

Spatial Distribution and Human Health Risk Assessment of Mercury in street dust resulting from various land use in Ahvaz, Iran.

Ahad Nazarpour^{1✉}, Navid Ghanavati², Michael J. Watts³

- 1- Department of Geology, Faculty of Science, Azad University, Ahvaz Branch, Ahvaz, Iran
- 2- Department of Soil Science, Faculty of Agricultural Science, Azad University, Ahvaz Branch, Ahvaz, Iran
- 3- Inorganic Geochemistry, Centre for Environmental Geochemistry, British Geological Survey Keyworth UK

Abstract

Mercury as a toxic element and its associated health hazard has been an important topic of research for urban pollution for many years. In this paper, the spatial distribution, pollution assessment, and health risk associated with Hg in roadside dust 96 street dust samples, representing differing land uses have been investigated. Land uses included residential areas (RA), industrial areas (IA), public gardens (PG), roadside areas (RS) and suburban areas (SA) in the city of Ahvaz, Iran were investigated. Compared with other cities, the concentration of Hg in Ahvaz was considerably higher with a mean value of 2.53 mg.kg⁻¹, ranging from 0.02 to 8.75 mg.kg⁻¹. Residential areas exhibited higher Hg in street dust than other areas, as demonstrated by spatial mapping illustrating hot-spots associated with old urban areas with high residential density, high volume traffic of roadside areas, and industrial districts: including oil-drilling activities, steel smelting related industries, and small industrial towns around Ahvaz. However, Hg concentrations in street dust near to the public gardens (PG) and suburban areas (SA) were not at elevated levels compared other land uses investigated in this study. A health risk assessment model of non-carcinogenic effects was evaluated for both children and adults. The HQ values also revealed that the main exposures route for children and adults decreased as follows: vapour>ingestion> dermal contact> inhalation. The Hazard Index (HI) in each area is less than the safe level (HI≤1) for children and adults, but higher for children. The HI value decrease as the following order: RS>IA>RA>SA>PG, which indicates potentially serious health hazards for children in the study areas.

Key words: Mercury, Health assessment, Street dust, Ahvaz, Iran

1. Introduction

Mercury (Hg) as an ever-present potential toxic metal is a considerable potential hazard to the environment and on human health (Sun et al. 2013; Farzin et al. 2008; Fu et al. 2012). Mercury in dust collected from the streets can easily enter the human body and bloodstream during inhalation, dermal contact, and ingestion (Wei et al. 2015). High levels of Hg cause adverse effects on brain, nervous system, gingivitis tremors, liver, and heart muscle (Farzin et al. 2008; Sun et al. 2013). The main origins of Hg in the environment are natural and anthropogenic sources (Chen et al. 2010; Solgi et al. 2014). Natural emission of Hg coming from volcanoes, weathering of rock and ores, soil evaporation, forest fires (Chen et al. 2010; Feng et al. 2003), whereas human activities such as municipal solid waste, pharmaceutical industry, mining, fuel combustion, electronic, paint and paper industries are the most important anthropogenic origins of Hg in the environment (Rodrigues et al. 2006; Biester et al. 2002; Chatterjee et al. 2014). Owing to the bioaccumulation, persistency, and toxicity of Hg, it was itemised as a priority pollutant in the controlled environment by the United Nations Environment Programme (UNEP), World Health Organisation (WHO) and Food and Agriculture Organisation (FAO) (Chen et al. 2010; Xinmin et al. 2006). Mercury mostly remains in gaseous and particulate phases, unlike other toxic metals that tend to exit only as particulate phase (Solgi et al. 2014). The dust collected from the streets in this study are a combination of airborne particulate matter with a diameter less than 10µm and is frequently constituted of fuel combustion dust, and secondary dust derived from relocated urban soil particles (Hu et al. 2011; Xinmin et al. 2006; Ying et al. 2009). The dust collected from the streets provides evidence of anthropogenic activity, geological and geographical sources. Comparison of soils and dust in an urban environment show that collected dust from the streets are easily affected by urban populations, particularly children (Hu et al. 2011). Moreover, previous works confirm that toxic metal enrichment in street dust are mostly higher than soil (Li et al. 2001; Lu et al. 2010; Manta et al. 2002; Ordonez et al. 2003; Shi et al. 2008; Wei and Yang 2010; Xinmin et al. 2006). In general, the study of ecological risk and health risk assessment of toxic metals like Hg, especially in street

✉ Corresponding Author. E-mail: A.nazarpour@iauahvaz.ac.ir , Phone: +989163721558, Fax: +986133348376

dust may be more significant than urban soils. Therefore, there are limited related literature for developing countries, especially in Iran, which focused on the toxic metal content, spatial distribution, pollution assessment, and source identification (Dankoub et al. 2012; Farzin et al. 2008; Karimi et al. 2011; Solgi et al. 2014; Soltani et al. 2015). The US EPA's human health risk and ecological risk assessment methods has been effectively applied to investigate toxic metals from street dust for children and adults (US EPA, 1986, 2001, 2002; Hu et al. 2011; Wei and Yang 2010; Wei et al. 2015).

