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Assessment of health risk of trace metal pollution in surface soil and road dust from e-waste recycling area in China

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Abstract

Informal recycling of e-waste and the resulting heavy metal pollution has become a serious burden on the ecosystem in Guiyu, China. In this investigation, we evaluated the trace metals concentration of community soil and road dust samples from 11 locations in Guiyu and 5 locations (consists of residential areas, kindergarten/school and farm field) in a reference area using graphite furnace atomic absorption spectrophotometer. The study spanned four seasons, 2012–2013, with a view to assess the risk associated with e-waste recycling in the study area. The concentration of Pb, Cd, Cr and Mn were 448.73, 0.71, 63.90 and 806.54 mg/kg in Guiyu soil and 589.74, 1.94, 69.71 and 693.74 mg/kg, in the dust, respectively. Pb and Cd values were significantly higher ($P \leq 0.05$) than the reference area and the mixed model analysis with repeated seasonal measurements revealed soil Pb and Cd levels that were 2.32 and 4.34 times, while the ratios for dust sample were 4.10 and 3.18 times higher than the reference area. Contamination factor, degree of contamination and pollution load index indicated that all sampling points had high level of metal contamination except farm land and kindergarten compound. The cumulative hazard index of Pb, Cd, Cr and Mn for children in exposed area was 0.99 and 1.62 for soil and dust respectively, suggesting non-cancer health risk potential. The significant accumulation of trace metals in the e-waste recycling area predisposes human life, especially children, to a potentially serious health risk.

Keywords

E-waste; trace metals; soil; dust; health risk; children

1. Introduction

Pollution of the natural environment by heavy metals is a worldwide problem because these metals are indestructible and most of them have toxic effects on living organisms when they

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exceed certain concentrations (Ghrefat et al., 2006). The main anthropogenic sources of heavy metals include various industrial points e.g. present and former mining sites, foundries, smelters and others such as piping, constituents of products, combustion by-products, traffic, industrial and human activities (Al-Khashman, 2004). Pollutants in surface soils can percolate into deep soil and be transported to water bodies and they may be accumulated by plants or become airborne (Chen et al., 1997). Reports have revealed that heavy metals not only decrease the productivities and qualities of crops, but also threaten the safety of ecosystem and human health (Raghunath et al., 1999). Wu et al. (2015) reported that the use of pond water near abandoned e-waste site for irrigation in Longtang, south China, resulted in considerable heavy metal contamination in the paddy soil.

Trace metals have direct influence on public health as they can easily enter the human bodies by dust ingestion, dermal contact or inhalation (Abrahams, 2002) and via the ingestion of heavy metal-polluted vegetables and crops (Zheng J et al., 2013). In particular, the ingestion of dust and soil has been widely regarded as one of the key pathways by which children are exposed to heavy metals and metalloids from paint, leaded gasoline, traffic exhaust and local industrial processes (Rasmussen et al., 2001). Recent study that focused on ambient air heavy metals in PM2.5 from informal electronic-waste recycling sites in China revealed higher level of Pb and Cd compared with reference area and possibility of their effects on the health of local residents, especially children (Zheng et al., 2015; Zhang et al. 2016 Cao et al 2016). Trace metal pollutants such as cadmium (Cd), chromium (Cr) and lead (Pb) have cumulative effects, causing growth retardation in children, kidney disease, cancer and many other adverse health effects (Xu et al., 2012; Xu et al., 2015, Zeng et al. 2016).

Electronic-waste (e-waste) recycling in developing countries is one of the sources of toxic trace metals because numerous metals are used in the production of electrical and electronic devices. The production of these devices is the fastest-growing sector of the manufacturing industry in industrialized countries. The United Nations Environment Programme, (UNEP) (2005) estimated that, 50–80% of the e-waste collected for recycling in industrialized countries end up in informal recycling centers in China, India, Pakistan, Vietnam and the Philippines. The number of electronic devices used per capita at the global scale is growing at a rate of about (4%) and will continue to increase as it is becoming the fastest waste stream world-wide (UNEP, 2005). E-waste recycling and disposal processes, particularly in the informal sector, can release a wide range of toxic substances such as trace metals (lead, cadmium, chromium and manganese) and organic pollutants [(polychlorinated dibenzo-p-dioxins (PCDDs) and furans (PCDFs), polybrominated diphenyl ethers (PBDEs) and polycyclic aromatic hydrocarbons (PAHs)], and therefore cause environmental damage and threaten public health (Leung et al., 2008; Xu et al., 2013; Xu et al 2013b; Yang et al. 2013). While over 1,000 different chemicals can be found in e-waste with some being toxic, metals are thought to comprise over 60.2 % of materials used in electronics and electrical equipment (Widmer et al., 2005). Thus, direct and/or indirect exposures of human to these chemicals and their toxic effects are of great concern.

Guiyu, a community in Shantou, China is one of the largest e-waste recycling and dismantling areas in the world with approximately 1.7 million tons of e-waste dismantled

annually in this region (Bi et al., 2007; Wu et al., 2012). It is located within a total area of 52 km^2 and has a population of 150,000 (Xu et al., 2013c; Liu et al., 2014) with nearly 160,000 migrant workers (Zheng et al. 2016). Guiyu accommodates millions of tons of e-waste from domestic and overseas sources each year. Over 6000 small scale family-run workshops are involved in the business of recycling or dismantling. Guiyu e-waste workers work under conditions of minimal or no protective measures resulting in exposure to toxic pollutants. Previous studies have revealed some health effects associated with trace metals from informal e-waste recycling (Chen et al., 2011; Xu et al., 2012; Yang et al. 2013; Zheng et al., 2013). A number of soil evaluations have been carried out on Guiyu to determine the extent of trace metal pollution but usually on the samples collected directly from e-waste processing site and such studies involved one time sampling (Alabi et al., 2012; Leung et al., 2008; Zheng J et al., 2013). This could not fully describe the extent of pollution in the entire town. Further more, potential impact of e-waste recycling on environmental matrices and inhabitants has not been fully explored. The current study sought to evaluate the extent of trace metals (lead, cadmium, chromium and manganese) pollution in soil and road dust matrices, collected from strategic places including; kindergarten, market places, recreational park and residential areas repeatedly over a year (4 seasons), using contamination indices. This was performed with a view to assess the potential health risk of such exposure to soil and road dust matrices in children and adults, and provide suggestions to stem the unfolding trend of e-waste toxicity in human community.

