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Eco-toxicity and metal contamination of paddy soil in an e-wastes recycling area

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ABSTRACT

Paddy soil samples taken from different sites in an old primitive electronic-waste (e-waste) processing region were examined for eco-toxicity and metal contamination. Using the environmental quality standard for soils (China, Grade II) as reference, soil samples of two sites were weakly contaminated with trace metal, but site G was heavily contaminated with Cd (6.37 mg kg^{-1}), and weakly contaminated with Cu (256.36 mg kg^{-1}) and Zn (209.85 mg kg^{-1}). Zn appeared to be strongly bound in the residual fraction (72.24–77.86%), no matter the soil was metal contaminated or not. However, more than 9% Cd and 16% Cu was present in the non-residual fraction in the metal contaminated soils than in the uncontaminated soil, especially for site G and site F. Compared with that of the control soil, the micronucleus rates of site G and site F soil treatments increased by 2.7-fold and 1.7-fold, respectively. Low germination rates were observed in site C (50%) and site G (50%) soil extraction treated rice seeds. The shortest root length (0.2377 cm) was observed in site G soil treated groups, which is only 37.57% of that of the control soil treated groups. All of the micronucleus ratio of *Vicia faba* root cells, rice germination rate and root length after treatment of soil extraction indicate the eco-toxicity in site F and G soils although the three indexes are different in sensitivity to soil metal contamination.

1. Introduction

Electronic-waste (e-waste) refers to the end-of-life electronic products including computers, printers, photocopy machines, television sets, mobile phones, and toys, which are made of sophisticated blends of plastics, metals, and other materials. Disposal of the e-wastes is an emerging global environmental issue, as these wastes have become one of the fastest growing waste types in the world. In China, More than 5.14 million home appliances and 4.48 million personal computers are becoming obsolete each year [1]. Moreover, being a developing country, over one million tons of e-waste from the U.S., Europe and other areas of the world are flooding into China every year, taking advantage of the lower labor costs and less stringent environmental regulations [2]. These discarded electronics are usually classified as hazardous waste as electronics contain heavy metals including lead, chromium, cadmium, mercury, and beryllium [3], and other hazardous materials. Furthermore, brominated flame retardants (BFR) can be a component of printed circuit boards, plastic covers and cables [4,5]. Also, plastics in electronics would create dioxins when burned [6]. Most of these substances are considered hazardous and pose threats to human and environmental health. Therefore, if not disposed appropriately, they could become a source of toxic heavy metals or persistent toxic substances (PTS) [7].

Some studies have been focused on the environment pollution caused by e-wastes in China since the last decade. The accumulation of heavy metals, polybrominated diphenyl ethers (PBDEs) and PAHs in air [8], water [9], sediment [10,11] and soil [12–14] has been reported in some sites of e-waste recycling locations. However, since these chemical data do not take into account the possible combined effects of different contaminants, as well as their bioavailability, they provide only part of the knowledge necessary to evaluate the toxic potential for wildlives and human. Also it is difficult to make clearly hazard assessments and predictions of possible eco-toxicological effects of e-wastes based only on total concentrations. Therefore, a complementally bioassay strategy should be developed with a focus on the most important eco-toxicological effects. The biological endpoints are chosen according to their importance of known biological targets. Genotoxicity and/or the genome disruption are ones of the first targets concerned. Micronucleus (MN) assay in *Vicia faba* root cells was selected to evaluated the Genotoxicity and/or the genome disruption [15], which is highly sensitive and capable of detecting mutagens, clastogens and carcinogens from the environment, and showed excellent correlations with tests in the mammalian systems and human lymphocytes systems

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[16,17]. Seed (Rice seed) germination assay was selected to evaluated the phyto-toxicity of soil, which is a sensitive assay regulated by OECD to test the phyto-toxicity of chemical material [18].

Taizhou, situated on the winding east coast of middle China, is widespreadly famed for its beautiful natural scenery. However, now it has become one of the biggest e-recycling areas in China during recent decades. It is pity that, except few reports of PBDE contamination in soil [13], the metal contamination and the available eco-toxicological data of this area are relatively limited. To understand the extent and nature of environmental pollution caused by these e-waste processing activity, this paper aimed to study the trace contamination of paddy soil in Taizhou. The objective of this study were (1) to establish a basic understanding of the level and extent of trace metal contamination in paddy soils around the e-wastes processing sites in Taizhou; (2) to examine mobility, solubility and potential bioavailability of heavy metals in the environment; and (3) to evaluate the eco-toxicity of paddy soils, and to find relationship between eco-toxicity and metal contaminations.

