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Geochemical, Topographical, and Meteorological Controls on Groundwater Arsenic Contamination in Sharda River Basin of Uttar Pradesh, India

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Abstract: Groundwater arsenic (As) contamination is a health threat for millions of people in the Gangetic plains of India. It is, therefore, critical to understand the mechanism of As enrichment to reduce the As exposure. Geochemical analysis of 30 groundwater samples collected across the banks of River Sharda was performed for the identification of major geochemical processes controlling groundwater geochemistry. Shallow wells (3-10 m) are found to be contaminated with As and is confined to newer alluvium of Holocene age. The average temporal decline of the groundwater level was observed across 10 monitoring stations is 0.067 m/yr. Decreasing rainfall, lesser recharge and huge groundwater extraction for irrigation might have impacted the groundwater to flow faster, while the increase in temperature and weathering regime favoured arsenic mobilisation. The long-term trends of rainfall show a decline of 1.97 mm/yr and the temperature increase is observed to be 0.0049°C/yr. These changes in rainfall and temperature also might have impacted the As mobilisation in groundwater. NO_3^- was found to be low in samples with low As concentrations, indicating the prevalence of reducing conditions. Whereas high concentrations of Fe were observed for high As samples, indicating their common source of origin. Also, the alkaline nature of aquifer and high concentrations of HCO_3^- might have contributed to As enrichment. Results from scatter plots and correlation matrix also support this sequential reduction leading to the reductive dissolution of iron oxyhydroxides and thus enriching the concentration of As in the groundwater.

Keywords: Arsenic; Groundwater; Geochemistry; Redox condition; Sharda River.

Introduction

Arsenic (As) is a toxic element. It is also carcinogenic and is widely accepted as a threat to public health (IARC, 2004; WHO, 2011; WHO/UNICEF, 2015). More than 296 million people worldwide are exposed to high As concentrations in groundwater (Chakraborti et al., 2018). Approximately 137 million people are exposed to unsafe levels of As in drinking water in South and Southeast Asia, which includes Bangladesh, Nepal, India, Pakistan, Cambodia, Laos, Myanmar, Thailand and Vietnam (Nickson et al., 1998; McArthur et al., 2001; Smedley, 2003; Stüben et al., 2003; Norra et al., 2005; Farooq et al., 2010; 2011; Chakraborti et

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al., 2016; Postma et al., 2016; Bhowmick et al., 2018). High As levels in groundwater have been reported in many parts of India (Bhowmick et al., 2018; Mazumder et al. 1988; Mukherjee et al., 2011). Drinking water with As>10 µg/L leads to serious health impacts causing skin lesions, keratosis and skin cancer (Mazumder et al., 2008; Shannon and Strayer, 1989; Rahman et al. 2009; Chen et al., 2017). Further, chronic As exposure significantly increases mortality due to cardiovascular diseases, skin, liver, bladder and lung cancer (Argos et al., 2010; Chen et al., 2011). Arsenicosis has been linked to mortality, impaired intellect and motor dysfunction in children (Rahman et al., 2009; Wasserman et al., 2014).

In the late 90's, clean groundwater (free of microbial contamination)led to an insurmountable increase in the number of handpumps in parts of India and Bangladesh. Since then, it has led to over-extraction of groundwater for irrigation purposes. Over-extraction of groundwater has significantly impacted the hydrologic regime of groundwater and, therefore, inhibiting natural and geochemical processes in the subsurface resulting in As mobilisation in groundwater. Over the past three decades, numerous studies have reported As contamination for groundwater in the states of West Bengal (Mazumder et al., 1988; Stüben et al., 2003; McArthur et al., 2004; Norra et al., 2005), Bihar (Chakraborti et al., 2016a; 2016b; Rahman et al., 2014; Saha, 2009), Assam (Nickson et al., 2007; Thambidurai et al., 2013; Jain et al., 2018), Manipur (Chakraborti et al., 2008), Arunachal Pradesh (Shah, 2015a), Tripura (Singh et al., 2008), Punjab (van Geen et al., 2019; Kumar et al., 2020), Uttar Pradesh (Saha, 2009; Singh et al., 2018; Bindal and Singh, 2019), Chhattisgarh (Shukla et al., 2010) and Jharkhand (Bhattacharjee et al., 2005). The incidence of high As level in groundwater is generally related to the geochemical environments of the aquifer (Mukherjee et al., 2011). The investigations suggest that the source of As lies in alluvial and deltaic sediments belonging to the quaternary Holocene sediments, which originated from the Himalayas and were drained by the river and deposited in their respective floodplains (Shah, 2008; Smedley and Kinniburgh, 2000).

Numerous studies have been conducted in middle Gangetic plains to understand the source and distribution of As and the processes controlling its mobility in groundwater (Chakraborti et al., 2016; Kumar et al., 2010; Ramanathan et al., 2015; Shah, 2010, 2015). McArthur et al. (2004) and Naseem and McArthur (2018) have proposed several mechanisms for As release in groundwater. One of the proposed mechanisms is that As is released under oxidising conditions, with the dissolution of sulphide-rich minerals such as arsenopyrite (Smedley and Kinniburgh, 2000). It may also be released by desorption of As (V) from As-bearing iron (Fe) oxides, hydroxides and oxyhydroxides at pH>8 (Stüben et al., 2003; Farooqi et al., 2007). However, the widely accepted mechanism is microbially mediated reductive dissolution of As-bearing Fe (III) oxides (Nickson et al., 2000). The reductive dissolution of Fe oxyhydroxides has been widely observed in the Gangetic plains and Bengal basin and is primarily responsible for As mobilisation under different hydrogeological settings (Biswas et al., 2014; McArthur et al., 2008; Nickson et al., 2000).

Uttar Pradesh and Bihar located in the middle Gangetic plains are affected by high As in groundwater (Saha, 2009). The major districts of Uttar Pradesh which are affected by As include Ballia (Chandrasekharam et al., 2007; Chauhan et al., 2009), Gorakhpur (Singh et al., 2018), Bahraich (Mehrotra et al., 2016), Ghazipur (Saxena et al., 2014), Gonda (CGWB, 2014), Kanpur (Chauhan et al., 2012) and Varanasi (Mukherjee et al., 2018). The As contamination has been reported from Buxar (Shah, 2014; Kumar et al., 2015), Bhojpur (Saha, 2009), Vaishali (Saha, 2009), Patna (Saha, 2009) districts of Bihar. Altogether in Uttar Pradesh, 22 million people might be effected to due high As in groundwater (Bindal and Singh, 2019). These districts are situated alongside rivers Ganga, Rapti and Ghaghra, which derive thick quaternary unconsolidated sediments from the Himalayas.

