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Research Paper

Effect of lead speciation on its oral bioaccessibility in surface dust and soil of electronic-wastes recycling sites

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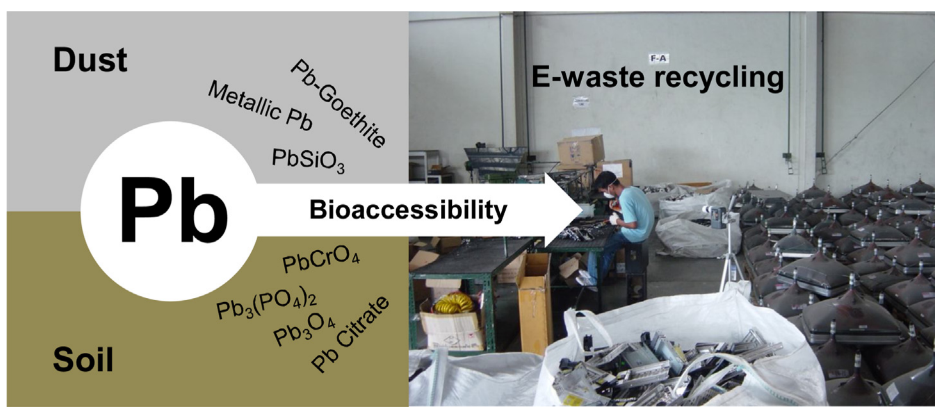
HIGHLIGHTS

- Bioaccessibility of Pb in e-waste surface matrices (dust and soil) was assessed.
- PBET test was used as *in vitro* bioaccessibility assay.
- Pb speciation was determined using X-ray absorption spectroscopy.
- Specific Pb bioaccessibility and species were identified.
- Influence of Pb species on bioaccessibility was discussed statistically.

GRAPHICAL ABSTRACT



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ABSTRACT

We measured bioaccessible lead (Pb) in simulated gastrointestinal fluids containing Pb-contaminated soil or dust from electronic waste (e-waste) recycling sites to assess the risk of Pb ingestion. The physiologically based extraction test (PBET) was used as *in vitro* bioaccessibility assay. Pb speciation was determined using X-ray absorption spectroscopy. The total Pb concentrations in dusts ($n = 8$) and soils ($n = 4$) were in the range of 1630–131,000 and 239–7800 mg/kg, respectively. Metallic Pb, a common component of e-waste, was ubiquitous in the samples. We also found Pb adsorbed onto goethite and as oxides and carbonate, implying soil mixing and weathering influences. Pb phosphate and organic species were only found in the soil samples, suggesting that formation was soil-specific. We identified other Pb compounds in

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several samples, including Pb silicate, Pb chromate, and Pb(II) hydrogen phosphate. A correlation analysis indicated that metallic Pb decreased bioaccessibility in the stomach, while a Pb speciation analysis revealed a low bioaccessibility for Pb phosphates and high bioaccessibility for organic Pb species. The health risk based on bioaccessible Pb was estimated to be much lower than that of total Pb due to the lower concentrations.

1. Introduction

Lead (Pb) pollution from electronic waste (e-waste) recycling poses serious risks to environmental and human health, as cathode ray tubes (CRT), circuit boards, and other electronics often contain Pb. Concentrations of Pb in surface soil [1–8] and dust [1,7–9] collected from e-waste recycling sites in developing countries exceed environmental standards and Pb has been reported in the blood of children [10], placenta [11], urine [12], scalp hair [13], and neonate umbilical cord blood and meconium [14] collected near e-waste recycling sites. Risk assessments based on hazard quotients (HQ) have indicated that Pb is the most hazardous element among metals and metalloids from e-waste dismantling and scrapping activities [8,9]. Recently, relationship between the Pb accumulation in humans and ingestion of surface matrices in e-waste recycling site was also reported [15,16]. Especially, soil was one of main contributing factors to the blood Pb level among groundwater, soil, rice, corn, chicken, and pork in the e-waste dismantling site [15]. However, no systemic studies have investigated the bioaccessibility of ingested Pb from e-waste. A survey on the bioaccessibility of Pb from indoor house environmental material provided the framework and techniques to study Pb from e-waste recycling.

