Human Health Risk Assessment due to Heavy Metals in Ground and Surface Water and Association of Diseases With Drinking Water Sources: A Study From Maharashtra, India

Govind Mawari¹, Naresh Kumar¹, Sayan Sarkar¹, Arthur L Frank², Mradul Kumar Daga³, Mongjam Meghachandra Singh⁴, Tushar Kant Joshi⁵ and Ishwar Singh⁵

¹Department Center for Occupational and Environment Health, Maulana Azad Medical College, New Delhi, India. ²Department of Environmental and Occupational Health, Drexel University, Philadelphia, PA, USA. ³Department of Internal Medicine and Infectious Disease, Institute of Liver and Biliary Sciences, New Delhi, India. ⁴Department of Community Medicine, Maulana Azad Medical College, New Delhi, India. ⁵Department of ENT, Maulana Azad Medical College, New Delhi, India.

ABSTRACT

BACKGROUND: Contamination of freshwater sources can be caused by both anthropogenic and natural processes. According to Central Pollution Control Board, Maharashtra along with 2 other states, contribute 80% of hazardous waste generated in India, including heavy metal pollution. Hence, it is important to quantify heavy metal concentrations in drinking water sources in such areas.

MATERIALS AND METHODS: Water samples were analyzed for toxic elements (F, As, Cd, Hg, Pb, Ni, Cu, Zn, Mn, and Cr) using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) Agilent 7500. Health risks due to ingestion and dermal contact was assessed. A total of 557 people were randomly selected, with consumers from all 4 types of water sources that is surface water, hand pump, wells, and municipal water. Spot urine samples were collected from 47 people after considering inclusion and exclusion criteria. Urine was collected for estimating mercury and arsenic levels in the study participants.

RESULTS: Arsenic contributes the most health risk from ingestion from water. Among surface water users, 14 people (32%) reported frequent loose stool (P-value < .05) (OR 2.5), and 11 people (23%) reported frequent abdominal pain (OR 1.9). Hand pump and well water users reported frequent abdominal pain (27%) (OR 1.4) and gastric discomfort (31%) (P-value < .05) (OR 3) respectively. The mean value of urinary Hg and As were 4.91 ± 0.280 and 42.04 ± 2.635µg/L respectively.

CONCLUSION: Frequent loose stool, gastric discomfort, and frequent abdominal pain were associated with the various sources of drinking water. Urine Hg levels were found higher than the NHANES (USA) Survey. It is recommended that frequent monitoring of drinking water should be enforced around the industrial hub, so that appropriate actions can be taken if present in excess.

KEYWORDS: Human health risk assessment, drinking water, heavy metals, urine arsenic, urine mercury

Introduction

Water of optimum quality is essential to human life, and water of acceptable quality is essential for agricultural, industrial, domestic, and commercial uses; in addition, water is also used for recreational activities. Therefore, major activities having potential effects on surface water are certain to be of appreciable concern.¹ The United Nations General Assembly (UNGA) has listed access to clean water and sanitation for all as one of the sustainable development goals to be attained by 2030.²

Contamination of freshwater sources can be caused by both anthropogenic and natural processes. Erosion, weathering, and other geological events are the natural sources of pollution,³ while human settlements, mining, industrial, and agricultural activities are among the anthropogenic factors.⁴ Heavy metals are also released into water bodies through sediment resuspension, desorption, reduction or oxidation reactions, and the degradation of organic tissues.⁵ All these factors increase the concentration of dissolved metals which may threaten the aquatic ecosystem and human health.⁶

Heavy metals are ubiquitous materials and prevalent contaminants in polluted environments, and their properties such as chemical stability, bioaccumulation, non-degradable nature, and long-lasting negative impacts have piqued public interest.⁷ Some dissolved metals are readily taken up by aquatic species and may enter the human body via drinking water, skin absorption, and ingestion of products, posing a health risk.⁸

DECLARATION OF CONFLICTING INTERESTS: The author(s) declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

CORRESPONDING AUTHOR: Mradul Kumar Daga, Internal Medicine and Infectious Diseases, Institute of Liver and Biliary Sciences, New Delhi 110070, India. Email: dmraduldaga@gmail.com
Cadmium alters the immune system and raises the incidence of pulmonary adenocarcinoma and proliferative prostatic lesions. Excessive blood lead can cause hypertension, weaken the skeletal, immunological, and endocrine systems, and reduce the intellectual capacity of children. It also disrupts renal and cardiac function in adults. For arsenic, inorganic compounds are more hazardous than organic compounds, while trivalent compounds are more dangerous than pentavalent or hexavalent compounds. Arsenic is a carcinogen that has several short- and long-term health effects on humans. Mercury causes widespread oxidative damage by increasing the production of reactive oxygen species (ROS). Zinc may cause infertility, renal problems, and CNS problems whereas copper induces depression and, in the long run may contribute to lung cancer. Chromium has been linked to cancer and tumors of the respiratory system. Therefore, assessment of heavy metals in drinking water is essential.

The hydrological environment is composed of 2 interrelated phases; groundwater and surface water. Impacts initiated in 1 phase eventually affect the other. For example, a groundwater system may charge one surface water system and later be recharged by another surface water system. The complete assessment of any impact dictates the consideration of both groundwater and surface water. Thus, pollution at one point in the system can be passed throughout, and consideration of only 1 phase does not characterize the entire problem.