2. Materials and Methods

2.1. Sample collection and preparation

Soil and road dust were collected from 11 points in Guiyu (exposed area) and 5 points in Haojiang (Reference area) with the aids of Global Positioning System (Figure 1). Samples were not collected directly from e-waste recycling site, rather residential areas, garden/park and school/kindergarten to evaluate the impact of recycling activities on the environmental matrices and inhabitants of Guiyu. Description of the sampling of the sampling points is shown in Table S1 The method of repeated sampling once in a season over a period of four seasons (spring, March 2012 to May 2012; summer, June 2012 to August 2012; autumn, September 2012 to November 2012; winter, December 2012 to February 2013) was employed. About 300 g soil sample was collected from bare soil surface within 60 cm^2 area of each collection point. Stones and other non-soil materials were excluded before being packed manually into zip lock bag with hand trowel (Fujimori et al., 2012) and then coded for identification. Road side dust at the selected areas was collected from a marked area of 30 × 120 cm. Pre-cleaned brush was used for gathering the dust and then packed into zip lock bag. This was followed by collection with vacuum cleaner (Black & Decker Vacuum Cleaner, FHV1200 Flex) adapted for road dust sampling and the content added to the previous collection. In our laboratory, the collected soil and dust were air dried for approximately 5 days, sieved in 2mm mesh size and then kept at −20 °C.

2.2. Digestion of samples and trace metal analysis

Four milliliter (4ml) of Aqua regia (HCl: HNO_3 = 3:1) was added to the solid sample (0.2g soil/road dust) in a Teflon vessel, placed in fume chamber and allowed to stay overnight.

The vessels were placed in a closed microwave oven (PreKem WX-4000, Shanghai Yiyao, China) and heated under pressure, following a four-step procedure viz. Step 1 (100°C, 5 atm, 1 min), step 2 (130 °C, 10 atm, 1 min); step 3 (160 °C, 12 atm, 4 min) and step 4 (190 °C, 20 atm, 5 min). After microwave digestion, the solutions were filtered into 50 mL capacity beaker using Whatman filter paper (No 42), and brought to volume with Milli-Q water (Navarro et al., 2006). The solutions were kept in polyethylene vials at 4°C until analysis.

The total concentrations of Pb, Cd, Cr and Mn in the filtrate were determined by graphite furnace atomic absorption spectrophotometry (Jena Zenit 650, Germany), which consists of an autosampler (MPE60) with an injection volume set at 10μL. The main parameters used for lead determination were a wavelength of 283.3 nm, a lamp current of 4.0 mA, a slit width of 0.8 nm, ashing at 650 °C and atomization at 1700 °C. The parameters for cadmium analysis were a wavelength of 228.8 nm, a lamp current of 2.0 mA, a slit width of 1.2 nm, ashing at 300 °C and atomization at 1300 °C. Evaluation of chromium was through a wavelength of 357.9 nm, a lamp current of 4.0 mA, a slit width of 0.8 nm, ashing at 1000 °C and atomization at 2400 °C while that of manganese was a wavelength of 279.5 nm, a lamp current of 5.0 mA, a slit width of 0.2 nm, ashing at 850 °C and atomization at 1900 °C. For quality assurance and control, solvents and chemicals used for analyses were of analytical grade. To remove contamination, all glassware used in the measurement of metals were soaked in 10% (v/v) HNO_3 for at least 24 h, rinsed with deionized water and then oven dried at 250°C. Blank and procedural control was included with every set of 6 samples. The results obtained from digested filtrates were blank corrected to eliminate background contamination. The analytical precision measured as relative standard deviation was routinely less than 5%.

2.3. Assessment of trace metal contamination levels

Contamination levels of trace metals in soil and road dusts were assessed using contamination indices including; contamination factors (*Cf*), the degree of contamination (*Cd*) and the pollution load index (PLI). *Cf* and *Cd* were calculated as suggested by Hakanson (1980) through equations 1 and 2, respectively.

$$Cf = \mathrm{Cs}/\mathrm{Cb} \quad (1)$$

$$Cd = \sum Cf \quad (2)$$

Where Cs is the measured concentration of the detected metal in either of the soil or road dust, and Cb is the background value of trace metals in the uncontaminated soil (Khairy et al. 2011). Hakanson (1980) classification of *Cf* was used to evaluate metal contamination level as shown in Table S2 (supplementary information). If the *Cd* value exceeds 20, then it is necessary to take immediate counter measures to reduce heavy metal contamination in the soil. Furthermore, each sampling area from the regions was evaluated in order to determine

the extent of trace metal pollution by employing pollution load index (PLI) by Thomilson *et al.* (1980) as follows:

$$\mathrm{PLI}=(Cf_1 \times Cf_2 \times Cf_3 \times Cf_4 \ldots . Cf_n)^{1/n} \quad (3)$$

Where n is the number of metals studied and *Cf* is the contamination factor calculated as described in Equation 1. The PLI provides comparative means for assessing an area quality. The rank of values of PLI and its implication is shown in Table S2.

2.4. Health Risk assessments on human via ingestion, dermal contact and inhalation

The average daily dose (ADD) (mg/kg/day) of a pollutant that can be contacted via soil/road dust through ingestion (ADD ingest), absorbed through the skin (ADD dermal) and inhalation (ADD inhale) as pathways can be calculated using equations 4, 5 and 6, respectively (USEPA, 1997):

$$ADD\ ingest = \frac{C \times ingestR \times EF \times ED}{BW \times AT} \times CF \quad (4)$$

$$ADD\ dermal = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times CF \quad (5)$$

$$ADD\ inhale = \frac{C \times inhaleR \times EF \times ED}{PEF \times BW \times AT} \quad (6)$$

where C is the soil or dust concentration of the contaminant (mg/kg), Ingest R is the Ingestion rate (mg/day), EF is the Exposure frequency (days/year), ED is the Exposure duration (years), BW is the Average body weight (kg), AT is the Average time (days), CF is the Conversion factor (1×10^{-6} kg/mg), SA is the Surface area of the skin that contacts the soil (cm^2), AF is the soil Skin adherence factor for soil (mg/cm^2), ABS is the Dermal absorption factor (chemical specific), InhaleR is the Inhalation rate (m^3/day) and PEF is the Particle emission factor =$1.36\times10^9 m^3$/kg