Ahvaz (31°19'45" N and 48°41'28"), the capital of Khuzestan province in Iran, with a population of approximately 1.32 million in 2016, is situated in south west Iran bordering Iraq, Kuwait and Saudi Arabia, whose deserts are the main source of dust events in the Middle East (Zarasvandi et al. 2011a and b). Ahvaz is one of the most important cities in Iran, owing to the presence of large industrial plants. For example, the National Iranian South Oil Company (NISOC), National Iranian Drilling Company (NIDC), Khuzestan Steel Company (KSC), Iran National Industrial Group (INIG) Khuzestan Oxin Steel Company (KOSC) and surrounding industrial towns has made Ahvaz as one of the most important industrial centres of Iran and it in turn, attracted many economic migrants to Ahvaz. In this study, the spatial distribution of Hg, ecological risk and human health risk assessment of Hg in urban street dust was investigated across five parts of the Ahvaz city areas. These included; residential areas (RA), industrial areas (IA), public gardens (PG), and roadside areas (RS) and suburbs (SA). The study outcomes are valuable for environmental scheduling and development through identification of hotspots, which will help to inform mitigation strategies.

2. Material and methods

2.1. Sampling and treatments

In July 2014, ninety six (96) street dust samples were collected from the 2 city, with locations shown in Figure 1. The number of samples for each of land uses were as follow: 23 samples for IA, 25 samples for RA, 25 samples for RS, 13 samples for SA, and 10 samples for PG. In addition, five dust samples were collected from new developed suburban area with no industrial activities high dense population and traffic volume as background samples (Fig. 1). Samples were selected using a commercial vacuum cleaner in the driest month of the year to prevent rain-washing away street dusts. A Geographical Information System (GIS) technique "create random points" were selected to create a sampling strategy that cover the whole of study area. Finally, the sampling points were classified according to the target land uses. The meteorological conditions were steady throughout the sampling, with no rainfall during one month earlier of sample collection. About 500 g of a composite street dust sample was collected by brushing an area of 1×1 m² from the pavements. The samples were sieved through a 220 mesh (63µm), air-dried and stored in polyethylene bags and labelled based on Tanner et al. (2008) and McKenzie et al. (2008).

Five grammes of dust collected from the street dust samples were mixed in highly purified nitric-hydrofluoric perchloric acids (5mL HNO₃- 4mL HCL- 2HClO₄) (Merck Ltd) to dissolve the street dust samples for measurement of Hg, based on the protocol GB/T 17441-1997 and GB/T 17238-1997 (Griepink and Tölg 1989). The Hg concentration of street dust samples were determined by Atomic Absorption Spectrometry (AAS) (220Z, Varian Co., Australia). The Quality assurance (QA) and quality control (QC) was assessed by measurement of blank and duplicate samples and certified reference material NIST 2710 which provided an accuracy of 100 ± 5% (n = 15), whilst the precision of duplicate samples was 4 to 6%.

2.2. Pollution Index (PI)

To evaluate the extent of pollution, a modified pollution index (PI) for Hg was assigned to each sampling point (Huang 1987). The PI was expressed as:

$$\begin{array}{l}
 \text{PI} = C/X_a \\
 \text{PI} = 1 + (X - X_a)/(X_c - X_a) \\
 \text{PI} = 2 + (X - X_c)/(X_p - X_c) \\
 \text{PI} = 3 + (X - X_p)/(X_p - X_c)
 \end{array}
 \qquad
 \begin{array}{l}
 C \leq X_a \\
 X_a < C \leq X_c \\
 X_c < C \leq X_p \\
 C > X_p
 \end{array}
 \quad (1)$$

Where C is the measured value of Hg in the study area, X_a is the maximum limit value of the Hg, X_c and X_p are the maximum limit value of the low and high level of pollution, respectively. The values of X_a, X_c and X_p, for Hg are 0.15, 1 and 1.5 mg.kg⁻¹ (Yu et al. 2004).