With the above background and data, studies have been conducted to establish the As contamination in the Sharda river basin, and also to identify the factors and hydrogeochemical processes controlling As-enrichment in the groundwater.

Materials and Methods

Study Area

Sharda river basin is situated in the Lakhimpur Kheri district of Uttar Pradesh. The district has a population of 4.02 million, and its population density of 524 people/km^2 is a part of the middle Gangetic plain (Census, 2011). Sharda river separates Nepal's border with India lying in the Terai region along the Himalayan foothills between 27.60°N and 28.60°N in latitude and 80.30°E and 81.30°E in longitudes. River Sharda flows from north to southeast and joins the Ghaghra River in the district Bahraich.

Geology and Geomorphology

The general slope of the area is from north-west to southeast. The area is marked by numerous streams and channels flowing in the southeast direction. The surface is interrupted by low riverbeds and high banks which spreads the stream on either side of the banks. Both the rivers, Sharda and Ghaghra, quite often change their course causing meandering which results in the formation of ox-bow lakes. The dominant soil type is black clay, which can absorb water and remain moist for longer durations. The texture of the soils varies from clayey loam to loam. The climate is sub-tropical, and the area receives moderate to heavy rainfall during the monsoon season. During monsoon, the Terai area is prone to flooding and consequent waterlogging (Shah, 2015b).

The region comprises of the quaternary alluvial deposits of the Ganga Plains and is divided into (a) older alluvium (Bhangar) of mid-Pleistocene, and newer alluvium (Khadar) belonging to the Holocene Period (Pant and Sharma, 1993). The older alluvium of Varanasi comprises of a polycyclic sequence of sand, silt and clay occasionally with calcrete. The newer alluvium, disconformably overlying the older alluvium Varanasi, belongs to the Holocene age. It represents the second phase of deposition in the Gangetic Plains and is sub-divided into an alluvial fan, channel alluvium and terrace alluvium, confined within the palaeo-banks of the rivers. Alluvial fans are the recent accumulations of material at the base of foothills, due to an abrupt drop in the channel gradient (Pant and Sharma, 1993). The terrace alluvium occurs within wide floodplains of the river characterised by an extensive development of relict features of abandoned channels, meander cut-offs, linear water bodies, which are related to the former active channels. In the active flood channels, the channel alluvium is seen in the form of point bars, channels bars and lateral sand bars. This is restricted to the active floodplain of the river Sharda.

In the study area, dug wells of approximately 3-15 m depth and shallow borewells and handpumps of 6-15 m depth exploit groundwater of unconfined aquifers which are made up of fine to coarse sand. The region comprises semi-confined and unconfined layers of aquifers, which is the source of drinking and irrigation water in the region (CGWB, 2014). The second layer of aquifers is confined between 18 m and 40 m, approximately. For the past two decades, there has been a substantial increase in the number of shallow handpumps and borewells leading to overexploitation of groundwater which might have impacted the hydrological regime in the region (Pahuja et al., 2010; NRDWP, 2012).

Sampling and Analytical Methods

A total of 78 handpumps were tested randomly for As using the ITS Econo-Quick test kit (George et al. 2012a) (Figure 1) and 30 groundwater samples were randomly collected from the villages lying on the floodplains of Sharda river. The villages were chosen based on their proximity to the river and geomorphic features. These ITS As Econo-Quick kit (part no. 481298) relies on the Gutzeit method which converts the As contained in the 50 ml sample into arsine gas, which gets trapped on a strip impregnated

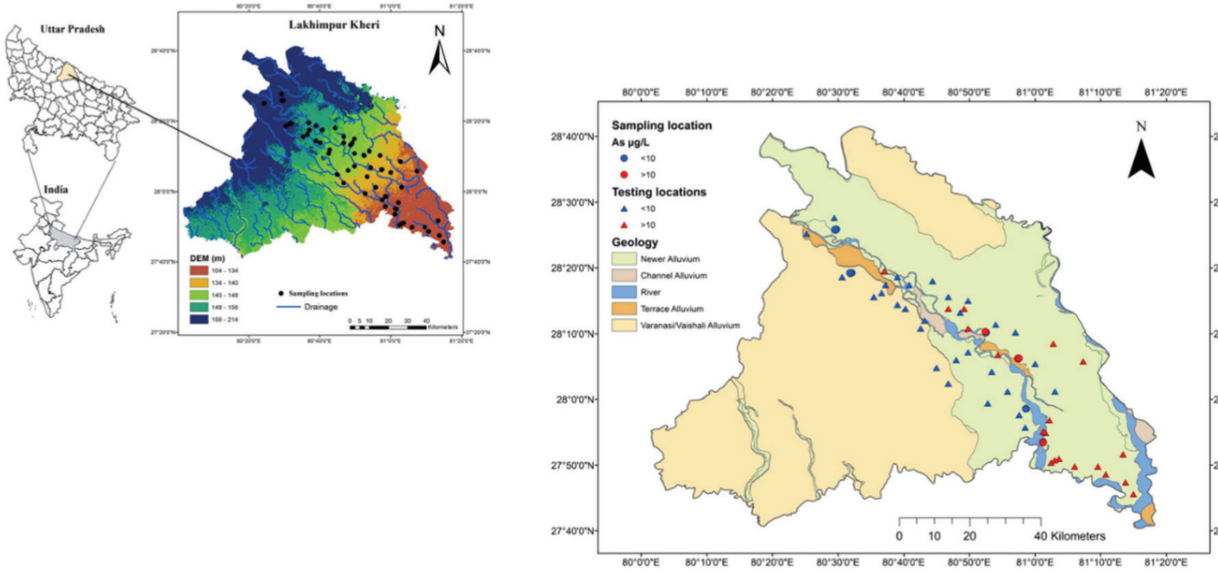

Figure 1: Topography and sampling locations within the study area.

with mercuric bromide. Additionally, tartaric acid, an oxidant to remove any interfering hydrogen sulphide, and a fine Zn powder as the reductant are added with the reaction time of 10 mins. The groundwater in the region was odourless (indicating absence of sulphide) and therefore the addition of tartaric acid was avoided in the testing of groundwater samples in the study region. The comparisons with laboratory measurements (George et al., 2012a; van Geen et al., 2014a) have shown that the kit correctly categorised ~90% of wells relative to the WHO guideline of 10 µg/L for As.

