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Original Research Paper

# Distribution Characteristics of Heavy Metals in E-Waste Recycling Sites

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## ABSTRACT

A field investigation was carried out on the distribution characteristics of heavy metals in the soils of electronic waste (e-waste) recycling location in Taizhou City, Zhejiang Province, China. The results showed that the concentrations of Cd, Cu and Pb were more than the Grade II values of soil quality standards from the State Environmental Protection Administration of China (SEPA), of which the most serious pollutants were Cd and Cu, followed by Pb. Based on the state standard values of soil quality in China, the comprehensive pollution index of Cd, Cr, Cu, Pb and Zn in the e-waste recycling locations was 4.3, that indicated that the e-waste recycling location's soil has been severely contaminated by heavy metals. Another result also showed that there existed compound pollution of Cu, Pb, Zn, Cr and Cd of the soils at test sites, and soils there were not suitable for agricultural cultivation any more.

## INTRODUCTION

Electronic waste, or e-waste for short, generally known as "waste electrical and electronic equipment", refers to any appliance using an electric power supply that has reached its end-of-life, including waste TV, waste computers, waste refrigerators, waste communication equipment and eliminated accurate electronic dashboards and such (Bhutta et al. 2011, Ha et al. 2009). E-waste contains many materials requiring special end-of-life handling, such as Cd, Ni, Cu, Pb and plastics capable of releasing, among other compounds, dioxins and furans, etc. (Sthiannopkao & Wong 2012, Wong et al. 2007 ). So the reasonable recycling of e-waste can bring about cheap materials and generous profits. But on the contrary, the primitive and unreasonable e-waste recycling processes cause many environmental questions, such as the harmful metals, polychlorinated biphenyls and polybrominated diphenyl ethers, which are harmful to human survival environment and health (Kiddee et al. 2013, Luo et al. 2011, Man et al. 2013, Xing et al. 2010). Since the last century, high technology industry such as electronic information has developed rapidly, electronic techniques have upgraded constantly to speed up, and more and more scrapped electronic products over the world are eliminated. In many countries, the most fast growing waste stream is electronic waste (Bhutta et al. 2011, Kiddee et al. 2013, Ni et al. 2014, Zhang et al. 2012). According to incomplete statistics, about 80% of the world's e-waste is exported to Asia, among which 90% flowed into China with the name of "Recycling" (Schwarzer et al. 2005).

Luqiao distinct, Taizhou city, Zhejiang Province, is one of the largest e-waste recycling centres. Local residents dismantle and recycle the e-waste by the way of baking, melting and strong acid leaching the circuit boards, or burning the current lead and electric cable in the open air. These primitive e-waste recycling activities have gained useful materials on one hand, such as precious metals, but have also led to the release of large quantities of toxic metals and organic pollutants into the surrounding environment (Fujimori et al. 2012, Luo et al. 2011, Xing et al. 2010). It has caused widespread attention in the past ten years. The State Environmental Protection Administration of China has strictly prohibited the open fire and random dumpling of the e-waste, but driven by the profit, all the efforts proved invalid**.** Although the environmental protection department of Luqiao distinct has carried out centralized rearrangement and focused all the e-waste recycling workshops on the same road, but due to the relatively primitive recycling methods, the pollution of the environment brought about by the recycling workshops still continuing. In addition, even though the environmental department sent directives to forbid nonstandard e-waste recycling activities, but local e-waste recycling activities still continued off and on; they stopped when the regulation was strict, but continued when the regulation was easy-going, thus the environmental pollution problems are still severe.

The present study was carried out in Fengjiang, a typical e-waste recycling location in Luqiao distinct, Taizhou city, Zhejiang Province, China. There million tons of e-waste, such as home electric appliances shell, panels and old wires, etc. have been dismantled intensively annually nearly 30 years. Most of the uncontrolled e-waste recycling sites are located on or close to agricultural fields. Potential contamination of soils may pose a significant threat to humans. The purpose of this research is to investigate the content level, pollution level and distribution characteristics of heavy metals by the uncontrolled e-waste recycling activities in the present. The results can be useful for the protection of the local community from potential health hazards.

## MATERIALS AND METHODS

**Soil samples collection:** Samples were collected at 0m, 100m, 200m and 300m away from an e-waste recycling centre in September, 2013. In each distance, three parallel sample sites were selected about 50m away from transverse distance. The control sample (CS) was collected from a farmland without e-waste recycling activities about 50 km away. In each sampling site, three parallel samples were collected. About 1 kg top soil (0-15 cm) was collected, then all the samples were wrapped with aluminium foil, put in polythene zip-bags, and transported to the laboratory, dried at room temperature, milled with agate sticks, made them through 200 nylon mesh, and stored in glass bottles at -4$^{o}$C until further processing. All the results were reported as dried weight.