For ingestion, intake rate (IngestR) of soil/dust was 100 mg/day for adult while it was 200 mg/day for children (Calabrese et al., 1987; USEPA 1997). The inhalation intake rate (InhaleR) for both adults and children was based on 20 m^3/day (USEPA, 1997). The exposure duration (ED) of 6 years and 24 years were used for children and adults, respectively, while exposure frequency (EF) was assumed to be 350 days/year for inhabitants of both exposed and reference areas, and lifetime exposure duration of 70 years was used in our assessment (Leung et al., 2007). A body weight of 16 kg and 70 kg were used for children and adults, respectively (Zheng J et al., 2013). Average time (AT) was obtained from ED x 350 day, which is equivalent to 2190 days for children and 8400 days for adult, while 25550 days was used for permanent cancer exposure risk. The value, 1×10^{-6} kg/mg, was used as Conversion factor (CF), while values used for adult and children Surface

area of the skin that contacts the soil/dust (SA) were 3300 cm^2 and 2800 cm^2, respectively. Skin adherence factor of soil (AF soil/dust) for adult workers and children used was 0.2 mg/cm^2 each; while dermal absorption factor (ABS) used for all trace metals evaluated was 0.001 (Chang et al., 2009). Particle emission factor (PEF) was 1.36×10^9 m^3/kg (USEPA 1997). A hazard quotient (HQ) for non-cancer risk was calculated to make the comparison with the health guideline and was determined by the relation;

$$HQ=ADD/RfD \quad (7)$$

The Reference Dose (RfD) in this equation 7 is the estimates of daily exposure below which adverse non-cancer health effects are unlikely. If the HQ < 1, then noncancerous effects are unlikely. If the HQ ≥1, then adverse health effects might be possible or if the ADD value is higher than the RfD, it is likely that the factor in question will cause adverse human health effect (USEPA 1993). The calculation used in the present study was based on the Risk Assessment Guidance for Super-fund, vol. I: Human Health Evaluation Manual (USEPA 1989). The values HQ were also added to generate a Hazard Index (HI) to estimate the risk of mixed contaminants (USEPA 1989). HI refers to the "sum of more than one Hazard Quotient for multiple substances and/or multiple exposure pathways (Man et al., 2010). The guidelines also stated that "any single chemical with an exposure level greater than the toxicity value will cause the Hazard Index to exceed unity and for multiple chemical exposures the Hazard Index can also exceed unity, even if no single chemical exposure exceeds its RfD". Thus, Hazard Index (HI) for non-cancer risk for each of the trace metals was obtained as;

$$\text{Hazard Index (HI)}=\text{HQingestion}+\text{HQdermal}+\text{HQinhale} \quad (8)$$

While the risk of mixed contaminants was expressed as cumulative HI for the metals viz;

$$\text{mHI}=\sum_{k=0}^{n}(\text{HI}) \quad (9)$$

As a result of non-availability of slope factors for carcinogenic risk through ingestion and dermal contact, this study only considered carcinogenic risk resulting from inhalation. For carcinogens, the lifetime average daily dose (LADD) was multiplied by the corresponding slope factor to produce an estimate of cancer risk (USEPA 1997; Baptista and De Miguel, 2005). Risk management decisions were most frequently made when the cancer risk ranges were 10^{-6} to 10^{-4} (USEPA 1997). The RFD and slope factors (for carcinogenic metals) of trace metal evaluated are shown in supplementary information (Table S3).

$$LADD=\frac{(C\times EF)}{AT\times PEF}\times\left(\frac{inhR\ child\times ED\ child}{BW\ child}+\frac{inhR\ adult\times ED\ adult}{BW\ adult}\right) \quad (10)$$

2.5. Statistical analysis

All statistical analyses were performed with SPSS (version 13, SPSS, Chicago, IL, USA). Data were tested for goodness of fit to a normal distribution with Kolmogorov – Smirnov's one-sample test. Since all the data were found to deviate from the normal distribution, the data were $\log_{10}$-transformed prior to performing student's t tests to compare the trace metal seasonal as well as overall geometric mean values between the exposed and the Reference areas. We used linear mixed models with consideration of repeated measurements in 4 seasons to calculate the overall difference in $\log_{10}$-transformed metal concentrations between the exposed and the Reference areas. Using this difference as the exponent on a base of 10, the product of the exponential function can be interpreted as a ratio of the exposed area metal concentrations to the Reference area metal concentrations.. The probability value of $P < 0.05$ was set as the level for statistical significance. All other calculation as indicated by equation were based on referenced standards.

3. Results and Discussion

3.1. Trace metal concentrations in soil and road dust

The concentrations of Pb in soil and road dust obtained from different sampling points and their average for different seasons in the e-waste exposed area and reference area are respectively shown in figures 2a and b.

The values of Pb obtained in soil for all seasons were highest in area at the back of e-waste market (ABEM) with autumn values of 4636 mg/Kg. The average mean value of soil Pb for each of the seasons in exposed area was significantly higher than the corresponding value in reference area ($P<0.001$). The distribution of road dust Pb was almost similar to that of soil Pb, except that of farm field. The Pb concentrations in road dust in most sampling points were not as high as values obtained for soil except at the farm field. Average values obtained for road dust Pb for all seasons were significantly higher compared to the values obtained for reference area (Figure 2b). ABEM as well as residential areas 2, 3 and 5 have soil Pb values above the China Grade III value (SEPA 1995) of 500 mg/kg for some of the seasons (Figure 2a) while road dust Pb was above 500g for all seasons at ABEM and at different seasons for farm field and some residential areas (Figure 2b).

The distribution of soil and road dust Cd show variations among different sampling points as shown in Figures 3a and 3b. The mean values of Cd in both soil and road dust of the exposed area showed significant difference in all seasons compared with reference area with highest value obtained in summer and spring for soil and dust, respectively.

The mean Cr value of 965.9mg/Kg obtained for soil in autumn at residential area 5 was the highest in all sampling points of e-waste exposed and reference areas (Figure 4a). Although there were variations in average soil Cr, there were no significant differences between the corresponding seasons when E-waste exposed area was compared with the reference area ($P> 0.05$) except in autumn. The average value of road dust Cr was significantly higher in reference area than the exposed area in spring and summer while there was no significant difference in autumn and winter. High value of Cr in road dust was observed especially for residential areas 9 and 10 in Reference area (Figure 4b).

The soil Mn values show variations between exposed and reference areas (Figure 5a). Highest value of soil Mn was obtained in winter at area in front of e-waste market. High values of Mn were also obtained for residential area 10 in Reference area for all seasons compared to other sampling points in this zone. Although average values of Soil Mn were higher in e-waste exposed area for summer and winter, there were no significant differences for all the seasons when compared with reference area. For road dust Mn (Figure 5b), summer value in e-waste exposed area was significant compared to the reference area while in winter, value obtained for reference area was significantly higher.

The variations observed in trace metal concentrations revealed the highest values in autumn, except for Mn in soil (winter) and Cd in dust (spring), while lowest values were obtained in spring in most cases. These respectively correspond with high and low period of activities in the exposed area. Spring is characterized with long period of vacation when there is lesser to no activities for longer period of time. Highest values of the trace metals in autumn may be attributed to more e-waste recycling in this period. Similar to our study, Cr was found to be the most enriched metals in PM2.5 from August to September (Fall/autumn period) in Guiyu and this was attributed to recycling of more e-waste items containing Cr during the period (Deng et al., 2006). The high values of Mn and Cr in the reference area might be due to extensive use of coal by thermal power plant in this region. Similar observation was reported for Cr and Mn in ambient $PM_{2.5}$ of Guiyu and Haojiang as reference area (Zheng et al 2016).