Handpumps were purged for 10 mins before sample collection, to avoid the impact of iron pipes. Samples tested were collected in two separate 200 ml polypropylene bottles for further laboratory analysis. For the determination of the cations, the sampling bottles were acidified using 1% HNO_3 (~pH 2). The samples for anion analysis were collected in separate 200 ml bottles and were left unacidified. The ancillary information such as type, depth and age of handpump were collected using a geo-tagged survey questionnaire. This geo-tagged survey was carried on an android-based platform that utilises Open Data Kit (ODK) and the data is collected directly on mobile phones. Electrical conductivity (EC) and pH were recorded onsite with the help of a portable Oakton probe. On each day of sampling, the Oakton probe for EC was calibrated with a standard solution and the measured values were within 8% and 16% of the expected values of 1990 µS/cm and 16.0 µS/cm, respectively. The Oakton probe calibration was done with pH standards of 7.0 and 10.0 and the readings were all within 0.3 of a pH unit. In the laboratory, acidified groundwater was analysed by atomic absorption spectrophotometer (Thermo-Fischer) for the measurement of As, Fe, Ca^{2+}, Mg^{2+}, Na^+ and K^+ (APHA, 2008). The instrument's detection limit was <0.1 µg/L, and the analysis was carried out for duplicates with an error range of <2%. Non-acidified samples were analysed for anions such as bicarbonates (HCO_3^-), nitrate (NO_3^-), sulphate (SO_4^{2-}), chloride (Cl^-) and dissolved phosphate (PO_4^{3-}) using ion chromatograph (Dionex) with a detection limit of <0.05 µg/L. The samples for anion analysis were treated with 1 N HNO_3 which prevents interference from hydroxide and carbonates (APHA, 2008). These anions/cations were selected for geochemical modelling as they are significant for understanding the redox-sensitive conditions that is essential for As mobilisation in aquifers of the region. All the maps were prepared using ArcGIS 10.1.

Results

Hydrochemistry of Aquifer

The groundwater samples collected were evaluated for all the necessary drinking water quality parameters. The observed values for all the parameters were compared with the values given by the Bureau of Indian Standards (BIS, 2012) (Tables 1). The pH of the groundwater samples varied between 6 and 8.3 with an average of 7.2. Results indicate that some of the groundwater samples were slightly alkaline. However, no such correlation exists between the pH and As concentration in groundwater samples. The value of EC ranged from 325 to 1124 µS/cm with a mean of 659.67 µS/cm. A higher value of EC suggests the prevalence of cations and anions, indicating high ionic strength and influence of surface recharge on groundwater. Spatial variation in water quality parameters suggests that the hydro-geochemistry of the area is highly heterogeneous. HCO_3^- was found as the most abundant anion in groundwater and ranged from 122 to 896 mg/L. The source for the high concentrations of HCO_3^- can be because of the presence of calcite in the aquifers. Moreover, the decomposition of peat deposits also leads to an increase in HCO_3^-. The flushing of CO_2-rich water from the unsaturated zone, where CO_2 is formed as a byproduct from organic matter decomposition, this might also lead to the formation of HCO_3^- (Buschmann et al., 2007). The concentration of NO_3^- ranged up to a maximum of 799 mg/L with a mean of 83.69 mg/L. This high concentration can result from anthropogenic activities such as the addition of fertilisers in the irrigation fields (Kumar et al., 2015). Additionally, due to lack of sanitation infrastructures, a lot of human sewerage seeps and contaminates the groundwater in the region. SO_4^{2-} concentrations varied between bdl and 116 mg/L. The presence of SO_4^{2-} might be due to decomposition of organic matter and excess run-off from surface. Another possible source of SO_4^{2-} could be pyrite mineral weathering along with anthropogenic inputs due to excessive usage of fertilisers. The concentration of PO_4^{3-} varies within bdl-0.5 mg/L, which might be due to the application of PO_4^{3-} rich fertilisers in the agricultural fields to enhance the productivity of wheat and rice crops (Mandal et al., 2019). Cl^- concentration ranges from 1.14 to 122.78 mg/L with a mean value of 22.61 mg/L. This high concentration of Cl^- in the groundwater samples may be associated with the percolation of sewerage which gets mixed with groundwater (Samantara et al., 2015). The details of the geochemistry data for 30 samples are

Table 1: Groundwater quality parameters for Lakhimpur Kheri along with values for Bureau of Indian Standard (BIS, 2012)

Groundwater quality parameters	*Acceptable limit (BIS, 2012)*	*Permissible limit (BIS, 2012)*	*Min*	*Max*	*Mean*
pH	6.5–8.5	–	6.0	8.3	7.2
Conductivity (EC)	200	600	325	1124	659.67
Manganese (Mn^+)	0.1	0.3	0.00	1.2	0.1
Iron (Fe^{3+})	0.3	No relaxation	0	11.28	0.74
Sodium (Na^+)	–	–	4.8	79.6	31.304
Potassium (K^+)	–	–	0.5	58	9.8
Calcium (Ca^{2+})	75	200	2.2	87.6	37.49
Magnesium (Mg^{2+})	30	100	1.4	61.6	29.86
Chloride (Cl^-)	250	1000	1.14	122.8	22.6
Sulphate (SO_4^{2-})	200	400	0	116	27.7
Nitrate (NO_3-)	45	No relaxation	0	799.3	83.69
Bicarbonate (HCO_{3-})	–	–	122	896	346
Phosphate (PO_4^{3-})	–	–	0.0	.5	0.35
Arsenic (As)	0.01	0.05	0	58.32	13.5

*All values are in mg/L, except for As in µg/L, pH, and EC (µS/cm).

given in Table 2. For cations, the order of dominance was found to be in the order Ca^{2+}>Na^+>Mg^{2+}>K^+. The Ca^{2+} and Mg^{2+} ranges from 2.2 to 87.6 mg/L with mean of 37.49 mg/L and from 1.4 to 61.6 mg/L with mean 29.86 mg/L, respectively. This indicates that weathering of calcium- and magnesium-rich minerals such as calcium carbonates, feldspar minerals, and dolomite is the prevalent process in the region. Earlier studies have also confirmed the presence of these minerals in the active floodplains of the River Ganges (Bhattacharya et al., 1997). Na^+ ranges from 4.8 to 79.6 mg/L with mean of 31.3 mg/L. K^+ ranges from 0.5 to 58 mg/L with mean, 9.8 mg/L. Additionally, the weathering of K^+ bearing minerals i.e., K-feldspar and anthropogenic activities such as the use of fertilisers might also contribute K^+ in groundwater. Fe ranges within bdl-11.28 mg/L with a mean value of 0.74 mg/L.