**Sample extraction and analysis:** We took 0.5000 ± 0.0005 g dried soil samples, put them in big glass tubes, treated with the nitric acid and perchloric acid digestion method (Zheng et al. 2010), and determined the contents of Cu, Cd, Zn, Pb and Cr ions with ICP-OES. Most of the chemical reagents were analytical pure and used without further treatment, and deionized water was used in the experiments. Quality control was conducted by using the national standard substances soil. At the same time, setting method blank, the analysis errors were within the allowable range.

## RESULTS AND DISCUSSION

**Physical and chemical properties of the soils in e-waste recycling sites:** Total phosphorus (TP), pH value, total nitrogen (TN), ammonium nitrogen $NH_4$-N, total organic carbon (TOC) and other physical and chemical properties of the soils in the e-waste recycling locations were investigated, and the results are given in Table 1. When comparing metal concentrations of the soil with quality standards of the State Environmental Protection Administration of China (SEPA), pH value is necessary. From Table 1, the pH value, TOC, TN, $NH_4$-N and TP had no significant difference, suggested that the basic physical and chemical properties of soil sampling sites had no significant differences. Compared with the second general survey of the national soils, the paddy soil's nutrient contents in the region (TOC: 24.5 mg/g; total nitrogen: 2.45 g/kg; total phosphorus, 0.41 mg/g) increased in some degree, while pH values were slightly lower (pH 6.0). Accordingly, the e-waste recycling activities in the investigation region did not lower the quality of its surrounding farmland soil's fertility, but reduced the soil pH value in some degree; this meant that the soil got acidified to some degree due to the e-waste recycling activities. There are lots of villager workers engaged in primitive e-waste recycling operations without the use of adequate protective equipment. Such primitive operations include, but are not limited to, stripping of metals in open pit acid baths. Then the waste acid is poured to farmland, terrace channels, etc. without any treatment. Doubtlessly, these activities led the soils to acidify in the long run. Soil acidification then made soil structure destroying and soil crusting. At the same time, the most important thing is that with the soil acidification, the heavy metals in the soil changed to water-soluble, or exchangeable metal ions, then the soil pollution in the study area might be on the rise. So it is worth concerning about the pollution of the neighbouring farmland near the e-waste recycling centre.

**Content level and distribution characteristics of heavy metal contamination in the soils of e-waste recycling sites:** The heavy metal concentration measured in soil samples is given in Table 2, together with the control site. The most abundant metals, Cr, Cu, Cd, Pb and Zn were investigated in the environmental samples. From Table 2, Cr, Cu, Cd, Pb and Zn contents of the e-waste recycling sites surrounding soils were significantly higher than the background values in the region (Cr: 58.51 mg/kg, Cu: 19.77 mg/kg, Cd: 0.20 mg/g, Pb: 24.49 mg/kg, Zn: 84.84 mg/kg) (Pan et al. 2007, Zheng et al. 2010). Cu and Cd were the most serious metal pollutants in the area, the maximum detected concentration of Cu was 519.3 mg/kg, and the minimum detected concentration was 249.0 mg/kg; the maximum and minimum were 10.4 and 5.0 times of the Grade II value of soil quality standards from SEPA. In addition, the maximum and minimum detected concentrations of Cd were 4.5 mg/kg and 0.8 mg/kg; they were respectively 9.0 and 2.7 times of the Grade II value of soil quality standards from SEPA. The survey also found that the maximum detected concentration of Pb is 56.9 mg/kg, this value was more than the Grade II value of paddy land, vegetable land, dry land quality standards from SEPA, indicating that the surrounding soils of the e-waste recycling locations were not suitable for agricultural cultivation anymore, and only could be used as orchard soils. Moreover, the survey showed that the Cr and Zn contents for the locations were low, not surpassed the Grade II values of soil quality standards from SEPA. This phenomenon appeared just for the e-waste recycling locations almost did not contain or contained little of e-wastes containing Cr, Zn,

Table 1: Physical and chemical properties of the soils.

| Distance (m) | pH ($H_2O$) | TP (µg/g) | TN (mg/g) | $NH_4$-N (µg/g) | TOC (mg/g) |
|---|---|---|---|---|---|
| 0 | 5.89±0.11 | 689.05±24.85 | 0.98±0.09 | 78.25±14.71 | 3.23±0.31 |
| 100 | 5.61±0.21 | 766.00±35.13 | 1.13±0.11 | 62.69±4.13 | 3.11±0.23 |
| 200 | 5.73±0.09 | 451.39±17.73 | 1.04±0.08 | 82.68±0.96 | 3.07±0.37 |
| 300 | 5.67±0.07 | 913.98±7.27 | 1.29±0.13 | 75.61±9.8 | 3.35±0.46 |