2. Materials and methods

2.1. 2.1. Sampling area

The research area is located in the southeast of Zhejiang province, China (Fig. 1), under the north sub-tropic monsoon climate. The mean annual rainfall is 1600–1700 mm, of which 60.2% occurs from May to September. Annual mean air temperature is 17 °C, with the highest of 40.8 °C and the lowest of −9.9 °C. The number of e-recyclers in Taizhou has grown exponentially and recycling bases have proliferated rapidly since early 1990s. Historically, local e-recyclers get e-wastes from two major sources: domestic pipelines and foreign imports mainly from U.S., Japan and Taiwan region of China. Different counties of Taizhou usually specialize in different stages of e-waste processing, from manual dismantling, circuit board cooking, acid bathing and stripping, to open burning and dumping. In fact, according to the Economic and Trade Commission of Taizhou city, as of January 2005, more than 40,000 peoples in Taizhou work in the e-recycling industry and around 2 million metric tons of e-waste annually are processed.

Fig. 1. Maps of Taizhou and sampling sites. The map of Taizhou city was used as Ref. [19]. See Table 1 for a description of sampling sites.

2.2. Sample analyses

Based on the distribution of e-recyclers, seven sites (A to G) were selected for study (see location and description in Fig. 1 and Table 1). All the seven sites have e-waste recycling history, but only at site F and site G such operation were observed around the paddy soils during a personal interview. Site G is by far the biggest e-recycling village with 10 years of experience. All the soil samples were taken from the flooded paddy soils at the same time in December 2007. Soil samples were collected from the topsoil (0–20 cm soil layer) using a stainless steel shovel and stored in clean polyethylene bags to minimize sample contamination. The soil samples were firstly air dried at room temperature after transported to the laboratory, then sieved (<1 mm) for removing stones, roots and coarse materials, and finally stored in a desiccator prior to analysis. The pH, total organic carbon (TOC), total phosphorus, total nitrogen, and clay contents were determined according to the general methods [20]. Sequential chemical extractions were performed for the metal contaminated soil samples to fractionate the metal solid phases [21]. For total elemental concentrations, the dried and powdered soil samples were dissolved using a combination of concentrated nitric acid (HNO_3) and perchloric acid ($HClO_4$). Metal concentrations (Cd, Co, Cr, Cu, Fe, Mn, Pb, and Zn) in the digested solutions or extraction solutions were determined using Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES, Iris Advantage 1000) by the Instrumental analysis center of the Shanghai Jiaotong University. Before using, the glassware and plasticware were soaked overnight in a 1:1 (v/v) concentrated nitric acid and concentrated hydrochloric acid, and then rinsed thoroughly with double distilled water. For quality control, reagent blanks and replicates were set at same time.

2.3. Soil extraction

Ten grams dried soil were used to make soil extraction with deionized water as extractant (liquid-to-solid ratio was 10:1). After an agitation extraction with a speed of 120 rpm for 8 h, the soil solution was centrifuged in 3500 rpm for 10 min, and then the supernatant was filtered through a 0.45-μm membrane filter. The filtration was immediately prepared for MN assay in *Vicia faba* root cells and germination assay in rice seeds, respectively.

2.4. Micronucleus assay in Vicia faba root cells

Micronuclei are small masses of chromatin which appear beside the main nuclei when the plant is contacted with aneugen agents (fusorial poisons) or clastogen agents (inducing chromosome breaking). MN assay using *Vicia faba* or *Tradescantia* has been being one of the most frequently applied genotoxicity assays for detecting clastogenicity of contaminated soils [22], which has been recommended for using in mutation screening or monitoring by the Royal Swedish Academy of Sciences, Committee 17 of the Environmental Mutagen Society and the World Health Organization [23]. The micronucleus assay was performed as described by Ma et al. [23] with some minor modification. *Vicia faba* seeds bought from the local market were stored at 4 °C under dry conditions before using. Before soaking in distill water for 24 h, the seeds were firstly surfacely disinfected with 5% calcium hypochlorite solution and thoroughly rinsed with distilled water. Then the seeds were allowed lying in two moist filter papers at 25 °C to germinate for 2 days. The seedlings with primary roots long to 2–3 cm were soaked for 4 h in darkness at 22 °C in soil extraction, distilled water (as negative control), and 50 mg L^{-1} $CuSO_4$ (as positive control), respectively, followed by 22–24 h recovery before fixation. The roots tips were isolated and fixed at 4 °C for