Even though the majority of the world’s population has access to water, it is often not suitable for human consumption and is rarely available in adequate amounts to fulfill the basic health needs. This is now a worsening problem worldwide. According to the World Health Organization (WHO), over 1.1 billion people worldwide consume contaminated water, and the majority of diarrheal diseases (88%) are caused by contaminated water and poor sanitation. Diarrhea is a major cause of childhood deaths. Water scarcity and low quality have a significant influence on long-term development, particularly in developing nations. Hence, it is crucial to evaluate water quality and risk assessment regularly in developing nations like India, with rapid growth in industrialization and urbanization. According to the Central Pollution Control Board (CPCB), Maharashtra along with 2 other states, contribute 80% of hazardous waste generated in India, including heavy metal pollution. The study area, Solapur, which is located in the Indian state of Maharashtra is home to many textile industries. Previous studies have documented the role of textile industries in heavy metal pollution. The potential health risk due to environmental release of hazardous chemical stimulated interest in ascertaining the health status of inhabitants who drink from different water sources. Water sources were then screened for presence of toxic elements (Pb, As, Cd, Hg, Pb, Ni, Cu, Zn, Mn and Cr) following which the human risk assessment was carried out. Finally, the Hg and As content of urine samples from the participant were assessed in a bid to further explain the observed incidence of gastrointestinal disorders amongst the inhabitants of the region.

Materials and Method
Study area and sample collection

The study was conducted in the industrialized city of Solapur district, which is located in the Indian state of Maharashtra. Handlooms and power-looms are the main industries in this area. The city experiences a tropical monsoon type of climate. The monsoon lasts from June to October and brings about 87% of the annual rainfall. The average annual rainfall in the study area is around 677.7 mm.

Water samples were collected from 7 locations around a 10 km radius of the industrial hub for 2 seasons viz. winter (January) and spring (April) in the year 2017 to understand water quality (Figure 1). Samples were collected as per IS:3025 (Part 1) methodology. Necessary precautions were taken while collecting, preserving, and transporting the samples. Samples were analyzed for pH, Total Dissolved Solids (TDS), Dissolved Oxygen (DO), Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Fluoride (F), Ammonia (NH3), Mercury (Hg), Arsenic (As), Cadmium (Cd), Lead (Pb), Nickel (Ni), Copper (Cu), Zinc (Zn), Chromium (Cr6+), and Manganese (Mn).

One sample (SW-1) was collected from the Sina River, another sample (GW-2) was collected from tap water, and the other 5 samples (GW-1, GW-3, GW-4, GW-5, and GW-6) were collected from hand pumps of that area to understand the quality of surface, municipal and ground water respectively. The selection of the water samples has been considered as per the utilization for domestic and drinking purposes. Water samples were collected in 1L polypropylene bottles; pre-cleaned with 5% HNO3 followed by rinsing repeatedly with deionized water, stabilized with 0.5% HNO3 and transported to the laboratory.

Spot urine samples were collected from the subjects after considering inclusion and exclusion criteria. The urine was collected for estimating Hg and As levels in the study subjects. Only apparently healthy individuals were included in the study of urine Hg and As levels. Subjects under treatment for tuberculosis, cancer, and chronic heart, lung, or kidney ailments were excluded. Also, pregnant and lactating women were not included as these conditions might modify some of the measured parameters.

Sample preparation and analysis

Parameters such as pH, temperature, and DO were measured on site while collecting the water samples. All the rest of the parameters were analyzed in the laboratory as per “Methods of Sampling and Test (Physical and Chemical) for water and wastewater” IS: 3025 and “Standard Methods for the Examination of Water and Wastewater” APHA. Preparation of the samples were made by diluting 1 mL of water sample to
10 mL with 0.3% HNO₃, and then, samples were analyzed for Fluoride (F) and heavy metals (As, Cd, Hg, Pb, Ni, Cu, Zn, Mn, and Cr) using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) Agilent 7500.

For urinary Hg and As examination, a 20 mL aliquot of urine was kept in a metal-free container (Tarsons) after collection from subjects, and further analysis was made with the help of Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) Agilent 7500.

Human health risk assessment

Chronic daily intake (CDI). In the present study, we have assumed that ingestion and dermal contact were the routes of exposure. The Chronic Daily Intake (CDI) through surface and ground water ingestion and dermal absorption was calculated by using the following equation:

\[
\text{CDI}_{\text{ingestion}} = \left( \frac{C \times IR \times EF \times ED}{BW \times AT} \right)
\]

\[
\text{CDI}_{\text{dermal}} = \left( \frac{C \times EF \times ED \times ET \times SA \times KP \times CF}{BW \times AT} \right)
\]

Where, CDI = chronic daily intake (mg/kg/day), C = mean concentration of heavy metal in water (mg/L), IR = ingestion rate (L/d), EF = exposure frequency (days/year), ED = exposure duration (years), BW = average body weight over the exposure period (kg), and AT = average time period of exposure (days), ET = exposure time (hour/day), SA = surface area of contact (cm²), KP = dermal permeability coefficient (cm/h),²⁷ CF = unit conversion factor (0.001 L/cm³).²⁸

The following assumptions were made to quantify exposure: People residing in the study area are assumed to drink 2 L/d of water. EF is taken as 350 days because it was assumed that a person will leave the area for about 2 weeks per year. For non-carcinogens ED is taken as 1 year. AT is the period over which exposure is averaged (1 year = 365 days) for non-carcinogens. BW was assumed as 58 Kg. ET was taken as 0.58 hour/day and SA was 18,000 cm².