Table 1 shows a mixed model accounting for repeated measurements at different seasons. The soil Pb and Cd levels were 2.32 and 4.34 times respectively higher in the exposed area compared with the reference area. For dust metal levels, Pb and Cd values were respectively 4.10 and 3.18 times higher in the exposed area compared with the reference area. The mean values of metals obtained in soil and road dust were comparable with report of other studies despite the fact that our samples were neither collected from e-waste site nor one time sampling as shown in Table 2. Pb, Cr and Mn values obtained from soil in the study area were lower compared to soil sample from an e-waste site in Guiyu as reported by Li et al. (2011) and Alabi et al. (2012). However, our values were higher than those reported for an abandoned e-waste area in Guiyu (Li et al., 2011) as well as values reported for e-waste recycling area in Taizhou, China (Zhou et al., 2012) and Manila, Philippines (Fujimori et al., 2012; Fujimori and Takigami, 2013). Trace metal concentrations from exposed area in this study were higher than metal concentration permissible level of State Environmental Protection Administration of China, SEPA, (1995) reported for Grade I (natural background) and Grade II (agricultural and related use), except for Cr. The trace metal levels in road dust obtained in exposed area in this study were lower compared to indoor and outdoor dust in Guiyu (Zheng J 2013). However, the Pb and Cd levels in our study were even higher than the Grade III (industrial activity) trace metal concentrations in China. Elevation of trace metals (Pb, and Cd) in the soils and road dust in Guiyu as obtained in the current study might have resulted from e-waste recycling as observed. Reports have shown that Pb levels in soil appear to be readily affected by anthropogenic factors (Gray et al., 2003; Martin, 2001). Higher concentration of trace metal in strategic places like residential areas, kindergarten area and farm land as observed in this study indicates that residents of Guiyu might have higher exposure risk.

3.2. Correlation analysis of trace metals from soil and road dust

Table 3 revealed positive correlations among trace metals evaluated in soil with significant difference at $P<0.001$. The correlation among metals in the road dust showed deviation from that of soil as there was no significant association between Cr and Mn, while negative significant association exists between Cd and Cr as well as Cd and Mn. This indicates high possibilities that trace metals from soil have common source(s) i.e. e-waste, while in road dust, Cr and Mn might not have common source compared to Pb and Cd. The contribution of Cr and Mn from vehicle tire and brake wear to road dust metal concentrations is not ruled out; while there can also be further contribution from traffic emission. Similar observation was reported for trace metals evaluated from urban road dust (Wei et al., 2009; Kong et al., 2012). Our inability to separate the trace metal contribution from vehicles from that of the e-waste in road dust is regarded as a limitation of this study.

3.3. Extent of pollution in soil and road dust from Guiyu as revealed by contamination indices

Pollution load index, contamination factors and degree of contamination are indices used to determine the level of pollution of different matrices including road dust and soil samples by different authors (Chen et al., 2005; Yekeen and Onifade, 2012). Table 4 presents soil contamination factor values which showed variations among the sampling points in the exposed area. Highest level of contamination was observed at area back of e-waste market with contamination values of Pb and Cd > 6. The degree of contamination could be used to group sampling points into: low (farm field and kindergarten area), moderate (Front of E-waste market, residential Area 1, residential area 4, residential area 6 and back of kindergarten), considerable (Residential area 2 and 3) and very high (back of e-waste market and residential area 5) based on average metal concentrations for the four seasons (Table 4). The road dust samples also revealed variations in values of contamination factor and degree of contamination. Similar to soil, area back of e-waste market still had the highest values. (Table 4). Further evaluation of the matrices with pollution load index showed that all soil and dust collection points had value above unity, except farm field and kindergarten area for soil while area front of e-waste market and residential area 3 for road dust. On average, based on high concentration of trace metals, a high value of degree of contamination and pollution load index were obtained for soil and road dust matrices in the exposed area and was regarded as being polluted.

3.4. Health risk assessment of exposure to trace metals from soil and road dust in e-waste recycling town

Human health risk assessments were evaluated for non-cancer risk in children and adults that might result from exposure to trace metals in soil and road dust in exposed area and the reference area through the combined pathways of ingestion, dermal contact, and inhalation while life time cancer risk was evaluated only via inhalation. The hazard quotient values for ingestion, dermal and inhalation of soil and road dust for all trace metals evaluated from average daily doses (Table S4) and references doses (Table S3) for non-cancer risk in children were higher in exposed area compared with the reference area, except for Cr only in road dust (Table 5). Pb had the highest hazard index for children with the values in exposed

area of 2.33 and 4.13 folds higher than that of the reference area for soil and dust respectively, while the values were 2.32 and 4.11 folds in the case of adults. The estimated cumulative hazard index of trace metals in children was 0.99 for soil, while the values of 1.62 obtained for road dust exceeded the unitary permissible limit (USEPA, 1997). The trends of estimated hazard quotient and hazard index for adults were similar to that of the children but generally of lower values (Table 5). However, Pb still had the highest hazard quotient and hazard index (>0.1) in both soil and road dust.

The hazard quotient obtained for ingestion for each of the metals was the highest among the routes of exposure for both matrices in the exposed and the reference areas for children and adults, except for Cr (Table 5). Similar trend of high hazard quotient had been reported for soil trace metals in India (Chabukdhara et al., 2013) and China (Yang Y et al., 2013). The hazard quotient we obtained for Cd and Cr were less than unity, which suggests non-cancerous risks. However, the mixed contaminants hazard index values of 0.99 and 1.62 obtained for soil and dust risk estimate respectively showed that children of exposed area might have health risk. Pb had highest percentage contribution in the exposed area probably because of its high level in different e-waste components. This can be corroborated by the study of De Miguel et al. (2007) and Kong et al. (2012), where it was reported that hazard index values above 0.1 would trigger adverse health effects in children. Our results were consistent with the studies on the dust collected in the waste printed circuit board recycling workshop (Leung et al., 2008; Xue et al., 2012).

Cancer risk as estimated for the Cd and Cr trace metals in both soil and road dust via inhalation showed higher health risk in exposed area compared with the reference area, except Cr in road dust (Figure 6). However, these values(Cd and Cr) as well as their cumulative cancer risk value for the two metals metal in both matrices (Figure 6) were lower than the limit of $10^{-6} – 10^{-4}$ (USEPA, 1997). This indicates that carcinogenic risks are relatively unlikely. Our assessment of non-cancer and cancer health risk using total trace metal concentrations from soils and road dust might have probably overestimated or under estimated the actual health risk compared to the use of bioavailable trace metal concentrations (Oomen et al., 2002). Nevertheless, the results of the present investigation provide some valuable information for concerned authority to look inward on the trace metal pollution status of the exposed area.

