

Heavy Metals Concentrations of Surface Dust from e-Waste Recycling and Its Human Health Implications in Southeast China

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Received July 27, 2007. Revised manuscript received
 December 27, 2007. Accepted January 5, 2008.

The recycling of printed circuit boards in Guiyu, China, a village intensely involved in e-waste processing, may present a significant environmental and human health risk. To evaluate the extent of heavy metals (Cd, Co, Cr, Cu, Ni, Pb, Zn) contamination from printed circuit board recycling, surface dust samples were collected from recycling workshops, adjacent roads, a schoolyard, and an outdoor food market. ICP-OES analyses revealed elevated mean concentrations in workshop dust (Pb 110 000, Cu 8360, Zn 4420, and Ni 1500 mg/kg) and in dust of adjacent roads (Pb 22 600, Cu 6170, Zn 2370, and Ni 304 mg/kg). Lead and Cu in road dust were 330 and 106, and 371 and 155 times higher, respectively, than non e-waste sites located 8 and 30 km away. Levels at the schoolyard and food market showed that public places were adversely impacted. Risk assessment predicted that Pb and Cu originating from circuit board recycling have the potential to pose serious health risks to workers and local residents of Guiyu, especially children, and warrants an urgent investigation into heavy metal related health impacts. The potential environmental and human health consequences due to uncontrolled e-waste recycling in Guiyu serves as a case study for other countries involved in similar crude recycling activities.

Introduction

Of the different e-waste processing activities carried out in Guiyu, China, the recycling of printed circuit boards is probably the most important input of heavy metals to the surface environment. The recycling procedure involves melting of solder from circuit boards over makeshift coal grills, and removal and sorting of electrical components such as chips, capacitors, and diodes which are sold to electrical appliance factories. Portable household fans are the only precautionary measures taken to reduce worker exposure to toxic solder fumes. Circuit boards are laden with heavy metals: Cu, used for its conductive properties, is either plated or etched onto surfaces of fiberglass epoxy resin boards; Pb is

used in Pb–Sn soldering and helps to prevent oxidation; and Co and Ni have magnetic and structural applications (1).

Dust is a significant environmental media that can provide information about the level, distribution, and fate of contaminants present in the surface environment. As the composition of settled dust is similar to atmospheric suspended particulates, it can be an indicator of pollutants such as heavy metal contamination in the atmosphere (2). Furthermore, the elemental composition and concentrations in dust reflect the characteristics of short- and long-term activities in the area (3). Previous heavy metal dust investigations in southeast China have included roads (4, 5), tunnels (6), urban parks (7), playgrounds (8, 9), children's nurseries (10) and households (11).

Humans can become exposed to heavy metals in dust through several routes which include ingestion, inhalation, and dermal absorption. In dusty environments, it has been estimated that adults could ingest up to 100 mg dust/day (12, 13). Children are usually exposed to greater amounts of dust than adults as a result of pica and play behavior (14, 15). Exposure to high levels of heavy metals can result in acute and chronic toxicity, such as damage to central and peripheral nervous systems, blood composition, lungs, kidneys, liver, and even death. Lead levels in dust have been significantly associated with Pb levels in children's blood (16), and a blood lead level (BLL) greater than an intervention level of 10 $\mu\text{g}/\text{dL}$ has been associated with a decrease in IQ (15).

Two recent studies have demonstrated elevated body loadings of heavy metals (17) and persistent toxic substances (18) in children and e-waste workers, respectively, at Guiyu. A study of 165 children aged 1–6 yr at a kindergarten revealed that 81.8% had BLLs greater than 10 $\mu\text{g}/\text{dL}$ (mean: 15.3 \pm 5.79 $\mu\text{g}/\text{dL}$). It was concluded that children with the highest BLLs were of parents who recycled circuit boards, whereas lower BLLs were found in children whose parents recycled plastics (17). Compared to a study (19) of children aged 1–5 yr living in Shantou City (30 km from Guiyu), the average BLL was 7.9 \pm 3.6 $\mu\text{g}/\text{dL}$; approximately two times lower than that of Guiyu children. A preliminary study by Brigden et al. (20) showed that Pb levels of single dust samples collected from the homes of two solder recovery workers were 4–23 times higher than that from a neighboring house where none of the residents were involved in e-waste activities. Therefore, workers inadvertently transported the contaminant from the workshops back to their homes, increasing the exposure of their children to the heavy metal. Elevated levels of heavy metals and POPs have been detected in the environment of Guiyu (21–26) and health checks on workers and children have uncovered digestive, neurological, and respiratory problems and high incidences of bone disease (27).