Hazard quotient (HQ) and hazard index (HI). To indicate the potential non-carcinogenic risk to human health posed by a hazardous material, the hazard quotient has been developed. It is the ratio of estimated heavy metals exposure of test water and oral reference dose. It indicates potential hazards to human health.
\[ HQ_{\text{ingestion}} = \frac{\text{CDI}_{\text{ingestion}}}{\text{RfD}_{\text{ingestion}}} \]
\[ HQ_{\text{dermal}} = \frac{\text{CDI}_{\text{dermal}}}{\text{RfD}_{\text{dermal}}} \]

Where, CDI = Chronic daily intake and RfD = Reference dose.

The oral reference dose (RfD) of F, As, Cd, Hg, Pb, Ni, Cu, Zn, Mn, and Cr are 0.06, 29 0.0003,30 0.001,30 0.0035,31 0.02,32 0.04, 0.3,32 0.014,32 and 0.003 mg/kg/day35 respectively. The dermal reference dose (RfD) of F, As, Cd, Hg, Pb, Ni, Cu, Zn, Mn, and Cr are 0.0582, 29 0.000123,32 0.00001,32 0.000021, 0.0000525,32 0.0054, 0.012, 0.06, 1.5 mg/kg/day29 respectively.

An index of equal to or more than 1 is considered as not safe for human health.32

To evaluate the potential risk to human health through more than 1 metal, a hazard index (HI) has been developed.36 The hazard index is the sum of the hazard quotients as described in the following equation.

\[ HI_{\text{ingestion}} = \sum HQ_{\text{ingestion}} \]
\[ HI_{\text{dermal}} = \sum HQ_{\text{dermal}} \]

It is assumed that the magnitude of the adverse effect will be proportional to the sum of multiple metal exposures. It also assumes similar working mechanisms that linearly affect the target organs.37 When the value of HI > 1, there is a greater possibility of non-carcinogenic health effects, and the probability increases with a rising value of HI.38 The hazard index for the elements F, As, Cd, Hg, Pb, Ni, Cu, Zn, Mn, and Cr through ingestion and dermal absorption has been calculated in the present study.

Cross-sectional health survey

A cross-sectional health survey was carried out in line with USEPA’s guidelines for Human Health Risk Assessment. The study area was limited to 25 kms around the industrial hub of Solapur. Stratified sampling techniques were used in the selection of villages, which were divided into various strata depending upon the direction of the fall out of pollutants from the industrial hub. A total of 557 people were randomly selected, with consumers from all 4 types of water sources that is surface water, hand pump, wells, and municipal water. To provide a more complete picture of major waterborne diseases, the questionnaire included both open-ended and closed-ended questions. Age, monthly income, education, smoking habits, body weight, drinking water source, employment, waterborne diseases, and human health risks data were collected and documented. The survey questionnaire is presented as Supplemental Material 1. The researchers conducted direct interviews with all survey respondents within the local communities in their native language and each participant were examined by medical professional to ascertain symptoms.

Table 1. Surface water characteristics.

| PARAMETERS | SW-1 | IS: 2296-1982 CLASS C NORMS |
|------------|------|---------------------------|
| pH         | 8.23 | 8.0                       | 6.5-8.5                    |
| TDS (mg/L) | 378  | 450                       | 1500                       |
| DO (mg/L)  | 6    | 4.4                       | 4                          |
| COD (mg/L) | 16   | 14                        | NS                         |
| BOD (mg/L) | 4    | 2.0                       | 3                          |
| NH₃ (mg/L) | Nil  | Nil                       | NS                         |
| F (mg/L)   | 0.4  | 0.9                       | 1.5                        |
| Hg (mg/L)  | <0.001 | <0.001                   | NS                         |
| As (mg/L)  | <0.05 | <0.05                     | 0.2                        |
| Cd (mg/L)  | <0.01 | <0.01                     | 0.01                       |
| Pb (mg/L)  | <0.05 | <0.05                     | 0.1                        |
| Ni (mg/L)  | <0.01 | <0.01                     | NS                         |
| Cu (mg/L)  | <0.05 | <0.05                     | 1.5                        |
| Zn (mg/L)  | <0.01 | <0.01                     | 15                         |
| Mn (mg/L)  | <0.01 | <0.01                     | NS                         |
| Cr (mg/L)  | <0.05 | <0.05                     | 0.05                       |

Fish survival after 96 h 90% 90% 90%

Results

Water parameters

The results of all the specified parameters were then compiled for both seasons and are tabulated in Tables 1 and 2. The water samples were observed to be neutral to slightly basic in nature with pH ranging from 7.06 to 8.23 for both seasons. For groundwater samples, TDS was between 410 and 1898 mg/L whereas for surface water was 378 and 450 mg/L for both seasons. F concentration ranged between 0.4 and 0.9 mg/L, Zn from 0.32 to 0.57 mg/L, and NH₃ was found to be <0.1 mg/L. No toxic compounds were observed in all 7 samples analyzed.

