Assessment of Arsenic and Heavy Metals Pollution in Chhattisgarh, India

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ABSTRACT

Ambagarh *tehsil*, in Rajnandgaon, central India, is a heavily polluted area. In this work, contamination with As and other toxic elements (Cr, Mn, Cu, Zn and Pb) has been monitored in water, soil, plant leaves and animal stools. Mean concentrations of total As in surface water, groundwater, surface soil, plant leaf and animal stool samples of 0.031±0.005 mg·mL^{-1}, 0.360 ±0.050 mg·mL^{-1}, 192±28 mg·kg^{-1}, 5.6±1.4 mg·kg^{-1} and 51±7 mg·kg^{-1}, respectively, were found. The speciation, sources and toxicities of the As and other metals are discussed, together with some associated health hazards, exemplified in domestic animals exposed to the contaminated water and food.

INTRODUCTION

Arsenic is a carcinogenic environmental and occupational pollutant, known to be very hazardous to health (Hong et. al., 2014). Arsenic pollution is prevalent in numerous countries, including Mexico, Chile, Argentina, India, Taiwan, Bangladesh and Myanmar (Eisler, 2004), and studies on contamination with arsenic and other toxic metals has been widely reported in the literature (Bandaru et. al., 2016; Belluck et. al., 2003; Bhattacharya et. al. 2010; Chaurasia, 2012; Karimi and Alavi, 2016; Lambrou et. al., 2012; Loeppert, 2010; Middleton et. al., 2017; Moreno-Jiménez et. al., 2012; Rahman and Hasegawa, 2011; Roychowdhury, 2008).

In the case of central India, arsenic contamination has been reported at hazardous levels over an area of 3000 km^2 in Rajnandgaon district (Chhattisgarh), and it has been shown to cause serious health hazards due to polluted water and food consumption (Pandey et. al., 2002; Patel et. al., 2005; Pandey et. al., 2007). As regards the sources of contamination, in the case of arsenic it appears to be mainly geogenic (Acharrya et.al 2001 and 2005; Pandey et. al 2002), while heavy metals pollution would be originated by the coal-based thermal power plants and integrated iron and steel industries in the area (Tiwari et al. 2015; Sajal & Towari 2014).

In this work, contamination with several toxic elements (As, Cr, Mn, Cu, Zn and Pb) has been investigated in which is believed to be the most contaminated site in this region (Ambagarh *tehsil*) by monitoring the concentrations of these pollutants in water, soil, plants and animals. In the latter, since excess metals are excreted out through urine and stool, stool samples have been used as bio-indicators (Sawidis et. al., 2011; Gupta, 2013).

MATERIALS AND METHODS

Study Area

Samples of surface water (SW), groundwater (GW) and surface soil (SS) were collected from 20 locations in 13 villages in the Ambagarh *tehsil*, namely Murethitola, Netamtola, Kaudikasa, Sonsaitola, Joratarai, Thailitola, Jadutola, Arajkund, Bihari Khurd, Sangali, Mangatola, Dhaditola and Meregaon (**Figure 1)**. Plant and animal samples were collected only from the most contaminated location, after the water and soil pollution screening: Koudikasa village.

Sampling procedure

The selected environmental samples (water, soil, plant leaves and animal stools) were collected in February 2017 using well-established methodologies (references). Details of the sampling locations and number of samples are provided in Table X.

Surface water was collected from tanks and canals, and the groundwater samples were taken from hand pumps in twice-cleaned 1 L polyethylene flasks (Nielsen & Nielsen 2006). The surface soil samples (≈1 kg, from a depth of 0–10 cm) were collected from the agricultural fields and stored in polyethylene bags.

Leaves from 45 plants commonly used for animal feeding in this area (detailed in **Table 3**, with 3 replicates per type of plant; ABCD g/sample) were manually collected from Koudikasa village and placed in polyethylene bags. They were thoroughly washed with deionized water, and dried in a glassroom for one week. Early morning stools samples from various animals (cow, buffalo, goat and sheep; number of samples/type of animal; ABCD g/sample) were collected in a polyethylene bags and dried in the sunlight in similar way. Solid samples from both plants and animals were further dried in a hot air oven at 50 °C for 24 h, they were crushed, and particles < 0.1 mm were sieved out.

Analyses

Water parameters (viz. pH, dissolved oxygen (DO), reduction potential (RP), electrical conductivity (EC) and total dissolved solid (TDS)) were measured at the spot using HANNA Instruments (Woonsocket, RI, USA) sensors. Total dissolved solid (TDS) were determined by evaporation of the water samples, previously filtered through a glass fiber filter, and by drying until constant weight, according to method 2540 D (APHA 2005).