The objectives of this study were to determine the concentrations and spatial distribution of seven heavy metals (Cd, Co, Cr, Cu, Ni, Pb, Zn) in surface dust at a printed circuit board recycling district, in particular workshops and nearby public places such as roads, a schoolyard, and an outdoor food market and to estimate the potential health risk to adults and children via dust ingestion. This is one of the few studies that characterize the extent of environmental pollution caused by e-waste processing and its associated human risks.

Experimental Section

Study Area. Guiyu, a village in Guangdong Province, southeast China, has a population of 150,000 and has been intensely involved in e-waste "recycling" for more than ten years. The extraction of electrical components and solder recovery from

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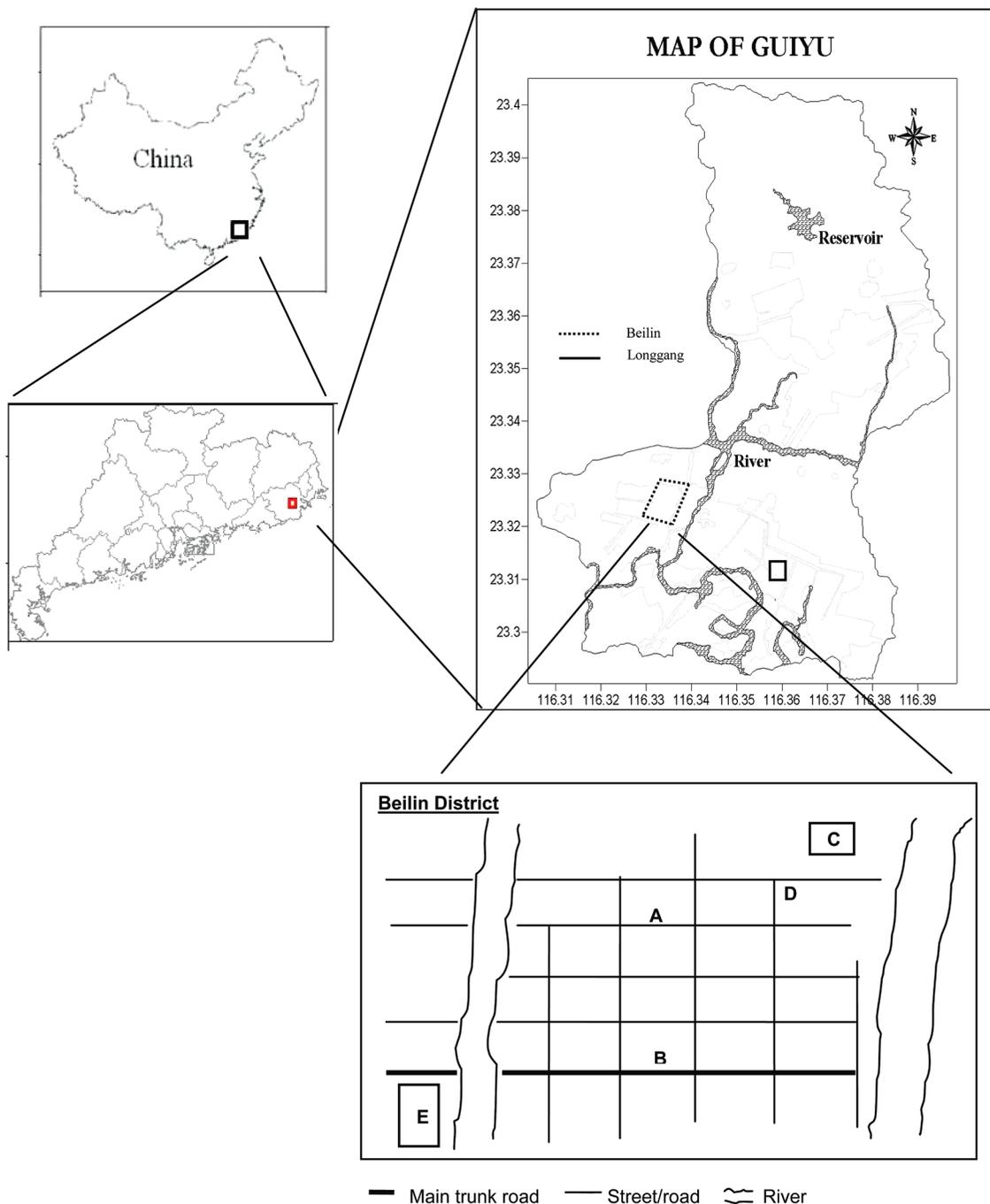