Simple, rapid, and inexpensive *in vitro* bioaccessibility assays are useful for estimating the relative bioavailability of Pb from incidental dust and soil ingestion. By measuring Pb solubilized from a solid matrix into simulated human digestive fluids, the potential amount of Pb available for absorption into the systemic circulation can be estimated. A gastrointestinal extraction system typically comprises several compartments maintained at body temperature (37 °C) with controlled pHs, exposure times, and added biogenic compounds. The physiologically based extraction test (PBET) is a common *in vitro* bioaccessibility assay that has been used to assess the bioaccessibility of Pb in contaminated soil [17–20] and house dust [21]. It begins with a 1-h incubation in the stomach compartment, followed by a 4-h incubation in the small intestine compartment [17]. The PBET has been shown to be well correlated with the results of available feeding studies using soil Pb [17,22,23]. However, the *in vitro* PBET has not been applied to heavy metals in dust or soil matrices derived from e-waste recycling facilities.

The chemical forms of Pb that used in electronic production were quite different with those in soil samples, which was reported to influence Pb's bioaccessibility [24,25]. In 2011, the Canadian House Dust Study (CHDS) revealed a significant correlation between bioaccessibility derived from X-ray absorption spectroscopy (XAS) speciation and that measured using simulated gastric extraction, indicating the utility of analyzing Pb speciation with XAS to predict bioaccessibility in house dust [25]. Dust and soil from e-waste recycling sites likely contain different Pb species from those found in house dust [26], because Pb is used in electronics as specific species. For example, CRT glass contains Pb oxides and Pb silicates, while metallic Pb is common in circuit boards and other electronics. Additionally, Pb in dust and soil exposed to air could change chemical form due to e-waste processes, weathering effects, and biogeochemical reactions. For example, it was reported that weathering altered the chemical form and bioaccessibility of zinc in house dust [27].

Our research group investigated Pb levels in dust and soil from e-waste recycling sites around Metro Manila, the Philippines, in 2010 [8]. We found that total Pb in dust from e-waste recycling sites had the highest level of chronic non-cancer toxicity risk and the Pb contamination originated from e-waste recycling activities. However, we did not investigate the chemical forms and bioaccessibility of Pb at that time. It is important to determine Pb bioaccessibility in surface soil and dust from e-waste recycling in order to conduct appropriate risk assessments. Additionally, the relationship between the chemical forms and bioaccessibility of Pb will improve our understanding of the characteristics of e-waste recycling activities.

In this study, we measured the bioaccessibility of Pb in e-waste surface matrices (i.e., dust and soil from the previously collected samples from the Philippines [8]) using an *in vitro* PBET assay, and examined bioaccessible Pb in simulated gastrointestinal fluids (i.e., the stomach and small intestine) to assess the risk of Pb ingestion. Pb speciation was determined using XAS. Combining these results from each sample with a dataset of total concentrations, bioaccessibility, and speciation of Pb, we examined the relationship between bioaccessibility and speciation to clarify the characteristics of Pb behavior in each gastrointestinal compartment.

2. Materials and method

2.1. Surface dust and soil from e-waste recycling

In 2010, we collected soil and dust samples from e-waste recycling sites in northern and southern Metro Manila: Caloocan (North, Metro Manila) and the provinces of Cavite and Laguna. We visited two formal sites in Cavite and Laguna and three informal sites in Caloocan (North) and Cavite. Previously, we measured total Pb concentrations in dust (150–130,000 mg/kg) and soil (23–7800 mg/kg) [8]. Based on the Pb concentration and type of e-waste recycling site, we selected 12 surface matrices (dust, $n = 8$ named D1–D8; soil, $n = 4$ named S1–S4) from these samples to assess Pb bioaccessibility and analyze the chemical forms of Pb. Table S1 lists the details of the soil and dust samples and provides a description of the sampling sites. According to a recent review by Chi et al. [28], e-waste recycling sites can be categorized as 'formal' or 'informal.' Formal sites are operated by approved companies that deal with a large amount of used products from affiliated clients and comply with environmental laws and regulations. Products at formal sites mainly consist of plastics, traditional forms of e-waste (e.g., CRTs, refrigerators, circuit boards, and wire cables), and more recent forms of e-waste (liquid crystal displays, and solar panels). In contrast, informal sites are illegal and have a small number of workers. Informal e-waste commonly consists of traditional forms of e-waste such as CRTs, circuit boards, and wire cables, etc. In the formal sector, Pb tends to accumulate in indoor floor dust without dilution effects, such as rain, wind, and soil mixing. At informal sites, there is generally an open-air environment in the workshop. In this study, we categorized sample types into only two groups, dust and soil, because the number of samples was limited to compare their differences by formal and informal (especially, for soil samples).