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Arsenic levels from different land-use settings in Pakistan: Bio-accumulation and estimation of potential human health risk via dust exposure

Marghoob Subhani [a,1], Irfan Mustafa [a], Ambreen Alamdar [b], Ioannis A. Katsoyiannis [c], Nadeem Ali [d], Qingyu Huang [b], Siyuan Peng [b], Heqing Shen [b,*], Syed Ali Musstjab Akber Shah Eqani [e,**,1]

[a] Department of Biological Sciences, University of Sargodha, Sargodha, Pakistan
[b] Key Lab of Urban Environment and Health, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, PR China
[c] Aristotle University, Department of Chemistry, Division of Chemical Technology, Box 116, 54124 Thessaloniki, Greece
[d] Center of Excellence in Environmental Studies, King Abdulaziz University, Jeddah, Kingdom of Saudi Arabia
[e] Public Health and Environment Division, Department of Biosciences, COMSAT Institute of Information and Technology, Islamabad, Pakistan

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ABSTRACT

The present study aims at assessing arsenic (As) levels in outdoor dust and human exposure risks at different land use setting (i.e., rural, industrial, urban) from Punjab, Pakistan. The results showed higher As concentrations (mg/kg) in all the sample types (i.e., dust, hair and nail) collected from industrial sites (9.78, 2.36, 2.5) followed by urban (7.59, 0.38, 0.88) and rural sites (6.95, 0.52, 1.12), respectively. In the current study, we also carried out human risk assessment via contaminated dust exposure, which suggested that dust ingestion is the major route of As contamination for the associated population, followed by the inhalation and dermal contact, at all studied land use settings. Hazard Index (HI) calculated for non-carcinogenic health risks for adults showed higher values at industrial (0.65) and urban (0.53) sites, which reflected that dust exposure is the major contributing source of human arsenic burden and may pose several adverse health effects. Carcinogenic risk values showed that at industrial areas the risk of carcinogenesis to the associated population is mainly due to As contaminated dust exposure. Hair (60%) and nail samples (70%) collected from industrial land use were found above the WHO threshold limit of 1 mg/kg, suggested high risks for human health in the studied area. The results of the present study would be useful for assessing the human health risks due to arsenic contamination via dust exposure in different parts of country.

1. Introduction

Arsenic (As) is a toxic metalloid with ubiquitous distribution into the environment (Hinwood et al., 2003; Silbergeld and Nachman, 2008), which originates either from anthropogenic and/or geogenic sources (Farooqi et al., 2007) and has been classified as Group 1 human carcinogen by the International Agency for Research on Cancer (Huang et al., 2014). It has been widely reported that human exposure to arsenic is of particular apprehension due to its several adverse health effects, caused via direct routes including ingestion of contaminated water and/or food (Farooqi et al., 2007; Waheed et al., 2013) and indirectly through the dust (Huang et al., 2014). Consumption of arsenic contaminated drinking water is one of the major public health concerns across the world (Farooqi et al., 2007). Nevertheless, in recent years several technological advances make it feasible to remediate the arsenic contaminated water and installation of arsenic removal units has reduced the arsenic exposure to the consumers (Katsoyiannis and Zouboulis, 2006; Katsoyiannis et al., in press). However, some additional sources of arsenic exposure have been recently reported, e.g., dust-borne contamination via ingestion, inhalation and dermal contact, especially to population inhibiting in the vicinity of urban hubs and industrial sites (Farooqi et al., 2007; Waheed et al., 2013; Huang et al., 2014). Hence, the situation gets further aggravated in the arid and semi arid regions such as Pakistan, due to low vegetation cover and high dust storm frequencies (Hussain and Mudasser, 2007).

In South Asian countries such as India, Bangladesh and

* Corresponding author.
** Corresponding author. Fax: +92 51 4442805.
E-mail addresses: hqshen@iue.ac.cn (H. Shen), ali_ebl2@yahoo.com (S.A.M.A.S. Eqani).
[1] Authors contributed equally.

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Pakistan, geological conditions are considered as one of the prime source of As exposure into the environment (Farooqi et al., 2007; Kazi et al., 2009). Several studies have reported As contamination of ground and surface water, soils and plants (Silbergeld and Nachman, 2008; Arain et al., 2009; Baig et al., 2011; Liu et al., 2010). Moreover, sorbed As onto parent rock material can be distributed into wider areas of Pakistan through flood drifts and/or seepage of ground water (Farooqi et al., 2007). However, atmospheric deposition and re-suspension of dust particles from dried soil surfaces cause dispersion of As into remote areas (Zheng et al., 2010). It has also been reported that non-dietary human As exposure through dust particles can elevate the body metal burden via inhalation, ingestion and/or dermal contact (Ferreira-Baptista and de Miguel, 2005); and might cause deleterious health effects including arsenicosis, hypo and hyper-pigmentation of skin, keratosis (ATSDR 2000; Farooqi et al., 2007) and cardiovascular diseases (Chiou et al., 1997; Tseng et al., 2003).