Hydrochemical Facies of Groundwater

The piper plot has been used to summarise the hydro-geochemical characteristics of groundwater (Piper, 1944). Na^+ and Mg^{2+} were found to be the dominant cation, while SO_4^{2-}, Cl^- and HCO_3^- were the dominant anions in the north and northwestern part of the study area (Figure 2a). The samples were classified as Ca-SO_4, Na-Cl and Na-HCO_3 type, which represents surface seepage and freshwater recharge in the aquifer (Singh et al., 2011, 2018). Further to the south, the groundwater type changes to Mg-HCO_3 type. The water type Mg-HCO_3 extends in the Sharda alluvial fan to south of the Ghaghara River, indicating temporary hardness in the groundwater. In the centre of the study region, the dominant groundwater type is Ca-NO_3, Na-Cl, and Mg-HCO_3 indicating the mixed type water facies influenced by anthropogenic activities. This might also be due to the mixing of surface recharge and surface sewage within the aquifer. The piper diagram shows that most of the groundwater samples with high As had Mg-HCO_3 water type (Figure 2a).

Ca^{2+} and Mg^{2+} were found to be dominant among cations, and HCO_3^- was the dominant anion indicating reducing conditions in the region. Few studies suggest that alkalinity in groundwater is influenced by rainfall and soil interaction. The carbonic acid (H_2CO_3) is produced by the interaction of dissolved CO_2 and rainfall (H_2O), resulting in the formation of HCO_3^- and H^+ ions. The HCO_3^- penetrates down in the soil and initiates the weathering of minerals present in the aquifer (Salama et al., 1999). This is supported by the presence of abundant Ca^{2+} and Mg^{2+} in the groundwater samples indicating weathering of carbonate and silicate rocks in the basin (Kumar and Singh, 2015). The dominant dissolution of both calcite and dolomite is shown in Figure 2b, which indicates the

Table. 2: The physico-chemical parameters of analysed groundwater samples

As	EC	Mn	Fe^{3+}	Na^{+}	K^{+}	Ca^{2+}	Mg^{2+}	Al^{3+}	Cl^{-}	SO_4^{2-}	NO_3^{-}	HCO_3^{-}	PO_4^{3-}
6.99	1090.00	0.02	0.42	30.50	6.90	72.40	35.80	0.00	26.87	28.67	476.51	178.00	0.00
0.00	325.00	0.01	1.64	57.50	0.50	2.20	28.30	0.00	51.87	47.90	0.01	349.60	0.00
0.68	1124.00	0.02	0.34	79.60	16.40	76.80	23.80	0.00	122.77	99.44	0.02	122.20	0.00
0.80	592.00	0.00	0.72	17.50	2.10	42.50	61.60	0.00	5.51	20.06	799.29	189.00	0.00
1.70	680.00	0.02	1.89	56.70	7.80	39.60	19.20	0.00	58.23	35.05	0.01	558.60	0.00
1.77	792.00	0.02	1.13	58.50	9.40	42.60	27.10	0.00	58.90	35.39	0.15	291.70	0.00
1.97	675.00	0.11	2.51	25.30	2.70	43.80	40.60	0.00	5.26	37.40	263.68	250.00	0.00
2.23	513.00	0.01	0.00	15.60	4.10	10.00	1.40	0.00	2.01	13.14	0.01	178.00	0.00
2.26	607.00	0.01	0.26	51.80	12.10	51.70	12.80	0.00	35.55	25.73	0.06	346.00	0.00
2.39	520.00	0.00	0.66	23.30	10.10	64.40	48.80	0.00	32.02	17.24	0.09	283.00	0.00
2.67	898.00	0.02	0.20	52.10	6.90	49.30	22.30	0.00	56.56	53.69	338.37	159.00	0.00
3.60	665.00	0.00	0.00	38.20	13.10	30.70	23.50	0.00	23.34	19.07	0.02	467.00	0.00
3.71	707.00	0.01	0.04	36.10	14.10	45.10	31.90	0.00	23.73	19.95	0.01	284.30	0.00
4.46	523.00	0.03	0.68	8.90	8.60	62.00	15.00	0.00	35.50	115.93	0.00	168.00	0.00
4.90	730.00	0.00	0.02	44.70	12.30	9.80	13.60	0.00	6.68	5.60	1.22	270.00	0.00
6.00	707.00	0.01	1.23	4.80	5.10	37.70	36.40	0.00	3.56	22.64	0.49	198.00	0.00
6.97	574.00	0.01	0.86	43.50	4.60	37.90	33.80	0.00	1.96	5.11	0.02	278.00	0.00
8.00	500.00	0.00	0.22	31.30	11.30	35.70	20.40	0.00	4.96	9.05	0.01	262.20	0.10
18.00	480.00	0.08	4.52	68.50	7.40	32.10	31.10	0.00	5.97	31.87	0.01	187.00	0.00
19.20	458.00	0.00	4.78	12.00	10.30	16.40	38.20	0.00	2.77	21.78	0.01	740.20	0.00
19.20	717.00	0.00	12.85	10.90	8.20	56.40	34.60	0.00	3.60	26.06	309.86	182.00	0.00
19.35	1028.00	1.16	14.30	62.00	58.00	87.60	47.80	0.00	55.15	72.65	315.33	669.00	0.50
20.13	665.00	0.01	3.79	10.40	7.80	34.10	25.20	0.00	3.95	23.71	0.02	164.70	0.00
24.00	635.00	0.00	12.01	19.70	12.40	10.00	38.60	0.00	8.33	7.57	0.01	550.00	0.00
24.08	520.00	0.00	3.07	8.20	2.50	20.10	20.20	0.00	3.15	2.23	2.76	509.00	0.00
38.90	644.00	0.01	4.53	24.50	12.00	19.80	35.30	0.00	17.76	14.83	0.01	583.00	0.00
40.19	630.00	0.04	8.46	10.00	8.10	13.50	17.30	0.00	7.05	0.01	2.27	164.70	0.00
40.72	685.00	0.18	0.19	21.00	5.20	23.20	39.70	2.90	1.14	2.23	0.01	384.30	0.10
47.00	537.00	0.01	10.20	7.40	7.20	11.90	30.80	0.00	2.60	0.01	0.12	517.20	0.00
58.32	569.00	0.00	25.45	8.70	7.50	45.50	40.80	0.00	11.55	16.36	0.23	896.30	0.00