FIGURE 1. Map of Guiyu.

circuit boards are mainly conducted in family-run workshops in the residential district of Beilin. Circuit board components litter the roads and the area is characterized by distinct acrid fumes and odor from molten solder which irritate the eyes and throat. Plastic recycling is carried out in the Longgang district of Guiyu. Plastics such as acrylonitrile-butadiene-styrene, high-density polyethylene, and polyvinyl chloride are manually separated from e-waste and mechanically shredded into small fragments. By immersing the fragments into large basins of water, they are crudely sorted according to differences in densities. The plastic fragments are then spread out on roads to dry and may be processed further by mechanical grinding to fine powder (20). The climate in Guiyu is subtropical monsoon with prevailing northwesterly winds in winter and southeasterly winds in summer.

Sample Collection and Preparation. Deposited surface dust samples were collected in December 2004, during the dry season when prevalence of dust was expected to be

greater. Replicate samples (each approximately 250–400 g) were collected using plastic brushes and dustpans by gentle sweeping motion to collect fine particulates. After each sampling, brushes and dustpans were cleaned with paper towels. Sampling locations included four printed circuit board recycling workshops (PCBRW), roads, and public places (schoolyard, outdoor food market) within Beilin. For comparison, samples were also collected from two roads in Longgang district, a road in Gurao town (no e-waste activities) located approximately 8 km northeast of Guiyu, and three locations within Shantou University campus located 30 km east of Guiyu. A map of Guiyu and the sampling areas are shown in Figure 1 and the sampling locations are described in Table 1. As it was not possible to obtain samples from within recycling workshops located on Street B-1, samples were obtained from similar workshops located 100–200 m southwest of the food market. It was assumed that the

TABLE 1. Description of Sampling Locations

sampling site	location (refer to Figure 1)	GPS coordinates	description
Beilin district, Guiyu			
PCBRW ^a -1 (<i>n</i> = 4)			Dust collected inside workshop, 3 grills
PCBRW ^a -2 (<i>n</i> = 4)			Dust collected inside workshop, 3 grills
PCBRW ^a -3 (<i>n</i> = 4)			Dust collected inside workshop, 2 grills
PCBRW ^a -4 (<i>n</i> = 4)			Dust collected outside workshop within small enclosure beside pile of printed circuit boards, underneath exhaust fan. Number of grills unknown
street B-1 (<i>n</i> = 16)	A	N23.328 E116.341	Street lined with PCBRWs on both sides, road approximately 7 m wide
street B-2 (<i>n</i> = 16)	B	N23.323 E116.341	A primary trunk road (2-lanes) (Guojing Road) approximately 10–12 m wide with sidewalks 2–4 storey residential buildings line both sides of the street where ground floors are reserved for small family owned shops for sorting and selling integrated circuits (ICs), electronics, cigarette, liquor, and tea Few printed circuit board recycling workshops Many pedestrians, cyclists and few motorists (mopeds, cars, 3-wheel trolley taxis), AADT < 300
schoolyard (<i>n</i> = 4)	C	N23.330 E116.345	Beilin Primary/Secondary School located on the northern perimeter of printed circuit board recycling workshop area
near school (<i>n</i> = 4)	D		Road dust samples were collected in front of entrance to school, small canteen across from school and at a nearby intersection
food market (<i>n</i> = 14)	E		An outdoor market selling fish, vegetables, meat
Longgang district, Guiyu			
street L-1 (<i>n</i> = 4)			Road dust collected around shredded plastic fragments placed on road to dry outside a plastics recycling workshop
street L-2 (<i>n</i> = 4)		N23.313 E116.360	Road dust collected around shredded plastic fragments placed on road to dry outside a plastics recycling workshop
Gurao town			
street G-1 (<i>n</i> = 16)		N23.361 E116.413	Street consisting of hotel, sewing factories, undergarment shops located 8 km from Guiyu
Shantou University			
SU-1 (<i>n</i> = 4)		N23.415 E116.628	Quiet tree-lined road in front of the teachers living quarters, AADT < 25
SU-2 (<i>n</i> = 4)		N23.421 E116.626	Quiet tree-lined road near a conference center, AADT < 25
SU-3 (<i>n</i> = 4)		N23.414 E116.634	Near carpark