Human health risk assessment

For the heavy metals which are below detectable limits, the value of the detectable limit was considered for calculation. CDI and HQ for toxic elements F, As, Cd, Hg, Pb, Ni, Cu, Zn, Mn, and Cr are presented in Table 3. For reference, an index of more than 1 is considered not safe for human health.32 In our study, HQ of more than 1 was noted for As (HQ\text{ingestion As} = 5.5108) for both surface and groundwater ingestion and it
Table 2. Ground water and Municipal water (GW-2) characteristics.

| PARAMETERS | GW-1 | GW-2 | GW-3 | GW-4 |
|------------|------|------|------|------|
|            | WINTER | SUMMER | WINTER | SUMMER | WINTER | SUMMER | WINTER | SUMMER |
| pH         | 7.07   | 7.21  | 8.01  | 7.8   | 7.25   | 7.4    | 7.13   | 7.5    |
| TDS (mg/L) | 1810   | 1860  | 821   | 864   | 465    | 590    | 1190   | 1206   |
| DO (mg/L)  | 4      | 0.8   | 4.5   | 3.7   | 5      | 3.4    | 3.5    | 2.8    |
| COD (mg/L) | 116    | 95    | 20    | 12    | 4      | 16     | 12     | 12     |
| BOD (mg/L) | 24     | 12    | 4     | 2.0   | Nil    | 3.0    | 4      | 2.0    |
| NH3 (mg/L) | 0.1    | <0.1  | 0.1   | <0.1  | 0.1    | <0.1   | 0.1    | <0.1   |
| F (mg/L)   | 0.5    | 0.9   | 0.8   | 0.8   | 0.8    | 0.9    | 0.9    | 0.8    |
| Hg (mg/L)  | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 |
| As (mg/L)  | <0.05  | <0.05 | <0.05 | <0.05 | <0.05  | <0.05  | <0.05  | <0.05  |
| Cd (mg/L)  | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 | <0.003 |
| Pb (mg/L)  | <0.01  | <0.01 | <0.01 | <0.01 | <0.01  | <0.01  | <0.01  | <0.01  |
| Ni (mg/L)  | <0.01  | <0.01 | <0.01 | <0.01 | <0.01  | <0.01  | <0.01  | <0.01  |
| Cu (mg/L)  | <0.05  | <0.05 | <0.05 | <0.05 | <0.05  | <0.05  | <0.05  | <0.05  |
| Zn (mg/L)  | <0.01  | 0.57  | <0.01 | 0.32  | <0.01  | 0.45   | <0.01  | 0.32   |
| Mn (mg/L)  | <0.01  | <0.1  | <0.1  | <0.1  | <0.01  | <0.1   | <0.01  | <0.1   |
| Cr (mg/L)  | <0.05  | <0.05 | <0.05 | <0.05 | <0.05  | <0.05  | <0.05  | <0.05  |
| Fish survival after 96 h | 70% | 75% | 90% | 90% | 90% | 90% | 90% | 90% |

| PARAMETERS | GW-5 | GW-6 | IS: 10500-2012 LIMITS |
|------------|------|------|------------------------|
|            | WINTER | SUMMER | DESIRABLE | PERMISSIBLE |
| pH         | 7.06   | 7.30  | 7.25   | 7.35 | 6.5-8.5 | No Relaxation |
| TDS (mg/L) | 1465   | 410   | 1898   | 1620 | 500 | 2000 |
| DO (mg/L)  | 3.5    | 3.1   | 3.2    | 2.6  | NS | NS |
| COD (mg/L) | 38     | 12    | 42     | 14   | NS | NS |
| BOD (mg/L) | 6      | 2.0   | 8      | 2.0  | NS | NS |
| NH3 (mg/L) | 0.1    | <0.1  | 0.2    | <0.1 | NS | NS |
| F (mg/L)   | 0.9    | 0.8   | 0.8    | 0.9  | 1  | 1.5 |
| Hg (mg/L)  | <0.001 | <0.001 | <0.001 | <0.001 | 0.001 | No Relaxation |
| As (mg/L)  | <0.05  | <0.05 | <0.05  | <0.05 | 0.01 | 0.05 |
| Cd (mg/L)  | <0.003 | <0.003 | <0.003 | <0.003 | 0.003 | No Relaxation |
| Pb (mg/L)  | <0.01  | <0.01 | <0.01  | <0.01 | 0.01 | No Relaxation |
| Ni (mg/L)  | <0.01  | <0.01 | <0.01  | <0.01 | NS | NS |
| Cu (mg/L)  | <0.05  | <0.05 | <0.05  | <0.05 | 0.05 | 1.5 |
| Zn (mg/L)  | <0.01  | 0.53  | <0.01  | 0.41 | 5 | 15 |
| Mn (mg/L)  | <0.01  | <0.1  | <0.1   | <0.1 | 0.1 | 0.3 |
| Cr (mg/L)  | <0.05  | <0.05 | <0.05  | <0.05 | 0.05 | No Relaxation |
| Fish survival after 96 h | 90% | 90% | 90% | 90% | 90% | 90% |
was found to be the major contributor in HI ingestion. The HQ of metals via intake of surface water follows the order: As > Cr > Pb > Cd > F > Cu > Mn > Ni > Hg > Zn, whereas ground-water follows the order: As > Cr > F > Cd > Pb > Cu > Mn > Ni > Hg > Zn (Figure 2). The HI of metals via intake of surface and groundwater are 7.1712 and 6.7823 respectively, whereas dermal absorption is 0.3042 and 0.1714 respectively. In the case of dermal absorption, it was found that in surface water HQCd > HQAs. The calculated HI of metal through ingestion and dermal contact signifies that the major health risk posed to the community living around the industrial hub is through ingestion of heavy metal contaminated drinking water.