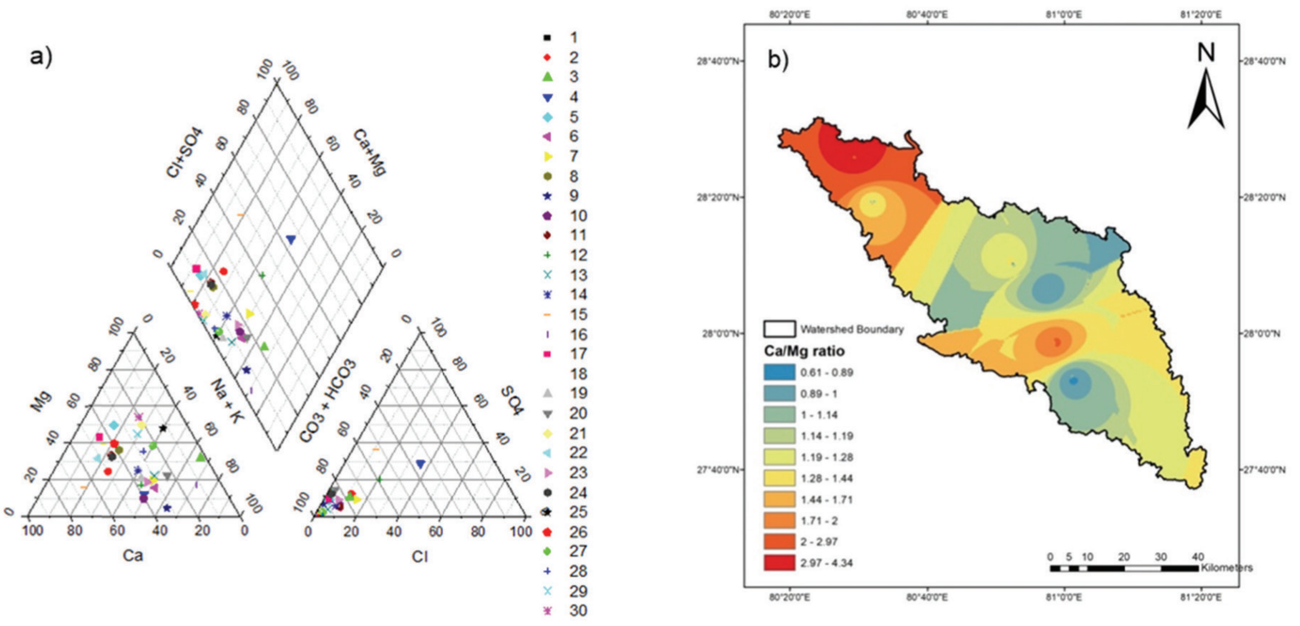

Figure 2: (a) Piper diagram of groundwater samples showing water facies, (b) spatial pattern of the ratio of Ca^{2+} and Mg^{2+} across the study area.

groundwater Ca^{2+}/Mg^{2+} molar ratio. The Ca^{2+}/Mg^{2+} molar ratio is equal to one indicating the dissolution of dolomite, while a greater value represents a calcite dissolution. Ca^{2+}/Mg^{2+} ratio, greater than 2, represents the dissolution of silicate minerals into the groundwater (Huq et al., 2018). While, 43.3% of the groundwater samples had a Ca^{2+}/Mg^{2+} ratio between 1 and 2, which indicates that dissolution of calcite. About 23.3% of the samples only had a higher ratio than 2, which showed the minimum effect of silicate minerals that contribute Ca^{2+} and Mg^{2+} to the groundwater. Around 33.3% of samples were indicative of the dissolution of dolomite with Ca^{2+}/Mg^{2+} ratio <1 (Figure 3).

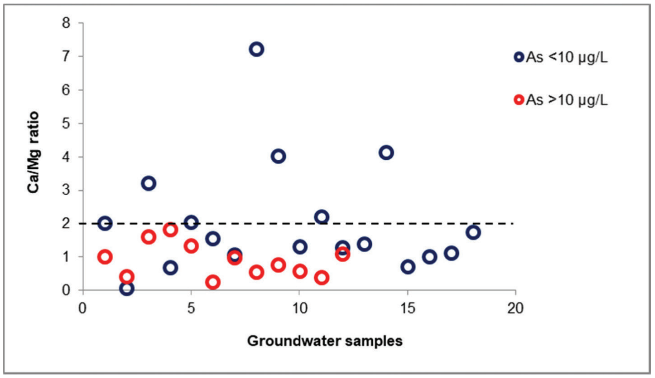

Figure 3: Scatter plot depicting the Ca^{2+}/Mg^{2+} molar ratio.

The spatial pattern of the ratio of Ca^{2+} and Mg^{2+} showed the variation from the northwest to the southeast and central parts of the plains. The ratio was the highest towards the north due to the increase in Ca^{2+} concentration through the weathering of silicate in the recharge area. Chemical composition of groundwater may also be altered by the mixing of freshwater recharge from rainwater, residence time and soil-water interaction (Sø et al., 2018; Saha et al., 2010). To further understand the contribution of the processes such as evaporite dissolution, silicate weathering and carbonate dissolution, a bivariate mixing diagram of Na-normalised Ca^{2+} versus Na^{+} normalised Mg^{2+} and HCO_3^- was plotted (Figure 4a). Most of the groundwater samples from older alluvium show dominance of silicate weathering (Figure 4a and 4b). Samples from younger alluvium tend to be more towards the carbonate dissolution than to silicate and dolomite weathering. The ratio decreased with the distance from the weathering zones towards the recharge zone in the northern part.

Topographic, Geological and Climatic Controls on As Contamination

Out of the total 30 samples, 40% (n = 12) were found to have As above permissible limit of 10 µg/L as recommended by WHO and the BIS (BIS, 2012; WHO, 2011). The As concentration is less than 10 µg/L in the northern part and more than 10 µg/L in the south and southeast part of the study area. It could be possible due to an accumulation of finer sediments deposited along the course of river Sharda or due to some local effects. Figure 1 also shows that all samples belonging to the older alluvium have As<10 µg/L and samples in the newer alluvium, were of mixed types. In our study region, the As contamination is spread along the right side of the river Sharda and between the doab regions of Sharda and Ghaghara river. This suggests the important role of depositional environment and geological age in controlling As mobilisation in the basin. Further, samples with high As concentrations are mostly present in the relatively flat land formed by the doab of Sharda and Ghaghara rivers (Figure 1). As the slope gradient reduces, it minimises the flow of sediments brought by the river to flow downstream and increases their deposition in flat regions/plains. These regions have an elevation range from 130 to 145 m. The elevation of the area plays an important role in the As occurrence in the groundwater (Singh et al., 2018). Samples with high As concentrations are found in areas with low topography/depressions (Figure 5). This result is consistent with the research performed by Buschmann et al. (2007), Shamsudduha, (2008) and Khan et al. (2016) from Bangladesh as well as from Bihar, where the handpumps with high As are located within topographic depressions (Saha and Sahu, 2016).