The PI value was categorised as: non-pollution for PI<1, low level of pollution (1<PI<2) and high pollution level (PI>3) (Yu et al. 2004).

2.3. Health risk assessment model

The human health risk assessment is a useful method to obtain further evidences for the risk management of heavy metals in the metropolitan environment. Based on the developed US Environmental Protection Agency (US EPA 1986) and Exposure Factors Handbook (US EPA 1997), mercury (Hg) exposure from street dust can occur via four main paths in adults and children.

- (1) Daily amount of exposure of direct hand to mouth ingestion of dust particles (D_{ing}),
- (2) Daily amount of exposure of mouth and nose inhalation of suspended particles (D_{inh})
- (3) Daily amount of exposure of dermal contact with dust particles (D_{dermal}).

Mercury exposure through each of the four paths can be expressed and calculated using Eqs. (2- 5) (US EPA 1997, 2001 and 2002) and (Zheng et al., 2010a, b).

$$ADD_{ing} = \frac{C \times IngR \times CF \times EF \times ED}{BW \times AT} \quad (2)$$

$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \quad (3)$$

$$ADD_{dermal} = \frac{C \times SA \times CF \times AF \times ABF \times EF \times ED}{BW \times AT} \quad (4)$$

$$ADD_{vapour} = \frac{C \times InhR \times EF \times ED}{VF \times BW \times AT} \quad (5)$$

The exposure parameters that were employed to the health risk assessment models are summarised in Table 1. The data are chosen by applying background values from US EPA (1997, 2001, and 2002). Then the daily contact dose for three pathways (D_{ing}, D_{inh}, and D_{dermal}), Hazard Quotient (HQ), and Hazard Index (HI) models were calculated Eqs. (6 and 7). HI denotes to the “total Hazard Quotient (HQ) of several materials or exposure paths” (US EPA, 1986). Therefore, a mixture of non-cancerous risk can be assessed by calculating the HI of every exposure pathway such as ingestion, dermal, and inhalation (US EPA, 1989). The possible non-carcinogenic and carcinogenic risks were computed by applying subsequent calculations (US EPA, 1989):

$$HQ_i = \frac{ADD_i}{R_f D_i} \quad (6)$$

$$HI = \sum HQ_i \quad (7)$$

The reference dose (R_fD) (mg kg⁻¹ day⁻¹) is an estimate of high level acceptable hazards for humans during regular contact, and *i* is the route of exposure. The threshold R_fD can be utilised to calculate whether there is a harmful impact throughout life. Reference dose for mercury used in this study were taken from the US Department of Energy’s RAIS

compilation (2004), which are $3.0E-4$ for direct ingestion, $2.10E-5$, for dermal absorption and $8.57E-5$ for inhalation through not only mouth and nose, but also vapor.

The ratio of average daily dose (ADD) and reference values can be utilised in determining the health effect of toxic metals. Values with lower than the reference dose indicate that there is no harmful health impact. On the other hand, the ADD value higher than the RfD , indicate that the contact pathway will cause harmful human health effects (US EPA, 1997).

Where:

$HQ \leq 1$: there are no harmful health effects

$HQ > 1$: there are harmful health effects

The HI value is the total of the HQs and implies the total risk of non-carcinogenic for a single toxic metal.

If $HI < 1$: indicate that there is no risk of non-carcinogenic impacts.

If $HI > 1$: arise a probability of non-carcinogenic impacts with the increasing HI value (US EPA, 2001, 2002).

3. Results and Discussion

3.1. Mercury concentrations

Statistical analysis of Hg in the Ahvaz street dust samples including local background value are presented in Table 2. Mercury concentrations varied from 0.02 to 8.75 $mg.kg^{-1}$ with an average value 2.53 $mg.kg^{-1}$. Positive coefficient skewness (0.74) indicating that Hg concentration values do not have a normal distribution pattern (Fig. 2). In addition, Kolmogorov–Smirnov (K-S) normality test, indicate that Hg do not follow a normal distribution (K-S; $P > 0.05$), which indicates the presence of samples containing high concentrations and hotspots of Hg in street dust. The mean value of Hg in Ahvaz street dust samples was 62 times greater than the background values (Table 2). These values imply that Hg in the street dust from Ahvaz were affected by anthropogenic sources.