Demographic overview

For the human epidemiological study, 557 people were recruited. Characteristics of the study population were presented in Supplemental Table 1. 68.2% of the participants were male. 55% of the participants had at least 10 years of formal education. 63.7% of the participants were currently using tobacco products. 12.9% of the participants drank alcohol.

Source of drinking water and prevalence of various diseases

A cross-sectional health survey was conducted within the study area to document the source of drinking water used and the prevalence of various symptoms and diseases such as cough, gastric discomfort, jaundice, frequent loose stools, frequent abdominal pain, and infectious diseases.

Out of 557 people, 43 people (7.7%) used surface water, 194 people (34.8%) used hand pump water, 64 people (11.5%) used well water, and 256 people (46%) used municipal water as their source of drinking water.

The data regarding the source of drinking water and the prevalence of various diseases are tabulated in Table 4 and Figure 3. Among surface water users, 14 people reported frequent loose stools (P-value < .05) (OR 2.5), and 11 people reported frequent abdominal pain (OR 1.9). Hand pump and well water users more frequently reported frequent abdominal pain (OR 1.4) and gastric discomfort (P-value < .05) (OR 3) respectively.

Urinary arsenic and mercury monitoring

For estimating Hg and As excretion through urine, urine samples were analyzed. Forty-seven subjects were selected after considering exclusion and inclusion criteria. The results of urinary Hg and As are demonstrated in Table 5. The mean value of urinary Hg and As are 4.91 ± 0.280 and 42.04 ± 2.635 µg/L respectively. No significant difference was found with respect to As and Hg levels in urine samples in the representative 47 samples with P-value being .854 and .431 for As and Hg respectively.

Discussion

This study provides information about the quality of drinking water used by the communities surrounding the industrial hub of Solapur. Sources of drinking water and prevalence of various symptoms and diseases were collected to see any correlation between water use and disease prevalence in the community. Furthermore, urinary As and Hg excretions were recorded to understand As and Hg exposure in the communities surrounding the industrial hub.

In the study area, the assessment of water quality was performed to understand its suitability for drinking, agricultural and domestic purposes. All the water parameters were within the Indian standards for ground and municipal water. TDS is
Figure 2. HQ ingestion and HQ dermal of ground and surface water.
composed of a variety of salts, including those of Ca, Mg, Na, K, and other elements, as well as carbonates, bicarbonates, chlorides, sulfates, phosphates, and nitrates. WHO has not defined its health-based limit in drinking water, because TDS occurs in drinking water well below its toxic levels. However, water with TDS level of less than 500 mg/L is generally considered to be good. Water becomes significantly and progressively unpalatable at TDS levels greater than 1000 mg/L. Consumers may find TDS beyond 1200 mg/L undesirable, and it may affect people who need to restrict their daily salt intake, for example diabetic, severely hypertensive, and dialysis patients. In groundwater samples (GW-1, GW-4, GW-5, GW-6) TDS was recorded above 1200 mg/L.

The main route of elimination of many metals from the human body is through urine, and urinary levels of metals have been used in demonstrating previous exposure within a few hours to days of ingestion. In the present study, we measured As and Hg levels in urine samples. The mean value of As in the urine sample was 42.04 µg/L, with a range of 10.00 to 82.00 µg/L. The result in the present study for urinary As levels were higher than the studies from mining areas in Zimbabwe and Guatemala. Urinary As levels in Michigan urban anglers were found lower than the present study. However, our results were lower than the studies conducted in mining areas of Ghana. The mean value of Hg in the urine sample was 4.91 µg/L, with a range of 1.00 to 10.00 µg/L. The results of urinary Hg were higher than those found in mining areas of Guatemala and Michigan urban anglers. Relatively similar results were reported regarding urinary Hg levels from mining areas of Zimbabwe and Colombia. However, our results were lower than the study conducted on non-occupationally exposed Indians by Panday et al. Our results for urinary Hg are higher than the NHANES Study in the US (Mean urinary Hg and As are 1.76 and 49.9 µg/L respectively). Human exposure to As and Hg can be from various routes including ingestion of contaminated food or water, inhalation, or dermal contact. Consumption of fish is the most common route of As and Hg exposure worldwide. Another probable cause of As exposure may be through drinking water. In our study, As and Hg in drinking water were found below detectable limits. So, other exposure routes should be investigated. A previous study from the same location as present study reported that As concentration in fruits and vegetables were within permissible limits. However, in garlic (Allium sativum) (0.123 mg/kg), okra (Abelmoschus esculentus) (0.17 mg/kg), fenugreek (Trigonella foenum-graecum) (0.235 mg/kg), sugarcane (Saccharum officinarum) (0.035 mg/kg), tamarind (Tamarindus Indica) (0.147 mg/kg), and sorghum (Sorghum arundinaceum) (0.356 mg/kg) Hg concentration exceeds WHO/FAO standard (0.03 mg/kg). This might be one of the reasons for mercury exposure in the study population.