For the monitoring of metals contents, the acid extraction procedure was used, according to method ABCD (reference): 0.25 g of solid sample were digested with aqua regia (3.0 mL HCl and 1.0 mL HNO_3) in a P/T MARS microwave oven (CEM, Matthews, NC, USA). The total Arsenic content was determined by hydride generation-atomic absorption spectrophotometry (HG-AAS) with a ABCD (Manufacturer, Location) apparatus. Other HMs (Cr, Mn, Cu, Zn and Pb) were analysed by graphite furnace-spectrophotometry (GF-AAS) and inductively coupled plasma–atomic emission spectrometry (ICP-AES) with a ABCD (Manufacturer, Location) and a ABCD (Manufacturer, Location), respectively. Four reference materials were used to check the quality of data: BCR 143 (sewage sludge), LGC 6138 (coal carbonisation site soil), NIST 1633b (coal fly ash) and NIST-1515, USA (apple leaf).

For arsenic speciation measurements, ABCD method (reference) was followed: 5.0 mL of 0.5 mol/L H_3PO_4 were added to 0.2 g of sample in centrifuge tubes, which were shaken overnight so that the acid and the sample reacted completely, ensuring good As extraction. The next day, the samples were taken off the shaker and placed in the centrifuge for 10 min at ABCD rpm. The extract was filtered using 0.45 µm PTFE disc filters. 2.0 mL aliquots were taken from each sample and were then diluted to 10 mL with sodium phosphate buffer (filtered), which was the mobile phase for HPLC. High-Performance Liquid Chromatography–Hydride Generation-Atomic Fluorescence Spectrometry (HPLC-HG-AFS) was used to detect the levels of As(III), As(V), monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA) present in the samples, according to the procedure described in [REFERENCE].

Apropos of quality control/quality assurance, standard calibration curves were prepared for the analysis of As and other elements ~~in the water samples~~ using the multielement standard solution (Sigma-Aldrich) for analysis. ~~The relevant reference material was used for analysis of the elements in the solid samples~~.

Average values of three analyses are presented. The confidence limit at 95% probability (p = 0.05) was used to denote the variability of the dataset.

RESULTS AND DISCUSSION

Pollutant contents in water

The concentrations of As Cr, Mn, Cu, Zn and Pb in surface water (SW) and groundwater (GW) are summarized in **Table 1**. The SW featured pH, DO, RP, EC and TDS values of 7.7±0.3, 6.4±0.2 $mg \cdot L^{-1}$, 523±23 mV, 619±15 $\mu S \cdot cm^{-1}$ and 780±25 $mg \cdot L^{-1}$, respectively. Slightly different physical characteristics were observed for GW, with pH, DO, RP, EC and TDS values of 7.2±0.1, 6.0±0.2 $mg \cdot L^{-1}$, 475±23 mV, 853±20 $\mu S \cdot cm^{-1}$ and 1171±30 $mg \cdot L^{-1}$, respectively. The concentrations of As, Cr, Mn, Cu, Zn and Pb in the SW samples were in the 0.017–0.048, 0.015–0.43, 0.089–0.165, 0.021–0.062, 0.033–0.110 and 0.003–0.006 $mg \cdot L^{-1}$ range, with mean values of 0.031±0.004, 0.055±0.039, 0.132±0.009, 0.036±0.004, 0.080±0.010 and 0.011±0.005 $mg \cdot L^{-1}$, respectively. Higher concentrations of arsenic and heavy metals were found in the GW samples, probably due to mineralization from bedrock, with values in the range of 0.187–0.582, 0.019–0.090, 0.156–0.310, 0.029–0.071, 0.080–0.142 and 0.008–0.031 $mg \cdot L^{-1}$ for As, Cr, Mn, Cu, Zn and Pb, respectively. Mean values of 0.360±0.50, 0.033±0.006, 0.238±0.018, 0.047±0.005, 0.114±.0.007 and 0.015±0.002 $mg \cdot L^{-1}$ were found for As, Cr, Mn, Cu, Zn and Pb. It is worth noting that the concentrations of As and Pb found in these water samples were significantly higher than the recommended limit of 10 $\mu g \cdot L^{-1}$ in drinking water (WHO, 2017). Moreover, As concentration in the water of the examined area was also higher than the values reported for other regions of the country (Chaurasia, 2012) and would be among the highest reported in the world (Mukherjee, A., Sengupta, M. K., Hossain, M. A., Ahamed, S., Das, B., Nayak, B., ... & Chakraborti, D. (2006). Arsenic contamination in groundwater: a global perspective with emphasis on the Asian scenario. Journal of Health, Population and Nutrition, 142-163) (Ahoulé, D.G., Lalanne, F., Mendret, J. et al. Water Air Soil Pollut (2015) 226: 302. http://dx.doi.org/10.1007/s11270-015-2558-4). According to the thorough bibliographic survey presented in the review paper by Shankar *et al*. (Shiv Shankar, Uma Shanker, and Shikha, "Arsenic Contamination of Groundwater: A Review of Sources, Prevalence, Health Risks, and Strategies for Mitigation," The Scientific World Journal, vol. 2014, Article ID 304524, 18 pages, 2014. https://doi.org/10.1155/2014/304524), Ambagarh *tehsil* would suffer one of the most severe cases of As contamination of groundwater across the globe (although even higher concentrations have been reported for the aquifers of Noakhali in Bangladesh, Rupandehi in Nepal, Ron Phibun in Thailand, and Muzaffargarh in Pakistan).