A study by Kumar et al. (2018) using groundwater level data from the three monitoring stations in the district showed a significant declining trend of groundwater level for both the pre-monsoon and post-monsoon season. The Mann-Kendall statistic for the groundwater level for pre-monsoon for the period of 1990-2016 over three monitoring stations shows the significant decreasing value of –3.107, –1.711 and –2.251, respectively. The analysis of groundwater level data for the last 25 years reveals an average declining trend at the rate of 0.067 m/yr (Figure 6). The data from 10 monitoring stations were used to study the trend of groundwater level fluctuation out of which seven monitoring stations showed a declining trend varying between 0.38 m/yr and 0.009 m/yr with an average of 0.12 m/yr (Table 3). The rainfall in the district is high, however, 70% of the total rainfall results in runoff; thus, little recharge of groundwater takes place. The long-term trends observed for rainfall show a declining trend of 1.97 mm/yr and the temperature shows an increasing trend of 0.0049°C/yr (Figure 7). Thus, the decrease groundwater recharge accompanied by higher runoff might also impact the mobilisation of As in groundwater.

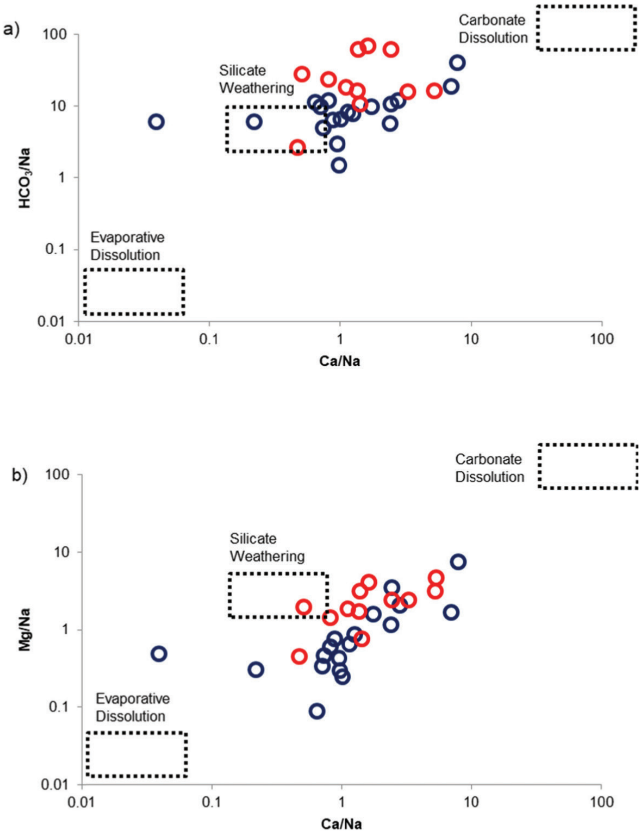

Figure 4: (a) Bivariate Na^+ normalized plot between HCO_3^- and Ca^{2+}, (b) bivariate Na^+ normalised plot between Mg^{2+} and Ca^{2+}.

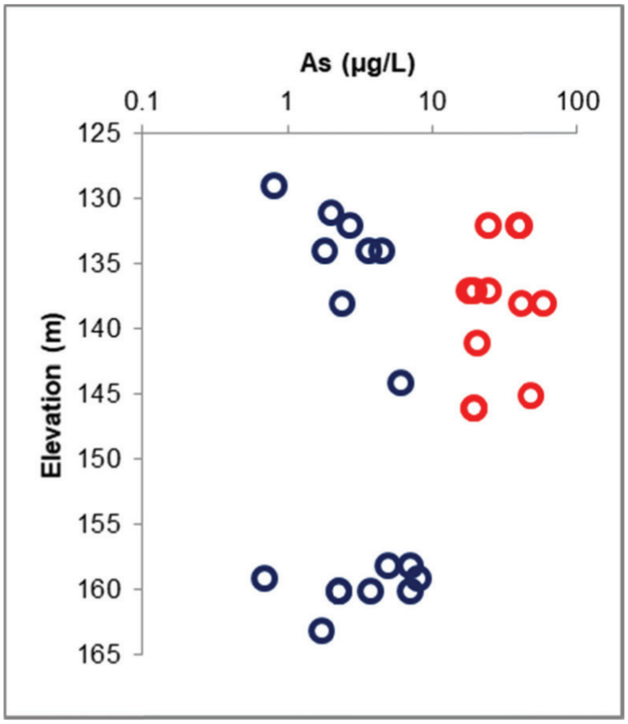

Figure 5: Distribution of As with respect to the topography of the study area.

Table 3: The trend observed for the 10 groundwater level monitoring stations

S. No.	*Monitoring stations*	*Trend line equation*
1	Mailani	y = -0.0322x + 5.861
2	Behjam	y = -0.3173x + 8.5192
3	Dharmapur	y = 0.1019x + 5.1207
4	Gularia	y = -0.0163x + 5.0072
5	Asogapur	y = 0.0048x + 3.9886
6	Chamlapur	y = -0.0425x + 2.9718
7	Chauapur	y = -0.0009x + 2.4387
8	ChandanChowki	y = 0.0593x + 3.6257
9	Dudhwa	y = -0.0438x + 10.321
10	Maigalganj	y = -0.3897x + 11.055

The groundwater level fluctuation as a result of climate variability may increase the groundwater temperature and thus enhance sulphide weathering majorly influenced by groundwater level decline. The increased oxidation of sulphide minerals owing to a decline in the water table may enhance the concentration of dissolved minerals which is influenced by the toxic environment in the aquifer (Appelo and Postma, 2005).