^a PCBRW: printed circuit board recycling workshop.

composition of dust in workshops of these two locations was similar since the circuit board recycling processes were the same.

Road dust samples were collected by sweeping from the middle of the road toward the edge of the road in an area of approximately 3.5 m × 3.5 m. Both sides of the road (spanning approximately 250 m) were swept. All samples were stored in paper bags (previously heated at 50 °C overnight to remove volatiles) and then sealed in polyethylene bags (Ziploc) for transport to the laboratory. The samples were then placed in a desiccator to rid them of moisture, sieved (<2 mm), and homogenized. This particle size range was chosen to facilitate comparison of heavy metal concentrations with soil guidelines (<2 mm). Dust samples (~5 g) were ground to a fine powder texture with mortar and pestle prior to chemical analyses.

Analyses of Metals in Dust. The ground dust samples were analyzed for Cd, Co, Cu, Cr, Ni, Pb, and Zn. Each sample (0.250 g) was placed into precleaned Pyrex test tubes and digested on an aluminum heating block (DK Heating Digester, VELP Scientifica, Milano, Italy) with HNO₃ (69%, 8.0 mL) and HClO₄ (2.0 mL) at 50 °C for 3 h, 75 °C for 0.5 h, 100 °C for 0.5 h, 125 °C for 0.5 h, 150 °C for 3 h, 175 °C for 2 h, and

190 °C until dryness (28). This was a “pseudo” total acid digestion because heavy metals strongly associated with or entrapped within silicates are not recovered. After cooling, HNO₃ (5%, 10 mL) was added and heated at 70 °C for 1 h with occasional mixing, and the cooled mixtures were decanted into polyethylene centrifuge tubes (15 mL) and centrifuged (3300 rpm, 10 min). The clear solutions were pipetted into clean centrifuge tubes for measurement of metals using inductively coupled plasma-optical emission spectroscopy (ICP-OES, Perkin-Elmer Optima 3000DV). All glass- and plastic-ware used were previously soaked overnight in HNO₃ (10%) and rinsed thoroughly with Milli-Q water. For quality control, reagent blanks and replicates made up 10 and 20%, respectively, of the sample population. The recoveries of standard reference materials (NIST SRM 2711 Montana Soil and NIST SRM 2709 Joaquin Soil) were satisfactory and the ranges were as follows: Cd (86–95%), Co (98–132%), Cr (69–78%), Cu (80–89%), Ni (90–100%), Pb (87–96%), and Zn (91–108%).

Statistical Analysis. Analysis of variance (ANOVA) was performed on all experimental data and means were compared using the Duncan’s Multiple Range test with SPSS version 11 software.

Risk Assessment. Risk assessment with regard to exposure to metal-contaminated dust by ingestion was carried out to estimate the noncancer toxic (chronic) risk of circuit board recycling workers and the general public including children living in Guiyu. Estimation of risk was calculated based on equations detailed in USEPA's Exposure Factors Handbook (29). Average daily dose (ADD) was determined by the following equation:

$$ADD = \frac{C \times \text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}}$$

where C is the mean heavy metal concentration (mg/kg) in dust. Conservative estimates of dust ingestion rates, IngR, were chosen for adult (100 mg/day) and child (200 mg/day) scenarios (29). An average body weight, BW, of 60 kg for adults (30) and 15 kg for children was assumed. In this study, exposure frequency, EF = 350 days/year; exposure duration, ED = 6 years; and the averaging time, AT = 2190 days. Noncancer toxic risk was determined by calculating the hazard quotient, HQ, where $HQ = ADD/RfD$ and RfD is an estimate of the daily exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime. Therefore, $HQ \leq 1$ suggests unlikely adverse health effects whereas $HQ > 1$ suggests the probability of adverse health effects (31). An $HQ > 10$ is considered to be high chronic risk (32). It was also assumed that the toxic risks due to the heavy metals were additive, therefore the HQ for each metal at a location scenario was summed to generate the hazard index (HI).

