larynx), tracheobronchial region (TB), and alveolar region (AR) [12,17] were calculated. In particular, the deposition efficiency of a size-specific particle in the head airway ($DF_{HA,i}$), tracheobronchial region ($DF_{TB,i}$), and alveolar region ($DF_{AR,i}$) can be estimated by

$$DF_{HA,i} = IF_i \times \left(\frac{1}{1 + \exp(6.84 + 1.183 \ln D_{p,i})} + \frac{1}{1 + \exp(0.924 - 1.885 \ln D_{p,i})} \right) \quad (1)$$

$$DF_{TB,i} = \frac{0.00352}{D_{p,i}} \times (\exp(-0.234(\ln D_{p,i} + 3.40)^2) + 63.9 \exp(-0.819(\ln D_{p,i} - 1.61)^2)) \quad (2)$$

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