Falling water tables enhance the exposure of rocks to oxygen, which diffuses through the unsaturated zone; the relatively low diffusivity of oxygen in water limits sulphide oxidation in the saturated zone (Pili et al., 2013; Todd et al., 2012). Therefore, the newly exposed rocks might contain fresher sulphides and thus more reactive mineral surfaces such as those without

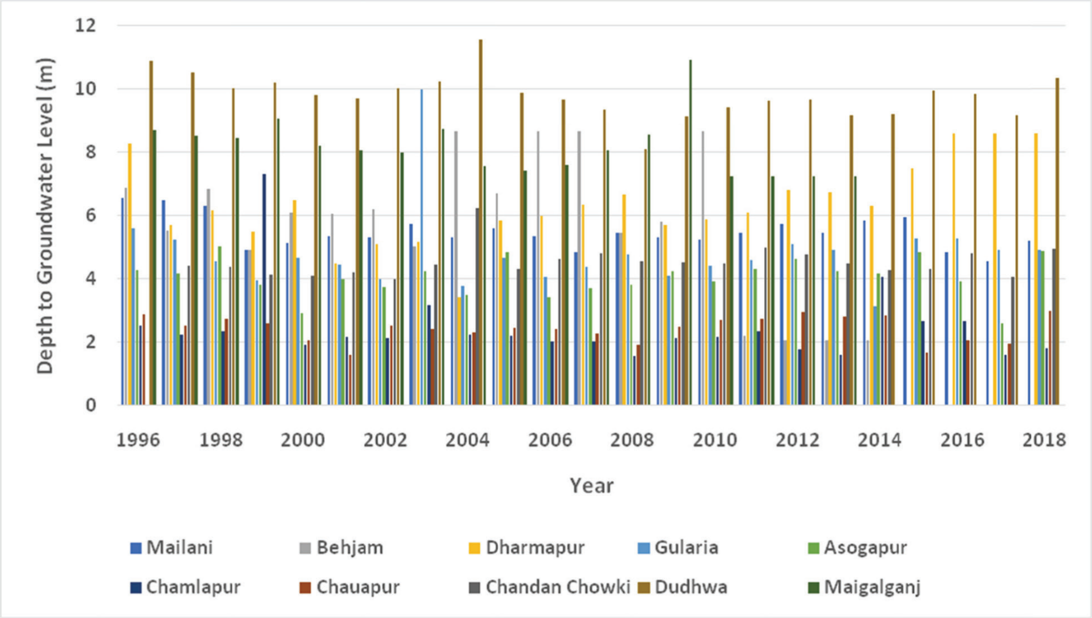

Figure 6: Groundwater level fluctuation with a declining trend in 7 out of 10 monitoring stations in the region.

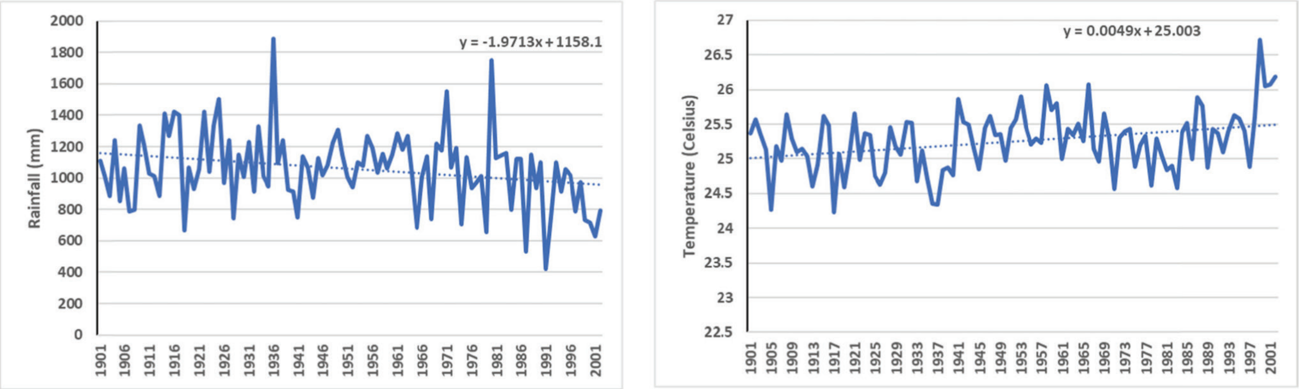

Figure 7: The declining trend of rainfall and the increasing trend of temperature in the region for ten stations.

coatings of secondary minerals. Reduced groundwater flow results in more geochemically evolved groundwater that would be prominent in contributing to flow increase during prolonged low-flow periods due to less recharge. This would also result in increased residence time and therefore extensive water-rock interaction leading to a more favourable environment for As mobilisation by reductive dissolution and alkali desorption. Even a slight change in groundwater temperature might result in decreased dissolved oxygen; thus, redox potential impacting the As mobilisation by reductive dissolution. In particular, a very high As concentrations may occur in downgradient areas that already contain geochemically evolved groundwater as observed in the groundwater samples of the study area. The experimental study also demonstrates that As gets precipitated during the SO_4^{2-} and Fe reduction which is associated with changes in the groundwater level (Kirk et al., 2010).

Depth Profiles

Most of the samples were from the shallow handpumps with an average depth of 0-20 m, but few households with depth up to 35-38 m were also sampled (Figure 8a-h). The presence of Ca^{2+}, Mg^{2+}, Fe, Cl^-, HCO_3^- in high concentrations indicates soil mineralisation at shallow depths. The shallow aquifers have no defined pattern for EC as well as other ions for samples with low As (Figure 8a). However, for samples with high As, there is a pattern found along with depth. Most of the samples with elevated As concentrations were confined to depths of 6-15 m (Figure 8c). The shallow aquifers are found to be contaminated with As in the state of Bihar whereas the deeper aquifers (>60 mbgl) are found to be free from contamination from both sides of the river Ganga (Saha et al 2010; Saha and Shukla 2013). These observations are directly in line with previous observations by Bhattacharya et al. (1997) and Mukherjee et al. (2018), where high As was reported at shallower depths of 6-13 m. In half of the samples, a high concentration of As was associated with a high concentration of HCO_3^- in the groundwater samples taken at a depth between 6 and 15 m (Figure 8d). The low concentration of HCO_3^- was found mostly in samples with low As at depth between 3 and 7 m. Studies suggest that this pattern corresponds to the most important anion species, which competes with As for adsorption sites at mineral surfaces (e.g., Fe and Mn oxyhydroxides and clay minerals), which consequently releases As into the groundwater (Mukherjee et al., 2018).

