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Spatial patterns of potentially hazardous metals in paddy soils in a typical electrical waste dismantling area and their pollution characteristics

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ABSTRACT

Environmental pollution caused by electrical waste (E-waste) dismantling has attracted much attention from environmental researchers in China and worldwide. A total of 90 topsoil samples, 10 soil profile samples and 14 groundwater samples, were collected from a typical E-waste dismantling area in southeastern China. The spatial and vertical variations of potentially hazardous metals (PHM) in paddy soils and their potentially ecological risks were studied. The results showed that the mean total concentrations of five PHM (Cd, Cu, Pb, Zn and Ni) in paddy soils were 0.38, 35.13, 35.40, 121.38 and 28.13 mg kg^{-1}, respectively. The total Cd, and Pb concentrations in paddy soils had strong variability coefficients of 121.05% and 109.38%, respectively. Compared with the background values in Zhejiang province, the PHM were enriched in paddy soils. Part of the study area was seriously contaminated by these metals. The soil was contaminated with Cd, with high $P_{i\ Ave}$ value (> 1). The shallow groundwater was also contaminated by Cd in the E-waste dismantling area. Results of vertical variations of PHM in soils revealed significant accumulation of PHM in the topsoil (0–20 cm, 20–40 cm). Soil pH and organic matter (SOM) was significantly correlated with most of the total PHM in soils. Kriging interpolation and Moran's I were used to identify the contaminated hotspots of these PHM. It was found that high soil Cd, Cu, Ni, Zn were located in Da Xi and Zhe Guo towns, which was attributed to E-waste dismantling and other anthropogenic activities. Soil Ni was mainly influenced by the parent material. The PHM in soils may pose a potential threat to local ecosystem and human health.

1. Introduction

Potentially hazardous metals (PHM) contamination of agricultural lands continues to attract a great concern from environmental researchers worldwide, due to their inherent toxicity, non-biodegradability, persistence and bio-accumulation (Adriano, 2001; Huang and Gobran, 2005; Zhao et al., 2011; Wu et al., 2014; Gao et al., 2016). In China, > 20 million ha of arable lands suffered PHM pollution such as Cd, Cr and Pb, causing 1.2 million tonnes loss of crops annually (Wei and Chen, 2001). The major anthropogenic sources of PHM include industries “three wastes”, sewage irrigation, overuse of agrochemicals, mines and smelters, among which the electrical and electronic waste (E-waste) is becoming a major source of PHM released in the dismantling process.

Electronic and electrical products (EEP), such as washing machines (combined dryers), computers, air conditioners, mobile phones, are ubiquitous around the world. Due to continuing technological innovation, the average lifespan of electronic and electrical products has been continually declining (Widmer et al., 2005), which led to a rapid growth of E-waste. It was estimated that the rate of E-waste generation globally was approximately 42 million tonnes per year (Balde et al., 2015). Management of E-waste has being recognized as a great challenge, not only because of the annually increasing volume of E-waste, but also because of the toxic materials generated during the dismantling process. China plays an important role in EEP manufacture and E-waste recycling worldwide (Song and Li, 2014). In 2009, approximately 50% of the global major home appliances (e.g. washing machines, air conditioners, computers and televisions) were exported (240 million units)

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from China (Zhu, 2010). The annual domestic generation of E-waste continues to rise, with an estimated over 6 million tonnes in 2014 (Balde et al., 2015). Besides the domestic E-waste, approximately 1.5–3.3 million tonnes of E-waste are illegally imported to China annually (Zhou and Xu, 2012) from developed countries, especially in North America and Europe, due to inexpensive labor and lax regulation. It is reported that most of the world's E-waste was processed in China in 2012 (Zhang et al., 2012), while the rest goes to India, Nigeria and other developing countries (Breivik et al., 2014).

Recently, China has started to establish formal E-recycling facilities, moving E-waste indoors with different protection methods from hazardous materials. However, driven by economic profits, the informal and primitive E-waste recycling technologies, including acid-leaching of computer chips and components, open burning of printed circuit boards and cables for component separation (Gullett et al., 2007), plastic chipping and melting, and manual dismantling of cathode ray tubes (Song and Li, 2014; Chi et al., 2014), were widely used in China. The E-waste dismantling regions were mainly located in the southeastern China, such as Shantou in Guangdong Province and Taizhou in Zhejiang Province. A large number of toxic metals like Cd, Cu, Pb are released into the surrounding environment of the E-waste processing sites (Cui and Zhang, 2008). Therefore, these PHM may enter the soil environment through atmospheric deposition and water irrigation (Nan et al., 2002). Previous researches have demonstrated that significantly correlations were found between the PHM and the health of E-waste workers, local residents, especially children through ingestion, inhalation, and other exposure pathways (Zheng et al., 2013; Zhao et al., 2015a, 2015b). Meanwhile, the accumulation of PHM in soils can also negatively influence soil quality, reduce crop yield and pollute agricultural products (Nagajyoti et al., 2010). Therefore, it is important to carry out a comprehensive investigation of PHM contamination in soils in E-waste dismantling region for the long-term improvement of local residents' health.