Pollutant contents in surface soil

The concentrations of As and other metals in the surface soil samples are shown in **Table 2**. The As, Cr, Mn, Cu, Zn and Pb contents were in the following intervals: 58–302, 45–146, 659–986, 40–105, 57–114 and 17–52 $mg \cdot kg^{-1}$, respectively, with average values of 192±28, 99±13, 795±44, 59±8, 80±4 and

31±3 mg·kg^{-1}, respectively. As and Mn were found in the soil at remarkably high concentrations (>value and >value mg·kg^{-1}, respectively [reference]), whereas other metals (Cr, Cu, Zn and Pb) were present at moderate levels (range in mg·kg^{-1} [reference]). In particular, the As concentration detected in the soil was at least an order of magnitude higher than the acceptable background levels of 5.0 mg·kg^{-1}, and among the highest reported the literature in India (Shrivastava, A., Ghosh, D., Dash, A. et al. Curr Pollution Rep (2015) 1: 35. http://dx.doi.org/10.1007/s40726-015-0004-2) and in other regions of the world (Belluck et. al.,–2003; Gillispie, E.C., Sowers, T.D., Duckworth, O.W. et al. Curr Pollution Rep (2015) 1: 1. http://dx.doi.org/10.1007/s40726-015-0001-5), only exceeded by some locations in Greece (up to 513 mg/kg; Casentini B, Hug S, Nikolaidis N. Arsenic accumulation in irrigated agricultural soils in Northern Greece. Sci Total Environ. 2011;409(22):4802–10) and some heavily polluted industrial areas in the USA (>1000 mg/kg; Smith, Euan Robert George, R. Naidu, and A. M. Alston. Arsenic in the soil environment. Diss. Academic Press, 1998) and in China (e.g., in Linfen area).

Pollutant contents in plant samples

As and other HMs contents in plant leaves are presented in **Table 3**. The concentrations varied from 0.3 to 27 mg·kg^{-1} (As), from 3.3 to 25 mg·kg^{-1} (Cr), from 18 to 159 mg·kg^{-1} (Mn), from 9.1 to 62 mg·kg^{-1} (Cu), from 29 to 238 mg·kg^{-1} (Zn), and from 0.8 to 8.0 mg·kg^{-1} (Pb). Mean concentration values of 5.6±1.4, 11±2, 89±10, 27±4, 86±12 and 3.7±0.6 mg·kg^{-1} were detected for As, Cr, Mn, Cu, Zn and Pb, respectively. The maximum accumulation of As, Cr, Mn, Cu, Zn and Pb in the leaves corresponded to leaves from *Vigna unguiculata, Diospyros melanoxylon, Mangifera indica, Hibiscus* sabdariffa, *Moringa oleifera* and *Zingiber officinale*, respectively, suggesting their applicability as bio-indicators for aforementioned elements.

The enrichment factors with respect to the soil mean values were in the following ranges: 0.001–0.130 (As), 0.055–0.417 (Cr), 0.020–0.178 (Mn), 0.108–0.738 (Cu), 0.309–2.53 (Zn) and 0.027–0.267 (Pb). Mean enrichment factor were found to be: 0.027±0.007, 0.099±0.011, 0.320±0.045, 0.913±0.132 and 0.124±0.021 for As, Cr, Mn, Cu, Zn and Pb, respectively. It is worth noting that the enrichment factor for As was the lowest one (i.e., enrichment factors for Cr, Mn, Cu, Zn and Pb were found to be 7.1, 3.7, 11.9, 33.8 and 4.6 times higher than that of As), while that for Zn was the highest. In this regard, leaves from several plants (*Abelmoschus esculentus, Acacia concinna, Aegle marmelos, Amaranthus spinosus, Dioscorea bulbifera, Hibiscus sabdariffa, Lablab purpureus, Moringa oleifera, Psidium guvava, Solanum lycopersicon, Tectona grandis, Trigonella foenum-graecum* and *Vigna radiate*) were identified as potential Zn hyperaccumulators (**Table 3**).

Apropos of As concentration in the plant leaves collected, it was much higher than values reported in other regions of India and the world (Bandaru et. al., 2016; Bhattacharya et. al. 2010; Karimi and Alavi, 2016; Lambrou et. al., 2012; Loeppert, 2010; Rahman and Hasegawa, 2011; Roychowdhury, 2008).