Results and Discussion

Heavy Metal Concentrations in Dust. The mean heavy metal concentrations of dust collected from the workshops, roads, and paved areas of public places are shown in Figure 2. Many of the results revealed large standard deviations indicating the highly heterogeneous nature of the dust samples. To evaluate the extent of heavy metal contamination in the dust, the concentrations were compared to several soil guidelines (Supporting Information Table S1). Currently, there are no guidelines or regulations for heavy metals in dust. In general, the metal concentrations of workshop dust were statistically higher ($p < 0.05$) than those from the other sampling sites.

Printed Circuit Board Recycling Workshops. Of the seven heavy metals investigated, Pb, Cu, and Zn levels of workshop dust were extremely elevated. The Pb concentration ranged from 22 900 to 206 000 mg/kg and exceeded the New Dutch List optimum value by 269–2426 times and the action value by 43–389 times suggesting the presence of fine elemental Pb particles in the dust. The mean Pb concentrations was $110\,000 \pm 61\,200$ mg/kg and was 29 times higher than the mean Pb levels of three floor dust samples from printed circuit board component separation workshops in East Delhi, India (20). Copper and Zn exceeded the New Dutch List optimum values by 31–994 and 7–73 times, respectively, and the action value by 6–188 and 1.4–14 times, respectively. Copper also exceeded the less stringent Chinese grade III guideline (industrial activity) by 3–89 times. The concentration of Ni varied quite remarkably between the workshops. In PCBRW-3, the concentration ranged from 76.1 to 118.6 mg/kg, and in PCBRW-2, the concentration was approximately 9–67 times higher (664–7967 mg/kg). The mean concentration of Ni in PCBRW-2 exceeded the New Dutch List action value by 19-fold. Mean Cd concentrations of PCBRW-1, PCBRW-2, and PCBRW-3 exceeded the action value with minimum and maximum concentrations ranging from 9.34 to 66.8 mg/kg. Mean Cr concentrations were all below the New Dutch List optimum value, however one dust sample from PCBRW-1 and two samples from PCBRW-2 were in exceedence. Cobalt concentrations of individual dust samples were generally

below the optimum value with the highest concentration (55.8 mg/kg) at PCBRW-2. Mean workshop dust concentrations for Cu, Ni, and Pb all exceeded the Canadian guidelines (industrial activity).

In general, no significant differences in metal concentrations were found between dust collected from inside workshops (PCBRW-1, PCBRW-2, PCBRW-3) and at PCBRW-4 (where dust was collected from a small enclosed area surrounding a vent of the workshop), indicating the transport of contaminated dust from within workshops via the exhaust fans.

Public Places. When comparing metal concentrations in dust from within workshops with that from roads and public places, Cd, Cu, Ni, Pb, and Zn were significantly higher ($p < 0.05$). Mean Pb concentrations from workshop floors were approximately 5 times higher than that of Pb collected from road (Street B-1) lined with workshops and 112 times higher than dust Pb from the main trunk road (Street B-2). Therefore, mean Pb concentration on Street B-1 was about 23 times greater than that on Street B-2; the latter had only a few recycling workshops which explained the lower Pb enrichment. Compared to the control sites, the mean Pb concentrations of Street B-1 and B-2 were approximately 370 and 15 times higher than Pb levels detected at Street G and SU, and Pb in Longgang dust was as much as 222 times lower than that on Beilin streets. Dust collected from Street L-1 and L-2 contained Cd, Co, Ni, Zn concentrations that were similar to those at Street B-2, the schoolyard, and the outdoor market, however, Cr (similar to those of workshops) and Cu levels were higher. For roads, similar trends were found for concentrations of Cd, Co, Cr, Cu, and Zn, whereby $\text{Street B-1} \gg \text{Street B-2} > \text{Street L-1, L-2} \geq \text{Street G and SU}$.