Studies on PHM contamination in surface soil at E-waste processing areas have been carried out (Wong et al., 2007; Shen et al., 2008; Fu et al., 2008; Fu et al., 2014; Wu et al., 2014). Luo et al. (2011) reported that paddy fields and vegetable gardens were primarily contaminated by Cd and Cu. Zhao et al. (2010) found that PHM contamination in agricultural soils close to E-waste-dismantling workshops was very serious. However, there is still little information on the spatial distribution of PHM at horizontal and vertical scales. Meanwhile, it is also important to evaluate the threat of toxic PHM in topsoil to shallow groundwater by leaching in order to control or eliminate PHM pollution and ensure shallow groundwater safety. In our study, a typical E-waste dismantling region with a large number of informal family-operated recycling facilities in southeastern China was chosen. The objectives of this study were (1) to characterize the spatial variation of PHM in paddy soils of a typical E-waste dismantling region; (2) to comprehensively evaluate the potential ecological risk of PHM in soils; (3) to study the correlation between the PHM in soils and the corresponding concentration in shallow groundwater.

2. Material and methods

2.1. Study area and soil samplings

The study area was located in the northwest part of Wenling City, Zhejiang Province, southeastern China (Fig. 1), with a total area of approximate 450 km^2 (121°10′ to 121°44′E, 28°13′ to 28°32′N). It has a subtropical marine monsoon climate with mean annual temperature of 17.3 °C and an average annual rainfall of 1693 mm. Paddy fields is the major land use type of arable land and paddy soil (Gleysols) is the dominant soil type in the study area. Based on the hydro-geological data (Agricultural Bureau of Wenling), the thickness of the porous unconfined aquifer is about 2.5–3.5 m, and the shallow ground water level is approximately 0.8–2.5 m. The vertical flow is the major movement of groundwater and the velocity of horizontal flow was very low due to the hydraulic gradient in Wenling. There are a large number of family-operated recycling facilities to deal with e-wastes from domestic market and other developed countries. The E-waste recycling in Wenling was initiated by a few individuals who engaged in the dismantling experiments in the early 1990s. Currently, in the Wenqiao Town of Wenling City, there are 30 villages and 384 households are heavily involved in unauthorized E-waste handling (Chi et al., 2014).

In October 2015, a total of 90 topsoil samples were collected from the paddy fields in the study area (Fig. 1). For each sample, five subsamples of soil were collected within a radius of 10 m surrounding each sampling location and then mixed to provide a composite sample. At least 1 kg soil (0–20 cm in depth) was taken for each sample. Meanwhile, A GPS Trimble Asset Surveyor (version 5.20) with sub-meter accuracy was used for recording the geo-referenced coordinates. The distribution of soil sampling sites in the study is shown in Fig. 1.

Moreover, to systematically assess the PHM pollution and vertical variation at different depths, a total of 10 soil profiles were chosen (5 soil profiles collected from Da Xi and Zhe Guo towns of E-waste recycling area (E-waste group) and another 5 soil profiles collected from Song Men town of reference locations far away from E-waste recycling area (Contrast group, an average distance of approx. 24 km from E-waste group), respectively) (Fig. 1). The E-waste and Contrast groups have similar agricultural management measures, which include the same rice varieties (Hybrid rice, Yongyou #6) planting and similar chemical fertilizer application (240 Kg N ha^{-1}, 130 Kg P_2O_5 ha^{-1}, 240 Kg K_2O ha^{-1}). For each soil profile, five soil samples were collected at depth intervals of 0–20 cm (topsoil, TS), 20–40 cm (shallow soil, SS), 40–60 cm (middle soil, MS), 60–80 cm (deep soil, DS) and 80–100 cm (bottom soil, BS). Therefore, a total of 50 subsamples from the 10 soil profiles were collected.

The shallow unconfined aquifers are very vulnerable to natural and anthropogenic activities such as precipitation, agricultural management (fertilization and irrigation) and toxic substances dumping (Wu et al., 2014). Eight groundwater samples were collected from eight tube-wells in the E-waste recycling area and another six reference groundwater samples were also collected from six tube-wells which were far away the E-waste recycling area (an average distance of approx. 24 km from E-waste dismantling area), in order to study the impact of PHM in soils on the shallow groundwater quality. The groundwater levels of 14 tube-wells were about 0.8–1.5 m during the sampling period in 2015. The groundwater samples were taken following several steps: (1) using the well water to clean the polyethylene bottle (150 ml); (2) collecting the groundwater from 10 cm below the top of the water table with a polyvinyl chloride (PVC) Baylor tube and filter the water through a 0.45 μm filter membrane (Whatman, Cliton, NJ, USA) to remove sand and other impurities; (3) using the filtered groundwater to fill the 150 polyethylene bottle and modifying the pH value < 2 with ultra-pure concentrated nitric acid to avoid the biological activity (4) storing the water samples at approximately 4 °C before analysis. Meanwhile, the real-time water temperature (T, °C) and pH value of groundwater was measured on-site using a portable electronic instruments (XB89-M267, Midwest, China).

2.2. Laboratory analyses

All soil samples were air-dried at room temperature (20–22 °C) and sieved to pass a 2 mm nylon mesh. A portion of each sample was ground in an agate grinder and sieved through 0.149 mm. The prepared soil samples were stored in polyethylene bottles for further chemical analysis. Soil pH and electrical conductivity (EC) were analyzed using an aqueous suspension (soil-to-water ration of 1:2.5 and 1:5, W:V, respectively). Soil organic carbon was determined by the wet oxidation using concentrated H_2SO_4 and $K_2Cr_2O_7$, and titrating with Fe $(NH_4)_2(SO_4)_2 \cdot 6H_2O$. Soil particle size distribution (sand, silt and clay) was measured by the hydrometer method.