Pollutant contents in animal faecal samples

The concentrations of As, Cr, Mn, Cu, Zn and Pb in stool samples was found to be in the following ranges: 41–60, 33–50, 212–301, 840–1220, 429–630 and 39–84 $mg \cdot kg^{-1}$, respectively, with mean values of 51±7, 39±6, 254±34, 1003±120, 529±71 and 63±14 $mg \cdot kg^{-1}$, respectively (see **Table 4)**. The highest concentration of toxic elements (As, Cu and Pb) was found in the goat stool samples. The elements were noticeably enriched in the faecal samples as compared to their average contents in the leaves (5.6, 11.4, 89, 27, 86 and 3.7 $mg \cdot kg^{-1}$ for As, Cr, Mn, Cu, Zn and Pb, respectively), at least by a factor of 9.1, 3.4, 2.8, 37.6, 5.9 and 16.8, respectively. Among them, two metals (Cu and Pb) was extremely enriched (>10-fold) in the stools; other elements (As and Zn) were moderately enriched (>5-fold); and Cr and Mn were poorly enriched (>2 - <5-fold) in the faecal samples [reference].

Arsenic speciation

The concentration of the different arsenic species found in the soil and stool samples are shown in **Table 5**. In general, inorganic As(III) and As(V) are more toxic than the organic As to the environment. In all samples, the concentration of As(V) was found to be the dominant species, ranging from 92.2 to 99.2%. The percentage concentration of As(III) in the soil and the stool samples of cow and buffalo was relatively low, ranging between 1.2 and 4.8%. Similarly, low concentrations of dimethyl arsenic (DMA) in the stool samples (except in the goat sample) were found, ranging from 2.9 to 7.2%. In all stool samples, monomethyl arsenic (MMA) was detected in trace quantities, ranging from 0.6 to 1.6%. No organic species (i.e., MMA and DMA) were detected in the soil samples.

Correlation coefficients

The correlation coefficients (r^2), presented in **Table 6**, were computed using the mean values of the elements (As, Cr, Mn, Cu, Zn and Pb) in the SW, GW, soil, leaves and stool samples. They showed high correlations among themselves ($r^2 = 0.91$-0.99), except for Zn, which showed moderate correlations ($r^2 = 0.42$–0.71). This suggests that they may share a common origin.

Toxicities

Arsenic contents in all samples were several times higher than the permissible limit (0.1 mg/kg) (FAO/WHO, 2010). Similarly, leaves showed heavy metals contents higher than the permissible limits in food (20 $mg \cdot kg^{-1}$ for Zn, 2.3 $mg \cdot kg^{-1}$ for Cr, and 0.3 $mg \cdot kg^{-1}$ for Pb, according to FAO/WHO, 2010). In particular, extremely high contents of As, Cr, Mn and Cu were detected in mango leaves **(Table 3)**. High Pb contents (>7.0 mg/kg) were accumulated in the leaves from *Aegle marmelos, Cynodon dactylon, Curcuma longa, Foeniculum vulgare* and *Hibiscus sabdariffa* (**Table 3)**. Similarly, Cr, Mn, Cu and Zn were accumulated in the leaves from *Amaranthus spinosus, Diospyros melanoxylon* and *Shorea robusta*; *Amorphophallus paeoniifolius* and *Mangifera indica*; *Abelmoschus esculentus* and *Hibiscus sabdariffa*; and *Acacia concinna* and *Moringa oleifera*, respectively, at concentrations beyond their respective tolerance limits of 20, 150, 50 and 215 mg·kg^{-1} (**Table 3**).

Aforementioned As levels pose a serious health hazard, provided that the consumption of water and food polluted with As may result in chronic arsenicosis. This multisystem disorder leads to increased risks of skin cancer with altered functions of organs, and usually manifests in skin lesions such as hyperkeratosis and pigmentation changes, in humans and animals (Kapaj et. al. 2006).

Conclusions

The environment of Ambagarh *tehsil*, in Rajnandgaon, India, is heavily polluted with As and other toxic metals (Cr, Cu and Pb) at hazardous levels, as evidenced by surface water, groundwater, soil surface, plant and animal samples collected in the area. Contents higher than those reported in other areas of India and in other regions of the world were detected. Arsenic was found to exist mostly in the inorganic As(V) form. On the basis of As and HMs enrichment factors, leaves from *Vigna unguiculata, Dalbergia melanoxylon, Mangifera indica, Hibiscus sabdariffa, Moringa oleifera* and *Zingiber officinale* were found to be the most dangerous for cattle feeding. Pollution in water and plants was reflected in the contamination of faecal samples of cattle with high levels of As (>50 mg·kg^{-1}).

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