For schoolyard dust, exceedences of the Canadian (residential/park) guidelines for Pb and Cu were by 3.3–6 and 2.5–13.2 times, respectively. Dust collected from four areas at an intersection near the school also revealed elevated concentrations which were higher than those found at the schoolyard, except for Cd. Lead concentrations near the school entrance and outside a small open air canteen located about 4 m from the school entrance were 1110 and 3113 mg/kg, respectively. Therefore, children eating at this canteen, which was in close vicinity to recycling workshops, would be exposed to contaminated dust. Nickel was also extremely high (2911 mg/kg) at this location and was comparable to concentrations found within the workshops. At the open air food market, there were exceedences of the New Dutch List optimum values for Cu, Ni, Pb, and Zn by 10, 5.4, 16, and 4.5 times, respectively. These high values are a concern because food market items (i.e., vegetables) which are often placed on top of newspapers or in plastic buckets on the ground could easily come into contact with contaminated dust especially during the dry season.

Comparison with Other Studies. A comparison of heavy metal levels in dust collected from roads, carparks, urban parks, playgrounds, and schools in Hong Kong and other cities, according to particle size, is shown in Supporting Information Table S2. In general, dust particle sizes investigated and heavy metal digestion methods used are various and thus do not allow for easy comparison among studies. Of the limited studies for the <2 mm range, some of the highest dust Pb levels were measured from roads in Coventry, England (ND–3300 mg/kg) (33) and a vehicle tunnel ceiling of Hong Kong (375–1410 mg/kg) (6). High Pb dust levels have also been reported in London households (max 36 900 mg/kg) (34) and near a closed-down Pb smelter in Granite City, IL (110–25 000 mg/kg) (35) which were similar to the lower-end concentrations detected in Guiyu workshops. Lead concentrations in Street L-1 and Street L-2 dusts were comparative to Guangzhou and Xian road dusts (5, 36), however Cu was found to be 8.5 and 17 times higher,

