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# Mercury contamination in deposited dust and its bioaccumulation patterns throughout Pakistan

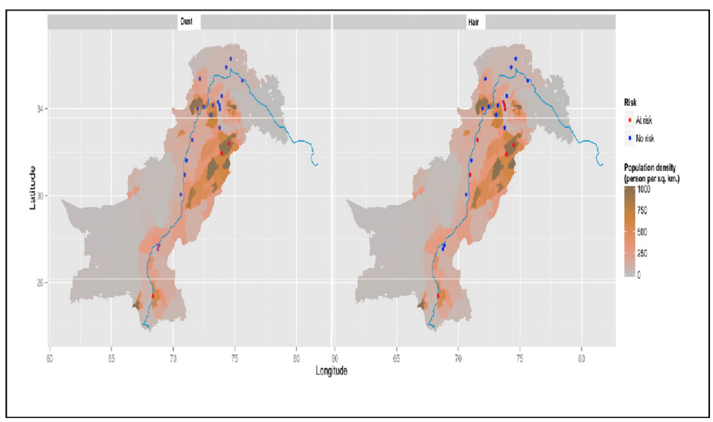
Syed Ali Musstjab Akber Shah Eqani [a,b,*], Avit Kumar Bhowmik [c], Sehrish Qamar [b], Syed Tahir Abbas Shah [b], Muhammad Sohail [b], Sikandar I. Mulla [a], Mauro Fasola [d], Heqing Shen [b,*]

[a] Key Lab of Urban Environment and Health, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, PR China
[b] Public Health and Environment Division, Department of Biosciences, COMSAT Institute of Information & Technology, Islamabad, Pakistan
[c] Stockholm Resilience Centre, Stockholm University, Stockholm, Sweden
[d] Dipartimento Scienze della Terra e dellAmbiente, Università, Via Ferrata 9, I-27100 Pavia, Italy

## HIGHLIGHTS

- THg in dust from different areas of Pakistan in context of urbanization.
- THg in dust samples were associated with population density by regression analysis.
- Association of paired dust and scalp hair from Indus plain areas.
- Human population (>40%) was at potential risk from THg in Urban areas.
- HI >1, reflected non-carcinogenic risk for children and for adults in industrial areas.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Mercury (Hg) contamination of environment is a major threat to human health in developing countries like Pakistan. Human populations, particularly children, are continuously exposed to Hg contamination via dust particles due to the arid and semi-arid climate. However, a country wide Hg contamination data for dust particles is lacking for Pakistan and hence, human populations potentially at risk is largely unknown. We provide the first baseline data for total mercury (THg) contamination into dust particles and its bioaccumulation trends, using scalp human hair samples as biomarker, at 22 sites across five altitudinal zones of Pakistan. The human health risk of THg exposure via dust particles as well as the proportion of human population that are potentially at risk from Hg contamination were calculated. Our results indicated higher concentration of THg in dust particles and its bioaccumulation in the lower Indus-plain agricultural and industrial areas than the other areas of Pakistan. The highest THg contamination of dust particles (3000 ppb) and its bioaccumulation (2480 ppb) were observed for the Lahore district, while the highest proportion (>40%) of human population was identified to be potentially at risk from Hg contamination from these areas. In general, children were at higher risk of Hg exposure via dust

* Corresponding authors at: Public Health and Environment Division, Department of Biosciences, COMSATS Institute of Information Technology, Islamabad, Pakistan.
*E-mail addresses:* ali_ebl2@yahoo.com (S.A.M.A.S. Eqani), hqshen@iue.ac.cn (H. Shen).

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particles than adults. Regression analysis identified the anthropogenic activities, such as industrial and hospital discharges, as the major source of Hg contamination of dust particles. Our results inform environmental management for Hg control and remediation as well as the disease mitigation on potential hotspots.



## 1. Introduction

Mercury (Hg) is a persistent and ubiquitous toxic trace metal, which originates from both natural and anthropogenic processes into environment in elemental, organic and inorganic forms (Hsiao et al., 2011; Sun et al., 2013). Natural and geogenic processes such as volcanic emissions and forest fires may induce Hg contamination into environment. However, anthropogenic activities such as mining, industrial, agricultural and medical wastes discharges have also reported to contribute towards severe Hg contamination of environment (Lu et al., 2009). One of the major pathways of human health exposure to Hg contamination is Hg-contaminated dust particles, which are transported in atmosphere both across long- and short-distances (Sun et al., 2013). Eventually, Hg can enter human bodies through ingestion, inhalation and dermal contacts to Hg-contaminated dust particles. Nevertheless, in contrast to atmospheric particulate material (potentially enter via inhalation route), deposited dust reflected more authenticated information about human exposure of trace metal impurities and points to the source (Mohmand et al., 2015). Upon entering human bodies, Hg is converted into methyl mercury, which may lead to serious implications including neuro-disorders, reproductive abnormalities, kidney failure, emotional instability, gingivitis and tremors (Hsiao et al., 2011; Azizullah et al., 2011).