4. Conclusion

Informal processing of electronic wastes results in release of trace metals which pollute soil and dust not only in the factory vicinity but the community as a whole. The trace metal concentrations (Pb and Cd) in soil and road dust from exposed area as observed in this study were significantly higher compared with the reference area. On the basis of the Environmental Quality Standards for Soils (GB 15618-1995) in China, the concentrations of Pb and Cd were observed to be above the Grade I and Grade II values but below the permissible limit of Grade III. The Pb and Cd in road dust were above Grade III limit. The values obtained for contamination factor, degree of contamination and pollution load indices indicate that soil and road dust from exposed area of Guiyu are polluted and as a result, measures to reduce the pollution resulting from informal e-waste recycling should be further

strengthened. The level of pollution reflected in estimated hazard index values of trace metal contaminants for non-cancer risk which is above the permissible limit in road dust for children suggests potential health risk. Human population can be exposed to trace metals from road dust and soil simultaneously, which may result to a higher level of health risk. The accumulation of trace metals in the recycling site is significant and may increase body burden especially in sensitive populations such as pregnant women and children; and thus constitute a potentially serious health risk.

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Fig. 1.
Soil and dust sampling points in E-waste recycling town (Guiyu) and reference area (Haojiang)

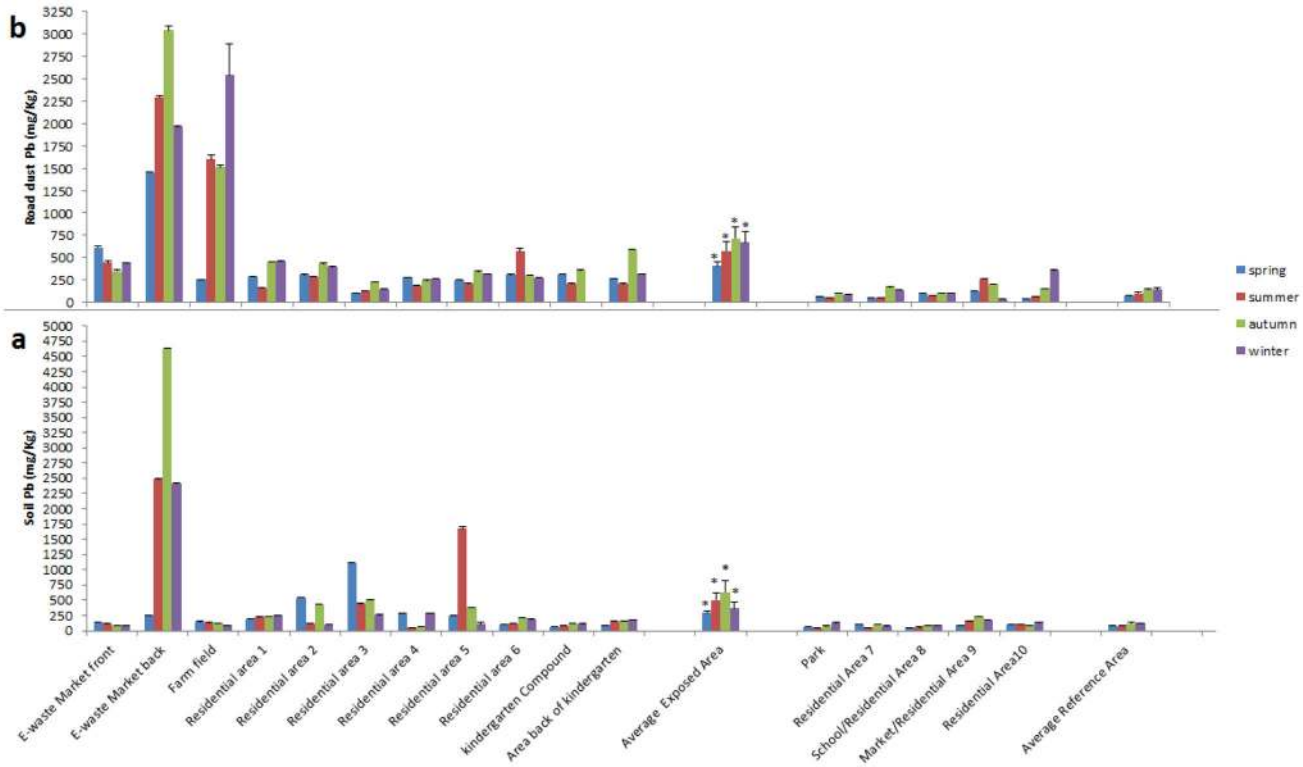

Fig. 2.

Soil (a) and road dust (b) Pb levels for different sampling points in e-waste exposed and reference areas for four seasons

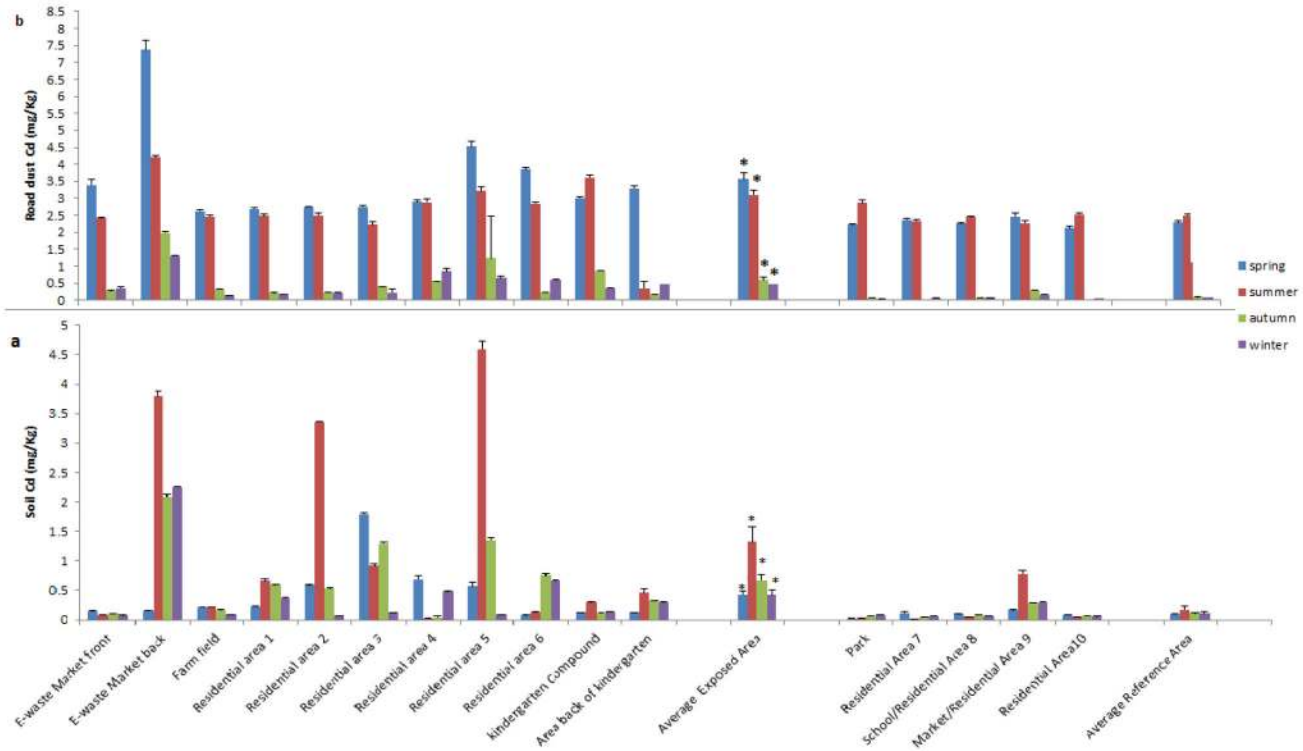

Fig. 3.