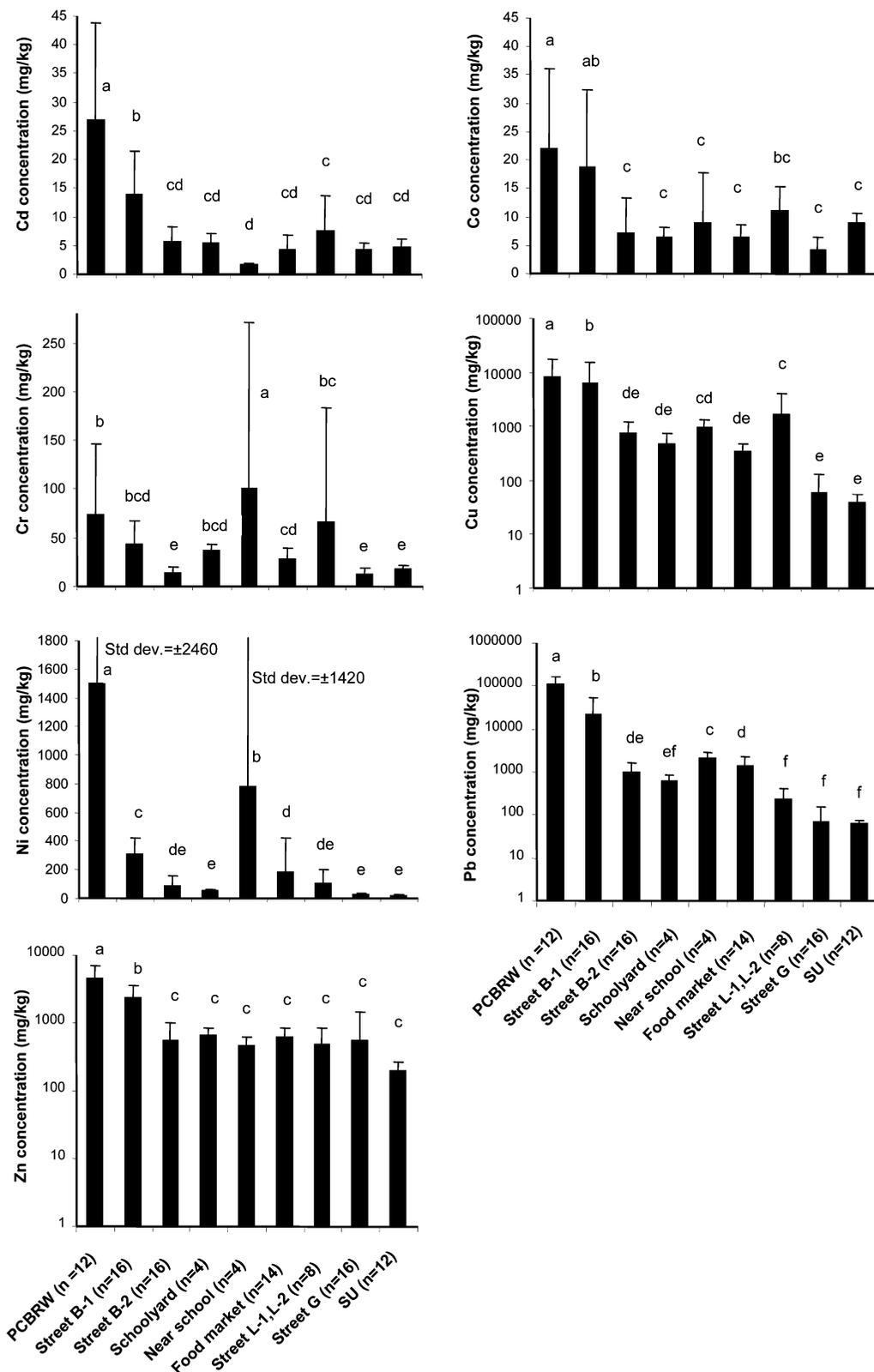


FIGURE 2. Heavy metal concentrations of dust samples (mg/kg) (<2 mm).

respectively. Street L-1 and Street L-2 dust Cu concentrations were significantly higher ($p < 0.05$) than all sampling sites except for workshops and Street B-1 and were higher than that reported by Brigden et al. (777 mg/kg) (20). When comparing metal concentrations of the Guiyu schoolyard with other dust studies in school playgrounds, Pb, Cu, and Cd were frequently higher (8, 10, 34), while Pb at playgrounds of Britain mining villages were ten times greater (34). It is noted that heavy metal concentrations in our study may be

greater for smaller particle sizes as studies by others have shown that metal concentrations increased with decrease in particle size (37).

Correlations Between Heavy Metals in Dust. There were significant associations between Co and Ni ($p < 0.01$) and Co with Cr and Cu ($p < 0.05$) in dust collected from the workshops. In Street B-1, Cd was significantly correlated with Pb ($p < 0.01$) and with Ni and Zn ($p < 0.05$). This suggests that elevated Cd levels were associated with higher levels of

other metals. Metal correlations for Street B-2 dusts (Cd and Co, Cu and Pb, $p < 0.05$) were different than that of Street B-1 dusts which seem to indicate different sources of heavy metals. The shops on Street B-2 mainly sold electrical components (i.e., integrated circuits) whereas the main industry on Street B-1 involved the removal of electrical components and solder from circuit boards by heating. Heavy metals found in Street B-2 dust may have been attributed to vehicle exhaust from heavier traffic. Lead and Co, and Ni and Zn were found to be significantly correlated ($p < 0.05$) at the schoolyard. Strong associations were found for Cd and Cr with many metals for Street L-1, L-2, G2-1 and SU. Chromium was significantly correlated with Ni. Significant correlations between metals indicate a common source.

