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Human exposure to toxic metals via contaminated dust: Bio-accumulation trends and their potential risk estimation

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HIGHLIGHTS

- Human exposure to trace metals via dust, in relation to different land use.
- Trace metal estimation by ICP-MS.
- Dust as major route of Cd, Pb, Co, Mn and Cr exposure for humans.
- Potential health risk from exposure to Cd (for children) and Pb via dust.
- Zn, Cr, Pb, Ni and Cd in hair and nail were beyond the ATSDR threshold.

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ABSTRACT

We assessed the levels of potentially toxic trace metals, Zinc (Zn), Lead (Pb), Manganese (Mn), Copper (Cu), Nickel (Ni), Chromium (Cr), Cobalt (Co), and Cadmium (Cd), in dust, hair, nail and serum, sampled in rural, urban and industrial areas of Punjab, Pakistan. Trace metals occurrence in all samples, in descending order, was: Zn, Pb, Mn, Cu, Cr, Ni, Co, Cd. The samples from the urban areas showed significantly higher concentration of toxic trace metals (Zn, Ni, Cr, Co, Mn, and Cd) than those from industrial (which conversely had higher levels of Pb and Cu), and than samples from rural areas. Bioaccumulation patterns showed that dust exposure is one of the major routes into human body for Cd, Pb, Co, Mn and Cr, while the burden of Zn, Cu, and Ni can be more linked to dietary sources. The concentrations of trace metals in the samples from Punjab were comparable and/or higher than those reported worldwide. In many cases, the levels of Zn, Cr, Pb, Ni and Cd in hair and nail were beyond the ATSDR threshold guideline values that may cause some serious health effects. Hazard Index (HI) calculated for trace metal concentrations in the human population of Punjab points particularly to health risks from Cd (for children in urban and industrial areas) and from Pb (for all sub-groups).

1. Introduction

Heavy metal pollution has recently become a severe menace globally, and especially in South Asia, where high exposure of the human population stems from lack of awareness and from depressed socioeconomic conditions (Malik et al., 2010; Abdullah et al., 2015). In developing countries, rapid urbanization and population rise has resulted into widespread conversion of vast undisturbed lands into agricultural and residential, commercial, and industrial areas (Faiz et al., 2009; Kurt-Karakus, 2012). It has also been widely reported that anthropogenic activities have become a major cause of environmental degradation via industrial discharge, traffic emission, excessive use of fertilizer and pesticides, and waste from municipal activities (Kurt-Karakus, 2012; Yoshinaga et al., 2014; Abdullah et al., 2015). Within this scenario, different industries, automotive, road construction, paints, metal smelting, contributed to introduce into the atmosphere harmful particles, like toxic metals contaminated particulate, aerosols and other inorganic elements. Agricultural activities as well

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contributed to atmospheric contamination via the use of pesticides, fertilizers, and farm machinery (Eqani et al., 2012). Highly toxic metallic impurities were thus introduced into the atmosphere, and became part of the dust that originates from the soil (Zheng et al., 2009; Hu et al., 2012), ultimately leading to risks for the environment and for human health. Street dust is an important factor of urban and rural pollution and has been widely reported to affect the health of the exposed population (Zheng et al., 2009). Humans are expected to be exposed to metal-contaminated dust via inhalation, ingestion and dermal contact, with several health risks (Sörme and Lagerkvist, 2002; Hu et al., 2012).

Trace metals are non-biodegradable and become biomagnified into body tissues, thus causing severe health risks, including disruption of the central nervous system, reproductive failure, genotoxicity, and gastro-intestinal problems (Zheng et al., 2009; Wu et al., 2010). Children and toddlers are more susceptible to trace metal exposure and toxicity than adults due to their hand-to-mouth activities and fast growth rates (Kurt-Karakus, 2012; Ali et al., 2013; Peña-Fernández et al., 2014). Diverse tissues, hair, nail, urine and serum, have been effectively used to study environmental burden of toxic metals (Kazi et al., 2008; Samanta et al., 2004; Zheng et al., 2009; Cebi et al., 2011; Abdulrahman et al., 2012). Among these biomarkers, hair and nail are non-invasive samples, easy to handle. Trace metals can easily integrate into their protein structures, due of their complex chemical nature i.e., sulfur containing amino acids and fibrous protein (Tamás et al., 2014). Moreover, human hair and nail reflect long term exposure, as growth rate ranges from 0.05 to 1.2 mm per week (Chatt et al., 1989). Conversely, being an invasive matrix, blood has several disadvantages, including bad participant response in volunteer epidemiological studies (Rockett et al., 2004). Moreover, blood and urine may only fingerprint the recent level of trace metals, and thus it is not suitable to evaluate long-term health risks, and they contain lower amounts of toxic chemicals making detection by chemical analysis more challenging (Wang et al., 2009).