Soil (a) and road dust (b)Cd levels for different sampling points in e-waste exposed and reference areas for four seasons

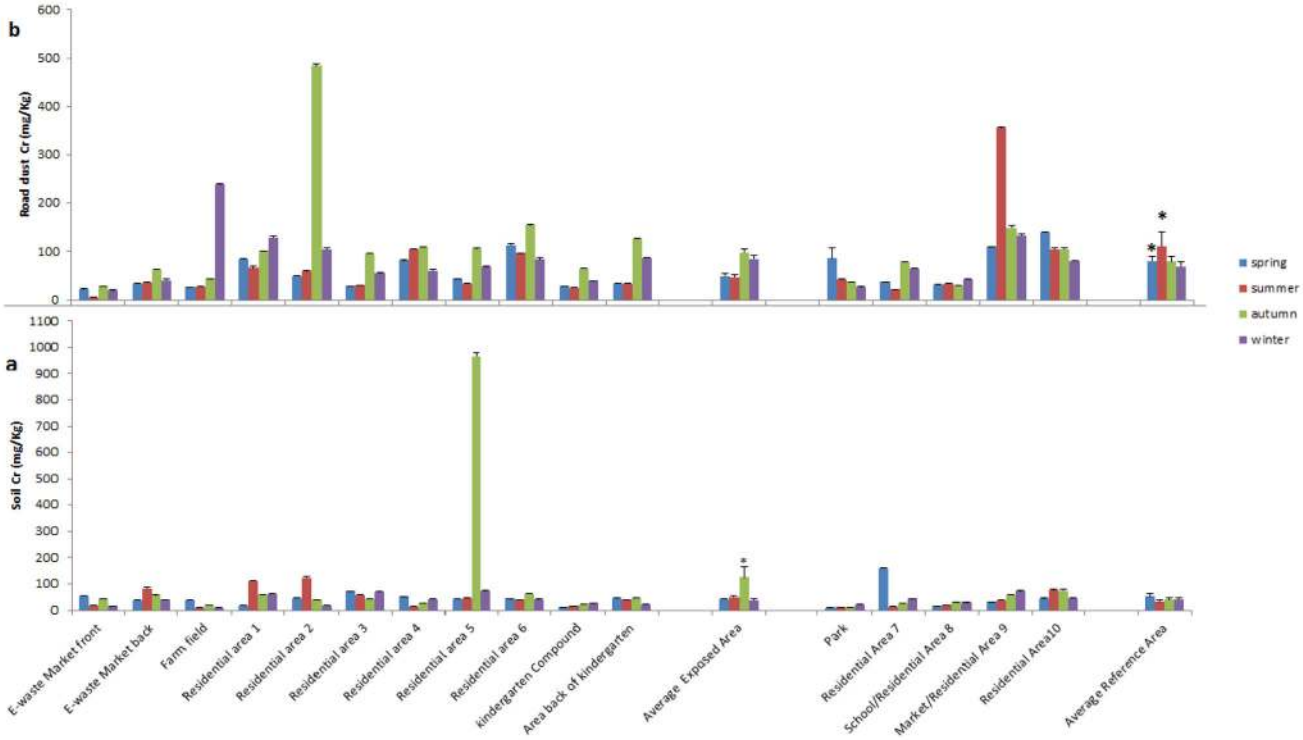

Fig. 4.

Soil (a) and road dust (b) Cr levels for different sampling points in e-waste exposed and reference areas for four seasons

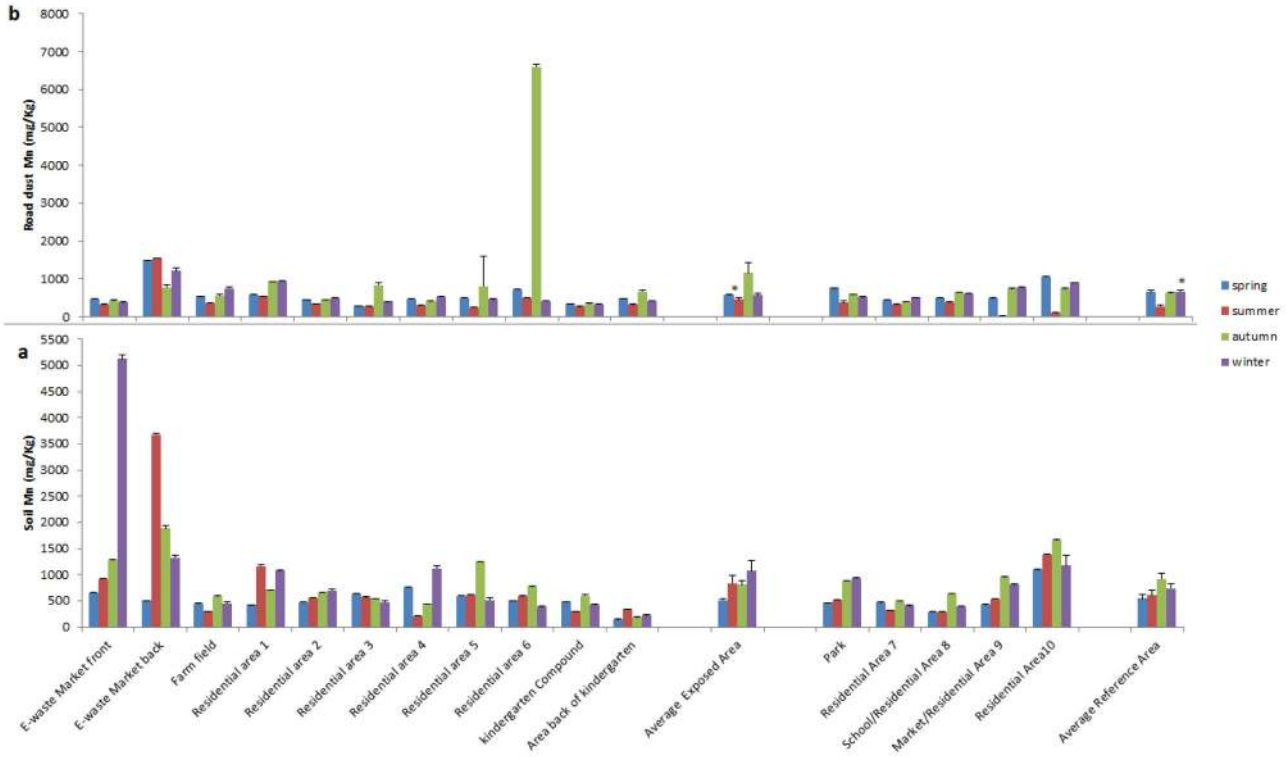

Fig. 5.