A spatial distribution map of Hg overlapping with the location of the main anthropogenic pollution sources might indicate the possible source of Hg in street dust from Ahvaz. As indicated in Fig (3) the main hot spots were distributed in the old urban area with high residential density, traffic emission, and long-term deposit of Hg. Furthermore, areas, which coincided with industrial activities such as oil-drilling, steel smelting related industries, and small industrial towns, also exhibited a high level of pollution (Fig 3). Mercury concentrations in the street dust depends on some factors including residential density, industrial materials, traffic volume, and technologies employed, as well as local metrological situations and wind direction (Chen et al. 2010). According to different land used, the mean value of Hg concentration followed the sequence: $RS > IA > RA > PG > SA$. This order indicate that roadside and industrial land uses as a result of more anthropogenic activities including traffic emission, steel smelting industry, oil-based mud drilling activity by NIDC in suburban area, Carbon Black company are the main sources of toxic metals in Ahvaz city, and have more potential of accumulation of toxic metals such as Hg. The mean value of RS was greater than the other types of land-use as shown in (Table 2). It is obvious that fuel combustion is one of the main important Hg sources (Sun et al. 2013; Tian et al. 2010), which accounts for about 20% of the total anthropogenic Hg emission in the studied city (Statistic Bureau of Wuhan, 1990-2011).

Hg concentration in Ahvaz street dust is high in comparison to many other cities in the world (Fig. 4). For example, whilst the mean concentration of Hg in the all samples in the study area is lower than Aviles, Spain (Lu et al. 2009), meanwhile, it was higher than that in Beijing, China (Xinmin et al. 2006); Shanghai, China (Shi et al. 2011), Baoji, China (Lu et al. 2009; Tang et al. 2013); Taipei, Taiwan (Zhang et al. 2014), Hong Kong (Tanner et al. 2008); Ottawa, Canada (Rasmussen et al. 2001); Kavala, Greece (Christoforidis and Stamatis, 2009); Luanda, Angola (Lu et al., 2009); and Shiraz, Iran (Keshavarzi et al. 2015) and higher than the UK environmental guideline value ($1.0 mg.kg^{-1}$; DEFRA and Environmental Agency, 2009).

3.2. Metal pollution index

The pollution index of Hg was calculated based on Huang (1987), and it ranged from 0.26 to 16.33 $mg kg^{-1}$, with the mean of 5.86 $mg kg^{-1}$ (Table 3). Descriptive statistical analysis and number of samples in each class of PI for street dust across each land-use category are presented in Table 3. Assessment of street dust samples shows that there were 7 dust samples (7%) with a $PI \leq 1$, 2 street dust (2%) had a PI between 1 and 2. Thirteen street dust samples (14%) had a PI between 2 and 3 and 74 dust samples (77%) had a $PI > 3$, exhibiting a high level of Hg pollution.

The proportion of pollution levels of Hg in Ahvaz street dust samples are presented in Table 3. The mean concentration of Hg in each land-use decreases as follow: RA>RS>PG>IA>SA. The concentration of Hg from the residential area (RA) (N=25) ranged from 0.93 to 13.83 mg kg⁻¹, with a mean of 6.68 mg kg⁻¹, was the highest category and higher than the mean Hg concentrations reported for cities shown in Table 3. In addition, as shown in Table 3, over 95% of the street dust samples in RA, indicate Hg pollution in the study area. In general, more than 84% of RS street dust, 82% of IA samples, 80% of RA samples, and 70% of PG street dust samples were highly contaminated with Hg. PI values in different land-use indicated serious anthropogenic activities, which resulted in the accumulation of Hg in street dust in Ahvaz.

Overall, these findings suggest that the street dust from Ahvaz city is polluted from anthropogenic Hg emissions. Dust samples with high and moderate Hg pollutions were located in areas with high dense pollution, high traffic volume, and manufacturing companies such as steel smelting, chemical industries, industrial towns, drilling pits, and uncontrolled oil-based mud.

3.3. Health risk assessment of mercury

The possibility of adverse effects and health risk of Hg in urban street dust was assessed. The results of the health risk assessment are presented in Table 4, and indicated that the average daily dose (ADD) and HQ values for Hg in street dust particles for adults and children are similar and decreases in the following order for adults and children: ADD_{vapour}>ADD_{ing}> ADD_{dermal}> ADD_{inhalation}. For adults and children, the path of vapour was higher compared to ingestion absorption dermal contact and inhalation exposure of re-suspended dust. The results also showed that for non-carcinogenic impacts, vapour seemed to be the major path of contact to dust collected from the street particles, therefore causing a higher health risk to adult and children for Hg, followed by ingestion, dermal contact, and inhalation absorption. HQ values for adults exhibited low variation across each land-use and the order of direct vapour>ingestion> dermal > inhalation contact are the major paths of contact for adults and children in each land-use area. The total HQ of studied vapour in studied land uses account for more than 80% of the HI value. In the case of children, inhalation of mercury vapour is the major path of exposure in all of the exposure pathways, which was compatible with Fang et al. (2011) and Sun et al. (2013).