In recent years, due to exponential population growth, Pakistan has undergone widespread environmental degradation, as a result of the rapid urbanization, industrialization and intensified agricultural activities (Khan et al., 2013; Abdullah et al., 2015). The contaminants released from densely populated areas move toward low lying rural areas via industrial effluent discharge, waste disposal into the rivers, traffic emissions and atmospheric deposition (Eqani et al., 2012; Abdullah et al., 2015). The situation is further aggravated by the lack of law enforcement and general public awareness about dust emission and air quality. To date, many studies have been conducted in Pakistan on health risks posed by trace metal toxicity, but they have been mainly focused on drinking water (Muhammad et al., 2011), fish (Rauf et al., 2009; Hussain et al., 2014), vegetables (Ismail et al., 2005; Khan et al., 2010), and soil matrices (Malik et al., 2010), and their main aim was to provide the baseline data. Till now, no detailed study has reported trace metal contents in dust and their associated health risks for the inhabitants of different land-use settings of Pakistan. The present study aims to assess the levels of trace metals into dust, and the concentrations in human hair, nail and serum in different land-use settings through Punjab, Pakistan, and to evaluate the risks posed to human health by the exposure to trace metal contaminated dust through inhalation, ingestion and dermal contact.

2. Materials and methods

2.1. Study area

Samples were collected from rural, urban and industrial areas of the Lahore and Sargodha districts of Punjab (Fig. S1). Lahore is one of the most densely populated and industrialized cities in Pakistan, with 9000 operational industrial units. The major sources of trace metal contamination into the atmosphere of Lahore are textile, chemical, and automotive industries, mechanical shops, vehicle service facilities, electric appliances, and factories producing plastic and PVC (Abdullah et al., 2015). Sargodha is located 206 km from Lahore, in an area mostly covered by agricultural fields. But even in this area, heavy traffic emission, excessive use of fertilizers and industrial effluents may provide great trace metals burden, together with industrial units that produce diesel engines, electric goods, pesticides, PVC, textiles, beverage, food, and sugar.

2.2. Sample collection

We collected dust and human hair and nail samples from two rural sites between Lahore and Sargodha city, and from urban sites within both Sargodha and from Lahore city. Most samples for industrial sites were collected from the famous industrial zone of Lahore, with few industrial samples collected near Sargodha city, where many brick kilns and small industries are situated and are considered important source of trace metals since coal is used as a source of energy in this area. Dust ($n = 30$), hair (30), nail (30) and serum (30) samples were collected from rural, urban and industrial areas ($n = 10$ from each land use) in both districts. The dust samples were collected by brushing the sampling area surface, using brushes from the respective household in order to avoid cross-contamination between sampling locations. After brushing, dust was wrapped in aluminum foil, sealed in polyethylene zip bags, and stored at room temperature. The dust samples were of fine particle size (<50 μm) which can be trapped by inhalation. Hair sampling was carried out by taking 3 cm of hair from the occipital region of both genders, in order to check for different exposure levels due to occupational habits, using non-metallic scissors (Wang et al., 2009). Information regarding spraying, dying and other treatments to the hair was also recorded. After collection, samples were transferred to plastic air-tight bags and sealed. Toe nail clippings were also collected in air-tight sealed bags. Serum samples were 6 cc of blood collected by a sterile BD 10 cc syringe, with the assistance of paramedic staff. The study was approved by the Ethical Review Committee of COMSATS, Islamabad and UoS, Sargodha, Pakistan. A detailed questionnaire recorded the occupation, smoking habits, and other information relevant to the possible contamination sources. All the collected samples were stored at 4 °C prior analysis.

2.3. Sample preparation

Before use, all the containers were soaked in 10% (v/v) HNO_3 overnight and repeatedly rinsed with ultra-pure water, obtained from a Milli-Q system (Millipore Corporation, Billerica, MA, USA). All the samples were weighed and digested overnight with 1 mL GR grade 65% (v/v) HNO_3 (CNW Corporation, Shanghai, China), then on the next day with 1 mL GR grade 30% (v/v) H_2O_2 (Sinopharm Chemical Reagent Co., Ltd, Beijing, China). For the dust samples, we also added 1 ml of $HClO_4$ to completely digest the samples. Afterward, the samples were mixed, sealed in Teflon microwave digestion tubes, and digested in an accelerated microwave digestion system (Mars CEM, CEM Corporation, Matthews NC, USA) at 800 W, 120 °C for 10 min and then 800 W, 170 °C for 30 min. All digested samples were filtered by using 0.22 μm nylon membrane and finally the volume was raised to 10 mL using ultra-pure water.

2.4. Analytical aspects

Trace metals (Pb, Cd, Cu, Co, Zn, Ni, Cr and Mn) were measured using ICP-MS Inductively Coupled Plasma Mass Spectroscopy