Soil (a) and road dust (b) Mn levels for different sampling points in e-waste exposed and reference areas for four seasons

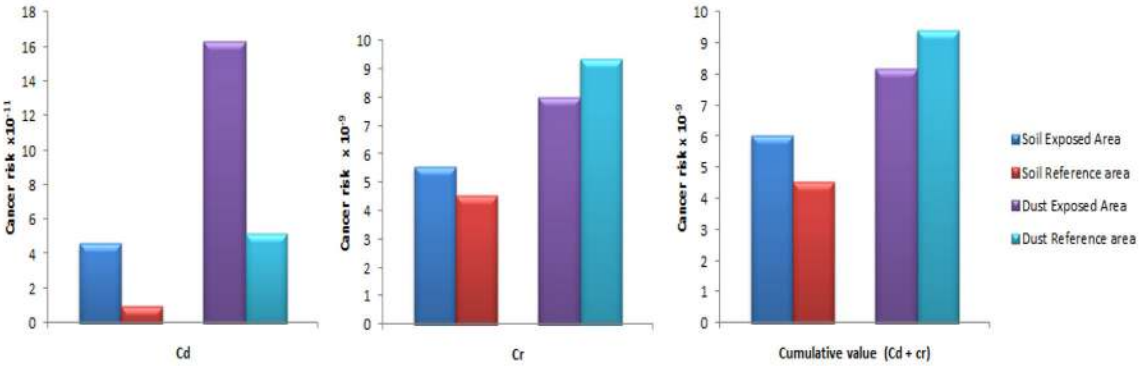

Fig. 6.

Cancer risks due to exposure to trace metals in soil and road dust in e-waste exposed and reference areas

Assessment was based on geometric mean; All values were lower than the range (10^{-6} – 10^{-4})

Table 1

Mixed model analysis comparing soil and road dust metal concentrations between e-waste exposed area and reference area using repeated measurements in all four seasons

Matrice	Metal	Geometric mean of metal concentrations		Ratio of exposed/reference area metal concentrations		P
		Exposed area (mg/kg, n=44)	Reference area (mg/kg, n=20)	Estimate	95% CI	
Soil	Pb	213.61	91.92	2.32	1.42, 3.80	0.0011
	Cd	0.32	0.07	4.34	2.27, 8.31	<0.0001
	Cr	38.56	31.76	1.21	0.79, 1.87	0.3718
	Mn	606.63	614.98	0.99	0.70, 1.39	0.9366
Road dust	Pb	392.58	95.72	4.10	2.74, 6.13	<0.0001
	Cd	1.14	0.36	3.18	2.11, 4.79	<0.0001
	Cr	55.46	64.93	0.85	0.58, 1.25	0.4092
	Mn	529.35	493.26	1.07	0.81, 1.42	0.6174

Table 2

Comparison of soil and road dust trace metal concentrations in current study with previous studies

Place	Pb	Cd	Cr	Mn	Times Samples collected	Unit	Reference
Soil							
Guiyu Environ:							
Mean	448.73*	0.71*	63.90	806.54	Repeated sampling (4 seasons)	mg/kg	This study
Geo mean	213.407	0.319	38.513	606.17			”
Haojiang Environ:							
Mean	101.19	0.12	41.74	704.06	Repeated sampling (4 seasons)	mg/kg	This study
Geo Mean	91.734	0.074	31.730	613.03			”
Manila, Philippines Soil from e-waste (outside)	250	-	-	-	once	μg/g wet	Fujimori and Takigami (2013)
Manila, Philippines Soil e-waste	7.57	0.039	-	-		mg/kg	Fujimori et al. (2012)
Guiyu: Soil from e-waste area	1431	31.96	153.6	374.1	once	mg/kg dw	Alabi et al. (2012)
Burnt plastic dump area Guiyu	104	1.7	28.6	-	once	mg/kg dw	Leung et al. (2006)
Printer roller dump area Guiyu	190	3.1	74.9	-	once	mg/kg dw	Leung et al. (2006)
Taizhou: Soil from e-waste area	124.74	0.90	25.64	-	once	mg/kg dw	Zhou et al. (2012)
Guiyu soil (abandoned area)	150	1.21	2600	300	Once	mg/kg	Li et al. (2011)
Guiyu soil residue from open burning	480	10.02	320	500	Once	mg/kg	Li et al (2011)
*E-waste recycling area in slum in Bangal ore, India (BES)	297	2.33	73	449	Once	μg/g	Ha et al. (2009)
*E-waste recycling facility in Bangalore, India (BEF)	126	0.478	54	619	Once	μg/g	Ha et al. (2009)
Area near the open-burning area of e-waste in Guiyu, China	97.8–123	ND	7.02–155	-		μg/g	Wong et al. (2007)
Area at the open-burning area of e-waste in Guiyu, China	856–7038	5.51–42.9	137–477	-		μg/g	Wong et al.(2007)
Road dust							
Guiyu							
Mean	589.74*	1.94*	69.71	693.74	Repeated sampling (4 seasons)	mg/kg	This study
Geo Mean	391.955	1.134	55.410	528.64			
Haojiang							
Mean	115.61	1.23	85.38	552.48	Repeated sampling (4 seasons)	mg/kg	This study
Geo Mean	95.481	0.356	64.908	490.42			
Around Manila, Philippines	6.44	0.044	-	-	Once		Fujimori et al.(2012)
E-waste - indoor dust	1467	59.0	-	-	Once	mg/kg	Zheng J et al. (2013)

Place	Pb	Cd	Cr	Mn	Times Samples collected	Unit	Reference
E-waste - outdoor dust	4489	120.	-	-	Once	mg/kg	Zheng J et al.(2013)
E-waste PCB baking workshop dust; Guiyu	1910	2.4	67	761	Once	mg/kg	Zhu et al. (2012)
E-waste plastic processing workshop dust; Guiyu	323	1.7	56	800	once	mg/kg	Zhu et al. (2012)
China[a]							SEPA (1995)
Grade I (natural background)	35	0.2	90	-		mg/kg	
Grade II (agricultural and related use)	300	0.3	200	-			
Grade III (industrial activity)	500	1	300	-			
Dutch standard optimum	85	0.8	100	-		mg/kg	VROM (1994)
Action	530	12	380	-			