Risk Assessment. Noncancer Toxic Risk. The calculated HQs and HIs using mean and maximum measured heavy metal dust concentrations and U.S. EPA reference doses (38) for the dust ingestion pathway for adult and child scenarios at various sampling locations are presented in Supporting Information Table S3. Of the seven heavy metals, the hazard quotient for Pb was highest at all locations. For a printed circuit board recycling worker, the HQ (calculated using mean concentration) was 50.2 indicating that the estimated oral ADD exceeded the "safe" oral Pb reference dose by 50 times. For an adult of 60 kg average body weight and oral RfD of $3.5 \mu\text{g}/\text{kg}/\text{day}$ (38), the tolerable daily intake of Pb would be 210 μg , however, due to the elevated Pb concentration in workshop dust, the daily intake of an adult recycling worker would be 10.5 mg, an amount indicating a high risk of adverse health effects. Health risk due to oral intake of Pb for the general public at Street B-1 was also considered high, but lower than at the workshops by 5-fold. When considering the contribution from the seven metals, Pb would contribute to 99, 97, 89, and 92% of the risk at the workshop, Street B-1, Street B-2, and the food market, respectively, whereas at the plastics processing area (Street L-1, L-2) and the non e-waste control sites, Pb would contribute to 45 and 56–60%. Except for Pb, the risks to adults contributed by the metals were minimal ($\text{HQ} \leq 1$). Copper contributed to the second highest HQs ($\text{HQ}_{\text{Cu}} < 0.35$).

In comparison to adults, the potential health risk to children at all locations was eight times greater. This was partly attributed to the higher ingestion rate used (200 mg/kg/day) in estimating the risk and the smaller body size. Risk due to Pb would be a grave concern for workshop ($\text{HQ}_{\text{Pb}} = 402$) and Street B-1 ($\text{HQ}_{\text{Pb}} = 82.6$) scenarios. Huo et al. indicated that children of circuit board recycling workers contained higher BLLs (17). Since children sometimes accompany their parents at the workshops, they can become exposed to metal-laden dust. Copper was found to be a pollutant of major concern ($\text{HQ}_{\text{Cu}} > 1$) at workshop and Street B-1. Overall, the accumulative risks due to the seven metals are a major concern ($\text{HI} > 1$) at all locations except for Street G and SU.

If the risk of potential adverse health effects were calculated using the maximum heavy metal concentrations measured in the dust samples, HQ_{Pb} would increase to 94.1 and 60.9 for the adult scenario at the workshop and Street B-1, and Pb at the food market would become a concern ($\text{HQ}_{\text{Pb}} = 1.6$). Furthermore, Pb at all locations, except SU, would be a concern for the child scenario since HQ_{Pb} would increase by approximately 2–7 times resulting in high risk. Nickel at the workshop would also be a concern ($\text{HQ}_{\text{Ni}} = 5.1$), and the risk of health effects due to Cu would increase by 4.3 and 6.6 times at the workshop and Street B-1, respectively.

The model used in this investigation provides a useful tool for risk assessment in identifying the relative human health risks of the heavy metals in dust at different locations, however, there are inherent uncertainties. These include

actual exposure duration, ingestion rate, and heavy metal concentrations in the dust owing to its heterogeneous nature. This study has only considered exposure to heavy metals through dust ingestion, however, risk due to consumption of contaminated food and water may also contribute to the ADD. In order to further improve and strengthen this risk assessment, the bioaccessibility (mobilization of contaminants from the ingested dust into the digestive juice chyme) and oral bioavailability (contaminant fraction that reaches the systemic circulation) of the heavy metals in dust could be investigated. Bioaccessibility could be assessed using *in vitro* digestion models that simulate the human gastrointestinal tract (i.e., stomach pH, food in stomach, intestinal absorption) (39).

This study shows that the crude recycling of printed circuit boards in Guiyu is a significant contributor of heavy metals in dust to the local environment. Roads and premises of nearby public facilities such as a schoolyard and outdoor food market have been shown to be adversely impacted by the recycling activity. Heavy metal concentrations, especially Pb and Cu, in workshop and road dusts were found to be severely enriched, posing potential health risks, especially to children. To the best of our knowledge, this study is the only human health risk assessment study conducted concerning dust exposure at an uncontrolled e-waste recycling site. It is hoped that the results can serve as a case study for similar e-waste activities in countries such as Africa, India, and Vietnam where e-waste is becoming a growing problem, so that the same mistakes could be prevented.

Acknowledgments

We thank colleagues and students of the Croucher Institute for Environmental Sciences, and Department of Biology, Hong Kong Baptist University for field and technical assistance. Financial support was sponsored by The Research Grants Council of the University Grants Committee of Hong Kong (Central Allocation Group Research Project HKBU 1/03C), Match Fund from Hong Kong Baptist University, and a private donation.

Supporting Information Available

Tables S1, S2, and S3. This information is available free of charge via the Internet at <http://pubs.acs.org>.

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ES071873X