In many parts of the world, drinking water is still a major contributor to the community burden of enteric disease

| Sources of Drinking Water | No. of People (%) | Cough OR | Gastric Discomfort OR | Jaundice OR | Frequent Loose Stool OR | Frequent Abdominal Pain OR | Diabetes OR | Tuberculosis OR |
|--------------------------|------------------|----------|-----------------------|------------|------------------------|--------------------------|------------|---------------|
| Surface water            | 43 (77)          | 2.2 (0.957-4.748) | 0.9 (0.350-2.220)     | 1.4 (0.919-2.339) | 0.6 (0.265-1.726) | 1.9 (0.916-3.911) | 0.6 (0.128-3.410) | 0.4 (0.128-3.410) |
| Hand Pump                | 194 (34.8)       | 0.9 (0.507-1.528) | 0.7 (0.398-1.114)     | 0.9 (0.367-1.997) | 0.5 (0.235-0.907) | 1.4 (0.919-2.339) | 0.6 (0.128-3.410) | 0.4 (0.128-3.410) |
| Wells water              | 64 (11.5)        | 3.1 (1.596-5.717) | 0.3 (0.143-0.949)     | 1.2 (0.657-2.349) | 0.5 (0.210-1.066) | 1.4 (0.919-2.339) | 0.6 (0.128-3.410) | 0.4 (0.128-3.410) |
| Municipal water          | 296 (46)         | 0.4 (0.253-0.780) | 0.9 (0.572-1.457)     | 1.4 (0.621-2.305) | 1.1 (0.712-1.708) | 0.7 (0.447-1.223) | 0.7 (0.447-1.223) | 0.7 (0.447-1.223) |
because available water sources are fecally contaminated or industrial contaminants make it unfit. When it comes to the severity and clinical importance, the effects of poor drinking water quality on human health can take many different forms, ranging from asymptomatic infections to gastroenteritis and diarrhea to serious sickness and eventually death.39 In our study, surface and well water users significantly reported frequent loose stools and gastric discomfort respectively. One possible explanation for this may be As exposure through drinking water as evident from the health risk assessment (HQₐₛ = 5.5). As is known to cause frequent loose stools and abdominal pain.52 Other possible reasons could be fecal contamination via runoff from agricultural fields around the Sina River. Some research suggests that when pathogens are present in well water, the home plumbing environment may encourage additional microbial growth, resulting in increased pathogen concentration in the water.53 This could well be a possible explanation for gastric discomfort in well water users. Other possible cause may be presence of latrine in the house.

Table 5. As and Hg levels in urine samples of representative samples.

|                  | ARSENIC – URINE (µg/L) | MERCURY – URINE (µg/L) |
|------------------|------------------------|------------------------|
| N                | 47                     | 47                     |
| Mean             | 42.043                 | 4.915                  |
| Std. error of mean | 2.6356                | 0.2801                 |
| Median           | 39.000                 | 5.000                  |
| Std. Deviation   | 18.0687                | 1.9205                 |
| Range            | 72.0                   | 9.0                    |
| Minimum          | 10.0                   | 1.0                    |
| Maximum          | 82.0                   | 10.0                   |
| 25               | 27.000                 | 4.000                  |
| 50               | 39.000                 | 5.000                  |
| 75               | 57.000                 | 6.000                  |
In our study only 105 participants out of 557 (18.9%) have latrine in their house. Previous study has documented that presence of latrine in house decreases the incidence of diarrhea. This could be a confounding factor in our study. This study only included the human health risk assessment of elements (F, As, Cd, Hg, Pb, Ni, Cu, Zn, Mn, and Cr) through ingestion and dermal absorption of drinking water. There are other routes of heavy metal exposure such as inhalation, which may increase the overall heavy metal intake but were not considered in this study. Another limitation is that many investigated heavy metals were found below detectable limits. Hence the value of the detectable limit was considered for calculation.

Conclusion
In our study frequent loose stools, gastric discomfort, and frequent abdominal pain were associated with the various sources of drinking water that is surface water, hand pump, wells, and municipal water. As per the observation from different villages for urine As and Hg levels in selected individuals, it was found that Hg levels were found higher than the NHANES (USA) Study. Moreover, it is recommended that regular monitoring of drinking water should be enforced around the industrial hub as metal accumulation can be toxic to consumers when they are drinking water. As per the observation from different villages for urine As and Hg levels in selected individuals, it was found that Hg levels were found higher than the NHANES (USA) Study. Moreover, it is recommended that regular monitoring of drinking water should be enforced around the industrial hub as metal accumulation can be toxic to consumers when they are drinking water. As per the observation from different villages for urine As and Hg levels in selected individuals, it was found that Hg levels were found higher than the NHANES (USA) Study. Moreover, it is recommended that regular monitoring of drinking water should be enforced around the industrial hub as metal accumulation can be toxic to consumers when they are drinking water.

Acknowledgements
None

Author Contributions
Govind Mawari (G.M.), Naresh Kumar (N.K.), Sayan Sarkar (S.S.), Arthur L Frank (A.F.), Mradul Kumar Daga (M.K.D.), Mongjam Meghachandra Singh (M.M.S.), Tushar Kant Joshi (T.K.J.), Ishwar Singh (I.S.). M.K.D., M.M.S., T.K.J., and G.M.; Designed, Supervised, and planned the research. M.K.D., T.K.J., M.M.S., and N.K.; performed the Patient Examination and lab experiments. G.M., and S.S.; collected the data. M.K.D., G.M., M.M.S., S.S., and N.K.; took the lead in writing the manuscript. M.K.D., M.M.S., T.K.J., G.M., A.F., I.S., and N.K.; Final manuscript editing. All authors provided critical feedback and helped shape the research, analysis, and manuscript.