In past several biomarkers e.g., blood, urine, nail and hair etc., have been used to monitor As exposure into associated human population (Liu et al., 2010; Hinwood et al., 2003). Nevertheless, several studies have reported As levels in hair and nail along with water and soil of different areas (Tsuji et al., 2005; Chiou et al., 1997; Lin et al., 1998) but very few studies has reported their association with dust and air particles (Huang et al., 2014; Tsuji et al., 2005). Similarly, a number of studies have reported As concentrations in human nail and hair, and reported them as an effective bio- monitoring tool for As exposure (Cottingham et al., 2013; Rakib et al., 2013; Samanta et al., 2004; Sanz et al., 2007; Anwar, 2005), due to their keratin tissue composition, containing high cystein residues that have affinity for diverse metals transported via body fluids, thus become part of these metabolic inactive tissues (Tsuji et al., 2005 and Huang et al., 2014). Hence, once As is accumulated into these non-invasive tissues, can easily be collected and used to monitor past As exposure into human body (Gualt et al., 2008; Hinwood et al., 2003). Therefore, dust exposure along with bio monitoring of hair and nail is an effective measure to assess mechanistic As toxicity in our environment.

To the best of our knowledge this is the first systematic study in Pakistan that has reported outdoor As dust concentrations and its association with biological samples (i.e., hair and nail of associated local population). Thus, in the present this study, we considered As contaminated dust to be one of the major sources of human exposure to arsenic. This was further validated by analyzing paired human hair and nail samples of affected population at similar areas. The main objectives of this study were: (a) to monitor As levels in dust and associated population from different land-use settings Viz; Rural, urban, and Industrial; (b) to compute human health risk estimation and major exposure routes via As contaminated dust exposure into local population.

2. Materials and methods

2.1. Study location

This study was conducted at two districts i.e., Lahore and Sargodha of Punjab province, Pakistan (Fig. 1). On the eastern side of district Lahore, River Ravi flows, one of the most polluted rivers of Punjab province, because of direct waste water discharge by water and sanitation agency (WASA) from different local drains and nallas. Apart from that, untreated effluent discharge from industrial zones including leather and paint industries are the main anthropogenic sources of metals contamination, including As, which ultimately penetrates into the groundwater aquifers (Farooqi et al., 2007; Mahmood et al., 2014). Whereas, district Sargodha is categorized as a saline hilly area, having parent sedimentary rocks rich in salt and minerals. Therefore, As contamination in Sargodha area is mainly of geogenic origin, attributed to the geographical characteristics. However, brick kilns and stone crashing industries are additionally contributing to arsenic burdens of the population living in the vicinity of these industries via contaminated dust exposure (Aslam et al., 1994).

The study area has also further divided into Rural, Urban and Industrial areas. Fig. 1 and Table S1 showed the map of the study area along with the sample collection sites. In this study, we collected rural dust and associated human hair and nail samples from two rural sites between Lahore and Sargodha city. Similarly, we selected urban spots from both Sargodha city and from Lahore city to collect all the sample types. However, most of industrial samples were collected from the famous industrial zone of Lahore and few industrial samples were also collected nearby Sargodha city, where many brick kilns are situated and known as important source of Arsenic, as coal has used as a source of energy in this area.

2.2. Sample collection

Hair ($n=30$) and nail ($n=30$) samples were collected from 30 different individuals along with composite dust samples ($n=22$) from corresponding studied areas, which were categorized as urban, rural and industrial sites. Using non-metallic scissors, 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 (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. After collection, samples were wrapped in aluminum foils and sealed into coded air tight polythene zipper bags and stored at -20 °C. The dust samples were collected by brushing the sample surface and to avoid cross-contamination between sampling locations, brushes from the respective household were used to collect the dust samples. After brushing, dust was wrapped in aluminum foil, sealed in polyethylene zip bags and stored at room temperature. The collected dust samples were of fine particle size (<50 μm) which can be trapped through inhalation, settled down from air. All the collected samples were safely transported to the laboratory and stored at 4 °C prior to analysis.

2.3. Analytical methodology

Hair and nail samples were washed according to International Atomic Energy Agency (IAEA 1985) protocol. The protocol includes the sequential washing with water, and acetone (extra pure, Merck). The washed samples were dried at 50 °C in a drying oven. Prior to digestion, all the tubes were soaked overnight (v/v) with 10% HNO_3 and rinsed thrice with ultra pure water Milli Q system A (Millipore Corporation, Billerica, MA, USA). Aliquot (0.1–0.5 g) of each dried and cleaned hair, nail and dust sample was weigh carefully and then digested with 1 mL GR grade 65% (v/v) HNO_3 (CNW Corporation, Shanghai, China) overnight followed by 1 mL GR grade 30% (v/v) H_2O (Sinopharm Chemical Reagent Co., Ltd, Beijing, China) at the next day. Finally, all samples were thoroughly 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. After cooling for 30 min, the vessels were opened carefully and each digested solution was transferred to a 5 mL volumetric flask. Then 100 μL of an internal standard mixture (10 ng/l to 2 ng/l of each) was added for As and the recoveries were recorded by using these samples. Samples were