Table 2: Heavy metal ions concentration in e-waste recycling location soils (mg/kg).

| Distance (m) | Cd | Cr | Cu | Pb | Zn |
|---|---|---|---|---|---|
| 0 | 4.5±0.4 | 27.7±2.1 | 519.3±35.0 | 56.9±12.1 | 145.3±23.1 |
| 100 | 2.8±0.1 | 25.1±1.9 | 495.6±13.5 | 23.7±2.6 | 127.9±8.7 |
| 200 | 2.7±0.3 | 22.3±1.2 | 299.1±21.2 | 20.2±2.1 | 114.3±7.0 |
| 300 | 0.8±0.2 | 19.3±0.8 | 249.0±28.6 | 11.8±3.2 | 94.9±7.4 |
| CS | ND | 7.1±0.2 | 32.1±2.1 | 9.6±1.1 | 44.9±8.9 |

such as tapes, video tapes, etc.

As given in Table 2, the content of Cd and Cu at each sampling site surpassed the Grade II values of soil quality standards from SEPA. The content of Pb in the center of the e-waste recycling location was more than the Grade II values of the paddy land, dry land and vegetable land quality standard from SEPA, indicating that the e-waste recycling activities contributed to the pollution of the surrounding soils. Besides, the contents of Cd, Cr, Cu, Pb and Zn decreased with increasing the distance away from the recycling centre in the investigation 300m away from the e-waste recycling locations. At the same time, the comprehensive pollution index of Cd, Cr, Cu, Pb and Zn in the e-waste recycling locations was 4.3, counting with the Grade II value of soil quality standards from SEPA. This indicated that the e-waste recycling locations have been seriously polluted by heavy metals, and are not suitable for agricultural cultivation anymore.

The pollution features of heavy metals in Luqiao were not similar to those in Wenling district, another city in Taizhou. Tang et al. (2010) found that paddy soils were heavily contaminated with Hg, and weakly contaminated with Cu, Cd, Pb and Zn in Wenling. Unlike Wenling city, Luo et al. (2011) investigated the heavy metal pollution condition of the farmland nearby the e-waste recycling locations in Longtang and Shijiao towns in Guangdong Province, and found that the over standard rate of Cu and Cd all achieved above 60%, while Pb was 48.5%, which was closer to the result of our study, perhaps for the e-waste types and the recycling processes in these two places were close to each other.

Xu et al. (2012) have investigated the heavy metal pollution situation of the same area in farmland, and found that total detection of Cu and Cd in the soils was also more than the Grade II value of soil quality standards from SEPA, and their concentrations were close to our study. The concentration of Pb and Cu were not over the Grade II value of soil quality standards from SEPA. The concentration of Cd reported by Xu et al. (2012) was much higher than our study. In addition, the related soil was acidified obviously (pH: 3.8~4.4), which might be due to the higher concentration of Cd in the soil. But in our investigation, the pH values of the soil arranged from 5.61 to 5.89, little higher than theirs. Pan et al. (2007) found that the content of Cu in the same area was 435.6mg/kg, which was close to the result of our study. Zheng et al. (2013) investigated the same area of farmland soil to investigated the heavy metal pollution characteristics and spatial distribution of the soil, and found that the content of Cu, Zn, Pb and Cd was 118mg/kg,169mg/kg,47.9 mg/kg and 1.21mg/kg, respectively. Among them the content of Cu was 1/2~1/5 of this survey, significantly the content of Cu and Cd were all lower than this study, perhaps due to accumulation speed of Cu and Cd is faster after nearly three years of e-waste disassembling, which significantly increased the content of Cu and Cd in soil.

## CONCLUSIONS

E-waste disassembling activities were usually operated within villages, due to the crude simple household workshops and nonstandard recycling ways, which have caused the serious environmental problems in Taizhou city, including the accumulation of heavy metals in the surrounding environment. The results of the present study suggested that the soil in e-waste recycling locations of Luqiao distinct was heavily polluted by heavy metals. Among the survey, heavy metals (Cd, Cr, Cu, Pb and Zn), except Cr and Zn, the rest of the investigated heavy metals were all surpassed the Grade

II values of soil quality standards from SEPA. Among them, the most serious pollution was by Cu and Cd, followed by Pb. The comprehensive index of Cd, Cr, Cu, Pb and Zn in the soils around the e-waste recycling locations abandoned with the Grade II values of soil quality standards from SEPA was 4.3. It has reached to serious pollution showing that there existed severe heavy metal pollution problems in the soils surrounding the e-waste recycling locations, and the soils were not suitable for farming anymore.

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