Ethics Approval
This study was conducted only after approval from the institutional ethical committee Maulana Azad Medical College.

Consent to Participate
Written consent was taken from all participating subjects.

Consent for Publication
All the authors give their consent for publication of the manuscript.

ORCID iDs
Sayan Sarkar https://orcid.org/0000-0003-3999-3110
Mradul Kumar Daga https://orcid.org/0000-0001-7774-7602

Supplemental Material
Supplemental material for this article is available online.

REFERENCES
1. Agrawal A, Pandey RS, Sharma B. Water pollution with special reference to pes- with cadmium exposures in older Americans with low zinc intake. J Trace Elem Med Biol. 2018;46:4863-4873.
2. United Nations. The 2030 agenda and the sustainable development goals: an opportunity for Latin America and the Caribbean. Published online 2018.
3. Staflow T, Saik J, Boev B, et al. Distribution of some elements in surface soil over the Kavadarci region, Republic of Macedonia. Environ Sci Technol. 2010;41:1515-1530.
4. Gu YG, Li QS, Fang JH, He BY, Fu HB, Tong ZJ. Identification of heavy metal sources in the reclaimed farmland soils of the pearl river estuary in China using a multivariate geostatistical approach. Environ Sci Technol. 2014;105:7-12.
5. Dong A, Zhai S, Zabel M, Yu Z, Zhang H, Liu F. Heavy metals in changjiang estuarine and offshore sediments: responding to human activities. Hai Yang Xue Bao. 2012;31:88-101.
6. Liao J, Xu X, Xie B, et al. Multi-phase distribution and comprehensive ecological risk assessment of heavy metal pollutants in a river affected by acid mine drainage. Environ Sci Technol. 2015;141:75-84.
7. Gao L, Chen J, Tang C, et al. Distribution, migration and potential risk of heavy metals in the Shima river catchment area, South China. Environ Sci Process Impacts. 2015;17:1769-1782.
8. Luo Y, Rao J, Jia Q. Heavy metal pollution and environmental risks in the water of Rongna river caused by natural AMD around tiegelongnan copper deposit, Northern Tibet, China. PLA Sci. 2012;17:e0266700.
9. Klaassen CD, Liu J, Diwan BA. Metallothionein protection of cadmium toxicity. Toxicol Appl Pharmaco. 2009;238:215-220.
10. Lin YS, Caffrey JL, Lin JW, et al. Increased risk of cancer mortality associated with cadmium exposures in older Americans with low zinc intake. J Trace Elem Med Biol. 2013;76:1-15.
11. Navas-Acien A, Guillard E, Silbergeld EK, Rothenberg SJ. Lead exposure and cardiovascular disease—a systematic review. Environ Health Perspect. 2007;115:472-482.
12. Nieboer E, Tuji IJ, Martin ID, Libena EN. Human biomonitoring issues related to lead exposure. Environ Sci Process Impacts. 2013;15:1824-1829.
13. Medina-Pizzali M, Robles P, Mendoza M, Torres C. Arsenic intake: impact in human nutrition and health. Rev Perú Med Exp Salud Publica. 2018;35:93-102.
14. Volko M, Rhodes CJ, Monoc J, Irakovic M, Mazur M. Free radicals, metals and antioxidants in oxidative stress-induced cancer. Chem Biol Interact. 2006;160:1-40.
15. Tchounwou PB, Yedjou CG, Patlolla AK, Sutton DJ. Heavy metal toxicity and the environment. Expert Rev. 2012;10:131-164.
16. Sani HA, Ahmad MB, Hussein MZ, Ibrahim NA, Musa A, Saleh TA. Nano-composite of ZnO with montmorillonite for removal of lead and copper ions from aqueous solutions. Proc Saf Environ Prot. 2017;109:97-105.
17. Zhang X, Yang Q. Association between serum copper levels and lung cancer risk: A meta-analysis. J Int Med Res. 2018;46:4863-4873.
18. Langård S, Andersen AA, Gylæth B. Incidence of cancer among ferrochromium workers. Br J Ind Med. 1980;37:114-120.
19. Sonal T, Kataria HC. Physico-chemical studies of water quality of Shahpura Lake, Bhupal (MP) with special reference to pollution effects on ground water of its fringe areas. Curv World Environ. 2012;7:139-144.
20. Shaheed A, Orgilj J, Montgomery MA, Jeoldad MA, Brown J. Why “improved” water sources are not always safe. Bull World Health Organ. 2014;92:283-289.
21. WHO. Guidelines for Drinking-Water Quality. 4th ed. World Health Organiza- tion; 1997.
22. Mazg BZ. Hazardous metals and minerals pollution in India: sources, toxicity and management. J Position Paper. Indian National Science Academy. Published online 2011.
23. Deepali KK, Gangwar K. Metals concentration in textile and tannery effluents, associated soils and ground water. NY Sci J. 2010;3:82-89.
24. Kailhren MA, Singh BR. Heavy metal contamination of soil and vegetation in the vicinity of industries in Bangladesh. Water Air Soil Pollut. 1999;115:347-361.
25. Bureau of Indian Standards. IS 3025-1 (1987): Methods of sampling and test (physical and chemical) for water and wastewater Part 1 - sampling. Published online 1987.
26. APHA. *Standard Methods for the Examination of Water and Wastewater*. American Public Health Association; 2017.