It can be observed that children are more likely to be exposed through hand-to-mouth transfer. However, daily outside activities of adults such as rubbing sweat, consumption of food outside, or not washing hands or faces before eating or drinking, may also cause the risk of ingestion and dermal exposure of Hg in re-suspended street dust on food, hands or faces. The inhalation and vapour of Hg from re-suspended particles through mouth and nose is likely to be negligible once associated with other pathways (Zheng et al. 2010a). Similar outcomes were found in earlier reports of exposure to toxic metals in metropolitan street dust (Ferreira-Baptista and De Miguel 2005; Zheng et al. 2010a). It is apparent that the increase of contact rate and inhalation and subsequently vapour exposure can result in the non-carcinogenic harmful special impacts to the tissues of children. Exposure to street dust in large amounts/dose by children may also cause non-carcinogenic results, as reported for other investigations (Ferreira-Baptista and De Miguel 2005; Sun et al. 2013; Zhang et al. 2014).

Analyses indicated that the HI value for Hg in each sample type for children and adults, showed that daily vapour inhalation and ingestion of street dust were 0.55 and 0.21, therefore, lower than the safe level (=1). The HI values for discrete land uses for children and adults is represented in Table 4. Compared to adults, the non-carcinogenic health risk of children due to greater sensitivity to Hg exposure is higher. The HI for each land-use were lower than the safe level (HI≤1) in both of children and adults, which decreases as the following order: RS>IA>RA>SA>PG, indicating that exposure of Hg in street dust would result in potentially adverse health impacts for children in each study area. The HI values also show that the highest level of risk is related to samples, which are close to roadside (RS) and industrial area (IA) land uses (Table 4). These results confirm that high traffic volume, and industrial activities are the main sources of Hg in Ahvaz street dust. On the other hand, the lowest HI are related to public garden (PG) samples which is lower than the safe level (≤1) in children and adults. With regarding, the high HI value for road side land use, vapour pathway is the main route of Hg in Ahvaz street dust samples, as evidenced by suspended particles from traffic emission. There were no non-carcinogenic risk of Hg in public gardens (PG) for adults and children. Non-carcinogenic risks for children from Hg indicated more potential harmful health risks, approximately 4-6 times greater than adults in the Ahvaz.

Meza-Figueroa et al. (2007) reported that HI values were greater than the safe level ($HI > 1$) in all studied samples for children with more hand-to-mouth action for what is a more subtle sub-population, by which dust containing Hg and other toxic metals can be freely inhaled. The lower body mass of children results in relatively greater absorption and accumulation of Hg ($\text{mg/kg-bodyweight/day}$) compared to adults (Wang et al. 2011). Consequently, the hazard health risk (HI) for children exposed to Hg in street dust is higher than that of adults. Vapour exposure is main pathway with higher risk for adults. This result was comparable to reported urban surface dust for Guangzhou, China (Wang et al., 2011), and can be described by the greater values of dermal exposure area (SA) and exposure duration (ED) of adults. Hence, the environmental and health risk effects of the enhanced Hg in the Ahvaz demand more detailed examination.

4. Conclusion

The Hg concentration in Ahvaz urban street dust ranged from 0.02 to 8.75 mg.kg^{-1} . Raw data for street dust was positively skewed and did not follow a lognormal distribution. The Hg content for street dust in each land-use decreased as $RS > IA > RA > PG > SA$. The main reason for the high concentration of Hg in RS and IA was related to high traffic volume, fuel combustion, and industrial activities. The result of mapping the distribution of Hg in Ahvaz street dust suggests that the high concentration of Hg mainly coincides with source points, such as industrial districts and the high-density residential area with an old urban history. The Hg content also indicates a decreased trend from the center to suburban areas of city.

The health risk assessment model was employed for calculation and results showed potentially high non-carcinogenic harmful impacts of Hg on children and adults. The HQ values for the major exposure pathway for children and adults decrease as follows: vapour > ingestion > dermal contact > inhalation. For two populations, children showed higher health risk than adults, with the HI value for total samples were 0.55 for children, and 0.21 for adults. The HI for each land-use were lower than the safe level ($HI \leq 1$) for children and adults and decrease in a similar order as: $RS > IA > RA > SA > PG$, indicating that exposure to Hg in street dust could have health consequences for children in the study areas.

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