In recent years, like in many developing countries, Pakistan has been facing severe environmental pollution due to its exponential population growth, vast industrial and urban activities, and lack of environmental management and awareness both at governmental and public level (Azizullah et al., 2011). Nevertheless, legally binding approach (LBA) is still under consideration by developing countries and being negotiated by voluntarily approach, which would further aggravated the problem in Asian countries (Andresen et al., 2012). According to a preliminary report of UNEP (2000), Pakistan is a potential hotspot of Hg contamination with a minimum disposal of 10,800 kg/year to a maximum of 36,900 kg/year into the environment, originating mainly from anthropogenic processes such as chlor-alkali plant, light products manufacturing units, cement industries, dental clinics wastes, incinerators and other coal-based primary and secondary unit operations (Abbas, 2009; Malkani, 2012; Khwaja et al., 2014). In the northern mountainous areas, amalgamation and smelting in gold panning activities have been practiced for decades along the Gilgit, Hunza and Indus rivers, which may also cause a widespread Hg contamination of freshwater resources in these and other low lying regions of Pakistan (Azizullah et al., 2011; Khan et al., 2012; Biber et al., 2014). However, Hg concentration in different environmental pathways such as dust and freshwater have rarely been monitored and hence, environmental management authorities are lacking a country level baseline data on Hg contamination.

Biomonitoring of Hg accumulation and its fate is important for disease mitigation as well as for remediation of Hg contaminated areas (Al-Majed and Preston, 2000; Adimado and Baah, 2002; Agusa et al., 2005). Based on exposure pathways, different strategies for Hg biomonitoring have been developed, such as monitoring of Hg concentration in human blood, urine, hair and nail samples (Adimado and Baah, 2002; Akira et al., 2004). Among them, scalp-hair and toe-nail samples have been preferred due to their high proneness to absorb Hg and other trace metals from environment driven by high proteins content, fibrous structure and high creatine levels (Mohmand et al., 2015; Akira et al., 2004; Adimado and Baah, 2002). Moreover, high representativeness of Hg contamination, ease of collection and Hg level quantification, and chemical non-invasiveness entail high suitability of these samples for monitoring Hg bioaccumulation in resource-constraint developing countries (Hsiao et al., 2011; Sun et al., 2013; Ohno et al., 2007). Furthermore, several studies reported a positive association of dietary (fish etc.) and/or non-dietary (dust, air etc.) mercury exposure with Hg concentration in human hair and nail samples (Adimado and Baah, 2002; Agusa et al., 2005). However, an analysis of bioaccumulation patterns of Hg through human hair and nail samples is missing at the regional scale of Pakistan and hence, population at potential risk of Hg contamination is largely unknown.

We provide the first detailed baseline data on Hg contamination in Pakistan by assessing Hg concentration level in deposited dust particles collected from five zones along the Indus river, which is a potential hotspot of Hg contamination and bioaccumulation with >100 million people. Concentrations of THg in dust particles were measured for their associations with natural and anthropogenic sources, and compared with the guideline values for identification of potential human health risks from exposure to THg contaminated dusts. Bioaccumulation patterns of THg were identified by analyzing human hair samples and the proportion of risky samples were identified for each site to indicate the proportion of population at risk.

## 2. Materials and methods

### 2.1. Sampling strategy and Hg concentration level assessment

Pakistan is characterised by diverse land use and altitudinal settings, including the mountains of Himalaya, Karakoram and Hindukush ranges in the North, flat-lying Indus Plain in the east, the upland Baluchistan plateau in the West and the coastal belt of Arabian Sea in the South (Fig. 1; S1; ISCGM, 2014). Hence, we divided Pakistan into five zones for sampling based on their land use types and altitudinal ranges (see Fig. S1 for details): 1) Swat Valley and Gilgit-Baltistan, which includes the high mountain ranges of Gilgit, Skardu and Hunza, 2) Kashmir valley, which includes mountain valleys of Kashmir area, 3) Lower Himalayan Mountains including the capital Islamabad, Abbotabad, Nowshera and Swabi, 4) Indus Plain-Agriculture, which includes agricultural areas located along the Indus river in Dera Gazi khan, Bhakkar, Layha, Mianwali, Sukkur and Kahirpur, and 5) Indus Plain-Industrial with highly urbanized settlements and industrial areas in Lahore, Sargodha and Hyderabad (Fig. 1). Details on the location, elevation and land use of zones are given in the supplementary Table S1. We collected samples of dust ($n = 110$) and of human hair ($n = 190$) from 22 sites in these zones (Fig. 1 and Table S1). The dimension of dust samples were determined by using the sieve (mesh size of 50 μm). The samples were of fine particle size (<50 μm), which can also be introduced by inhalation. A room (on the ground floor at all sites) with four-ways cross window was selected at each site and a wooden table had placed inside each room. Composite samples were collected over a period of 8 dry days (of each month) during the summer months of May–June 2013. The dust that had settled on the surface was periodically brushed off and collected in the sampling bags. The area receiving the dust was also measured for the computation of amount of dust fall per unit area and per unit of time. To avoid external contamination of the samples, dust particles were collected using a plastic, metal-free brush in a plastic collection pan. For details on sample collection and transport, readers are referred to Eqani et al. (2016). The scalp hair samples were collected