27. USEPA. Regional screening levels (RSLs)—generic tables. Published online 2017.

28. United States Environmental Protection Agency (USEPA). *Risk Assessment Guidance for Superfund Volume I: Human Health Evaluation Manual (Part E, Supplemental Guidance for Dermal Risk Assessment)*. Office of Emergency and Remedial Response, US Environmental Protection Agency; 1989.

29. Zabin SA, Foaad MA, Al-Ghamdi AY. Non-Carcinogenic risk assessment of heavy metals and fluoride in some water wells in the Al-Baha region, Saudi Arabia. *Hum Ecol Risk Assess*. 2008;14:306-313.

30. Antoine JMR, Fung LAH, Grant CN. Assessment of the potential health risks associated with the aluminium, arsenic, cadmium and lead content in selected fruits and vegetables grown in Jamaica. *Toxicol Rep*. 2017;4:181-187.

31. Wang X, Sato T, Xing B, Tao S. Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. *Sci Total Environ*. 2005;350:28-37.

32. USEPA. A review of the reference dose and reference concentration processes. In: *Risk Assessment Forum, U. The Environmental Protection Agency*. 2002.

33. USEPA. Toxicological review of zinc and compounds. In support of summary tables. *Toxicol Rep*. 2017;4:181-187.

34. Wang P. Potentially Toxic Metal and metalloid fractionation contamination in sediments of daya bay, South China Sea. *Koruja Ind*. 2015;64:255-262.

35. Ahmad W, Alharry RD, Zubair M, Ahmed M, Hameed A, Rafique S. Toxic and heavy metals contamination assessment in soil and water to evaluate human health risk. *Sci Rep*. 2021;11:7006.

36. Mehrabruh G, Zarehbruk S. Concentration and health implication of heavy metals in drinking water from urban areas of Tigray Region, northern Ethiopia. *Momona Ethiop J Sci*. 2011;3:105-121.

37. Zhang T, Ruan J, Zhang B, et al. Heavy metals in human urine, foods and drinking water from an e-waste dismantling area: identification of exposure sources and metal-induced health risk. *Ecotoxicol Environ Saf*. 2019;169:707-713.

38. Lakete S, Moonga G, Wahl AM, et al. Biomonitoring of arsenic, cadmium and lead in two artisanal and small-scale gold mining areas in Zimbabwe. *Environ Sci Pollut Res Int*. 2022;29:4762-4768.

39. Basu N, Abare M, Buchanan S, et al. A combined ecological and epidemiologic investigation of metal exposures amongst indigenous peoples near the Marlin Mine in western Guatemala. *Sci Total Environ*. 2010;409:70-77.

40. Watringway WA, Irvin-Barnwell E, Li Z, Ragin-Wilson A. Biomonitoring of toxic metals, organochlorine pesticides, and polybrominated biphenyl 153 in Michigan urban anglers. *Environ Res*. 2022;203:11851.

41. Basu N, Nam DH, Kwasnaza-Aniah E, Renne EF, Niragio JO. Multiple metals exposure in a small-scale artisanal gold mining community. *Environ Res*. 2011;111:463-467.

42. Mambrey V, Rakete S, Tobollik M, et al. Artisanal and small-scale gold mining: a cross-sectional assessment of occupational mercury exposure and exposure risk factors in Kadoma and Shurugwi, Zimbabwe. *Environ Res*. 2020;184:109379.

43. Calao-Ramos C, Bravo AG, Paternina-Uribe R, Marrugo-Negrete J, Diez S. Occupational human exposure to mercury in artisanal small-scale gold mining communities of Colombia. *Environ Int*. 2021;146:106216.

44. Panday VK, Parameswaran M, Soman SD. The distribution of mercury in drinking water from urban areas of Tigray Region, northern Ethiopia. *Sci Total Environ*. 1986;48:223-230.

45. Centers for Disease Control and Prevention (CDC). Fourth national report on human exposure to environmental chemicals: updated tables, January 2019, Volume One. Published online 2017.

46. Mergler D, Anderson HA, Chan LHM, et al. Methylmercury exposure and health effects in humans: a worldwide concern. *AMBIO*. 2007;36:3-11.

47. Kowk RK. A review and rationale for studying the cardiovascular effects of drinking water arsenic in women of reproductive age. *Toxicol Appl Pharmacol*. 2007;222:344-350.

48. Villasenusa I, Bollinger JC. Arsenic in drinking water: sources, occurrence and health effects (a review). *Rev Environ Sci Biotechnol*. 2008;7:307-323.

49. Mapili K, Rhoads WJ, Coughter M, Pieper KJ, Edwards MA, Pruden A. Health risk factors in Kadoma and Shurugwi, Zimbabwe. *Environ Res*. 2020;211:109151.

50. Daniels DL, Cousens SN, Makoae LN, Feachem RG. A case-control study of occupational human exposure to mercury in artisanal small-scale gold mining communities of Colombia. *Environ Int*. 2021;146:106216.