Reported values for authors are mean except Ha *et al.* (2009) which is geometric mean

*T-test analysis with log transformed values to compare exposed area and reference area (P<0.05; two tailed)

[a]Standard Reference in China

Table 3

Correlation between trace metals in soil and road dust from study area

Matrices	Pb		Cd		Cr		Mn	
Soil								
	R	P	R	P	R	P	R	P
Pb	1		0.834**	<0.001	0.560**	<0.001	0.388**	<0.001
Cd			1		0.488**	<0.001	0.216**	<0.001
Cr					1		0.436**	<0.001
Mn							1	
Road Dust								
Pb	1		0.054	0.390	–0.064	0.308	0.240**	<0.001
Cd			1		–0.322**	<0.001	–0.321**	<0.001
Cr					1		0.368**	<0.001
Mn							1	

** Spearman correlation significant at the 0.01 level (2-tailed)

* Spearman correlation significant at the 0.05 level (2-tailed)

Table 4

Contamination factor, degree of contamination and pollution load index obtained for soil and dust samples from e-waste exposed area

Sampling Point (SP)		Contamination factor				Degree (*Cd*)	PLI
		Pb	Cd	Cr	Mn		
Soil							
1	E-waste Market front	1.025	0.827	0.789	2.834	5.475	1.173
2	E-waste Market back	24.172	17.242	1.296	2.622	45.322	6.135
3	Farm field	1.203	1.398	0.453	0.632	3.686	0.833
4	Residential area 1	2.203	3.853	1.450	1.190	8.696	1.956
5	Residential area 2	2.926	9.500	1.336	0.842	14.604	2.365
6	Residential area 3	5.718	8.608	1.468	0.785	16.579	2.744
7	Residential area 4	1.714	2.575	0.798	0.895	5.982	1.332
8	Residential area 5	5.946	13.759	6.733	1.052	27.490	4.906
9	Residential area 6	1.519	3.375	1.108	0.795	6.797	1.458
10	Kindergarten compound	0.918	1.385	0.445	0.641	3.389	0.776
11	Area back of kindergarten	1.436	2.475	0.915	0.315	5.141	1.006
Overall Exposed area		4.435	5.917	1.531	1.146	13.029	2.605
Road dust							
1	E-waste Market front	4.006	1.310	0.219	0.727	6.262	0.836
2	E-waste Market back	18.952	3.023	0.510	2.272	24.757	2.854
3	Farm field	12.767	1.130	0.983	1.005	15.885	1.326
4	Residential area 1	2.932	1.130	1.115	1.359	6.536	1.497
5	Residential area 2	3.069	1.170	1.167	0.800	6.206	1.353
6	Residential area 3	1.312	1.138	0.611	0.810	3.871	0.927
7	Residential area 4	2.098	1.461	1.038	0.764	5.361	1.249
8	Residential area 5	2.407	1.956	0.738	0.913	6.014	1.335
9	Residential area 6	3.140	1.527	1.312	3.728	9.707	2.201
10	Kindergarten compound	2.468	1.592	0.464	0.580	5.104	1.014
11	Area back of kindergarten	2.962	1.885	0.824	0.855	6.526	1.408
Overall Exposed area		5.101	1.577	0.816	1.256	8.750	1.695

Table 5

Health risk assessment of children exposure to trace metals from soil and road dust from e-waste town compared to reference area

	Children					Adults				
Trace metals	**Pb**	**Cd**	**Cr**	**Mn**	**m HI**	**Pb**	**Cd**	**Cr**	**Mn**	**m HI**
soil exposed area										
HQingest	7.31 E-01	3.82 E-03	1.54 E-02	1.56 E-01		8.71 E-02	4.55 E-04	1.83 E-03	1.86 E-02	
HQdermal	1.36 E-02	1.07 E-03	2.15 E-02	1.09 E-02		3.83 E-03	3.00E-04	6.05 E-03	3.11 E-03	
HQinhalation	5.34 E-05	2.81 E-07	1.36 E-03	3.73 E-02		1.27 E-05	6.69 E-08	2.83 E-04	8.88 E-03	
HI	7.45 E-01	4.89 E-03	3.83 E-02	2.04 E-01	9.92 E-01	9.09 E-02	7.55E-04	8.16E-03	3.06 E-02	1.30 E-01
Soil reference area										
HQingest	3.14 E-01	8.39 E-04	1.26 E-02	1.58 E-01		3.74 E-02	1.00 E-04	1.50 E-03	1.88 E-02	
HQdermal	5.87 E-03	2.35 E-04	1.77 E-02	1.12 E-02		1.65 E-03	6.60 E-05	4.95 E-03	3.14 E-03	
HQinhalation	2.30 E-05	6.17 E-08	9.69 E-04	3.78 E-02		5.48 E-06	1.47 E-08	2.31 E-04	9.02 E-03	
HI	3.20 E-01	1.07 E-03	3.13 E-02	2.07 E-01	5.59E-01	3.91 E-02	1.66 E-04	6.68 E-03	3.10 E-02	7.69E-02
Dust exposed area										
HQingest	1.34 E+00	1.35 E-02	2.21 E-02	1.36 E-01		1.60 E-01	1.61E-03	2.64 E-03	1.62 E-02	
HQdermal	2.51 E-02	3.79 E-03	3.10 E-02	9.62 E-03		7.05 E-03	1.07 E-03	8.70 E-03	2.71E-03	
HQinhalation	9.80 E-05	9.96 E-07	1.71 E-03	3.26 E-02		2.34 E-05	2.37 E-07	4.06 E-04	7.76 E-03	
HI	1.37 E+00	1.73 E-02	5.48 E-02	1.78 E-01	1.62 E+00	1.67 E-01	2.68 E-03	1.17 E-02	2.66 E-02	2.08 E-01
Dust reference area										
HQingest	3.26 E-01	4.32 E-03	2.59 E-02	1.26 E-01		3.89 E-02	5.14 E-04	3.09 E-03	1.50 E-02	
HQdermal	6.10 E-03	1.21 E-03	3.63 E-02	8.97 E-03		1.71 E-03	3.39 E-04	1.02 E-02	2.51 E-03	
HQinhalation	2.39 E-05	3.17 E-7	2.00 E-03	3.02 E-02		5.71 E-06	7.56 E-08	4.76 E-04	7.20 E-03	
HI	3.32 E-01	5.53 E-03	6.42 E-02	1.65 E-01	5.67 E-01	4.06 E-02	8.53 E-04	1.38 E-02	2.47 E-02	8.00 E-02

*HQ: hazard quotient HI: Hazard Index; mHI: mixed contamination hazard index

-Assessment was based on geometric mean