Discussion

Almost 90% of the samples have EC in marginal category (500-1500 μS/cm). However, no relationship was found between As and EC in the groundwater of study area. The spatial distribution of As is found to be variable and heterogeneous in nature. Low concentration of As was found in the groundwater samples located in regions of steeper slopes. The lower the slope more higher is the As concentration (Wang et al., 2018). This can be seen from Figure 6 which confirms that low-lying plains would have high As concentrations in groundwater (Buschmann et al., 2007). Similar relationships have been identified by other studies in the Bengal and Mekong river basins (Mukherjee and Fryar, 2008; Buschmann, 2007). Sharda river basin has frequent monsoon floods, forming a lot of abandoned channels. This leads to waterlogging, which causes surface water retention, resulting in the reaction of CO_2 and water and leading to the formation of carbonic acid, which then percolates into the subsurface (van Geen et al., 2008). Since carbonic acid is a weak acid, it breaks downs into HCO_3^- and H^+ ions. This contributes to HCO_3^- in the aquifer, leading to reducing conditions in the aquifers (Postma et al., 2016). The groundwater samples have a very high concentration of HCO_3^-, thus, resulting in reducing conditions in groundwater. Low NO_3^- was observed in high As samples. As a result of denitrification, the reduction of the NO_3^- to ammonia occurs along with the formation of HCO_3^-. Few high NO_3^- samples were found that could be due to anthropogenic activities (Mueller et al., 2018). It can also be seen that there is an overall negative correlation between As and NO_3^- (-0.21) although the value is not very significant (Table 4). Furthermore, this sequential microbially mediated process of denitrification, metal reduction, sulphate reduction (NO_3^-, Mn, Fe and SO_4^{2-}) plays a significant role in mobilisation of As in the groundwater (Mahanta et al., 2015). The dissolution process of iron oxyhydroxides causes surface desorption of As from these oxides, resulting in increased concentrations of As along with HCO_3^- and Fe in groundwater. Figure 8b shows that in a few samples, the high concentration of As was associated with a high concentration of HCO_3^- which was found to be confined to a depth between 6 and 15 m.

The low HCO_3^- concentrations are found mostly in samples with low As at depth between 3 and 7 m (Figure 8c and 8d). This pattern corresponds to the most important anion species, which competes with As for adsorption sites at mineral surfaces (e.g., Fe and

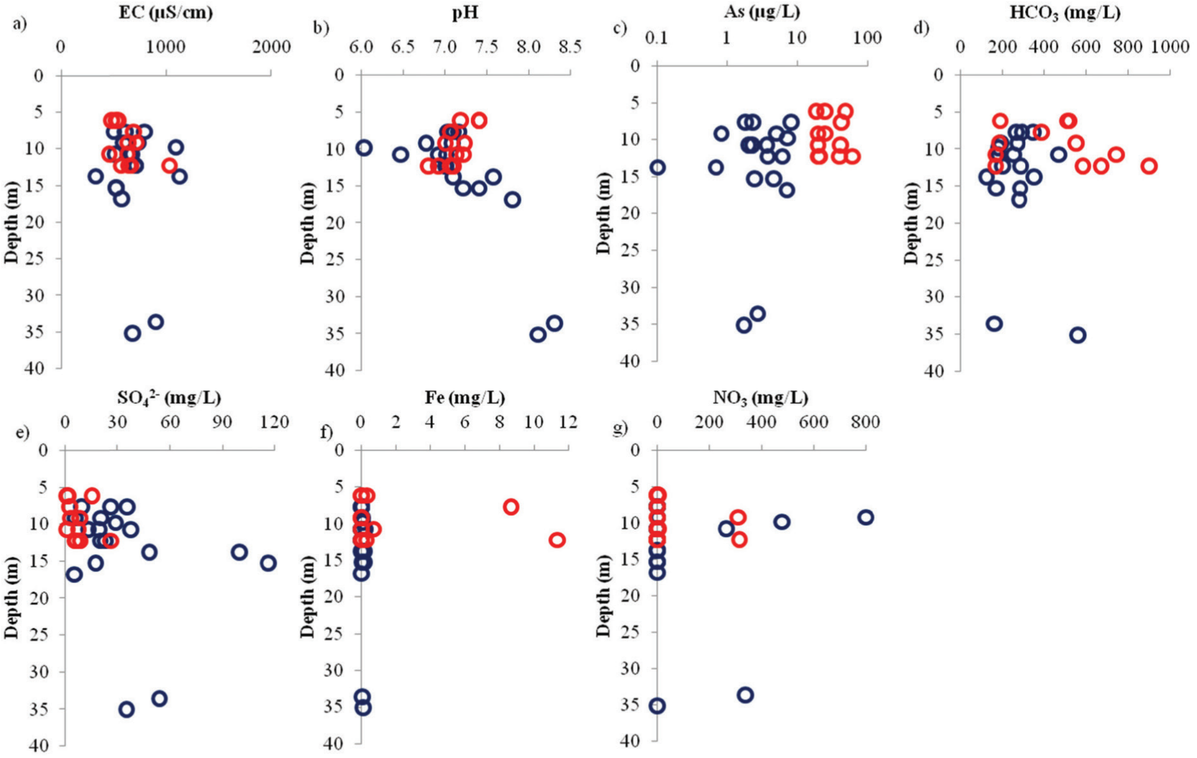

Figure 8: Depth profile of the major cations and anions along with pH and EC with As in log plot, blue circle denotes As < 10 µg/L and the red circle indicate As > 10µg/L.

Mn oxyhydroxides, and clay minerals), consequently releasing As into the groundwater (Mukherjee et al., 2011; Mahanta et al., 2015). We have also observed a high positive correlation among Fe and Mn (0.86) and Al (0.59) indicating their common source of origin. This indicates that minerals comprised of these metals such as Mn oxides, Fe oxides and Al oxides are the sources for As. This observation is supported by poor correlation (-0.21) between As and NO_3^- (Figure 8g), which highlights that apart from dissolved O_2, NO_3^- is another thermodynamically favoured electron acceptor for microbial degradation of dissolved organic matter in the shallow aquifers of the study area (Rowland et al., 2017).

Moreover, there is a positive correlation observed between Fe and PO_4^{3-} (Table 4). The competitive exchange of anions by PO_4^{3-} and desorption from metal oxide surfaces due to the increased alkalinity may also be responsible for mobilisation of As (Stollenwerk et al., 2007). In contrast, PO_4^{3-} is adsorbed strongly onto solid phases of mainly Fe and Al oxides in soils (Zahid et al., 2008). However, the amount of PO_4^{3-} released into water is related to the concentration of PO_4^{3-} that exceeds the capacity of Fe to create insoluble iron phosphate. However, PO_4^{3-} may be released into groundwater due to microbially mediated reductive dissolution of Fe-oxyhydroxides (McArthur et al., 2004). Furthermore, K^+ and PO_4^{3-} also have a positive correlation and could reflect their common source of origin which can be anthropogenic, mainly the fertilisers.

Figure 9 depicts that there are few samples that have high Fe and Mn along with high As however not all the samples show a similar relationship. This lack of correlation between Fe and As indicates the decoupling between mobilisation of As and Fe (Table 4). Mueller et al., 2018, also suggested that the process which mobilises As and Fe are decoupled, which could be due to reprecipitation of Fe after reduction of Fe oxyhydroxides, while the mobilised As remains in a soluble form. This low Fe concentration can also be due to its precipitation to a stable phase from an unstable Fe-oxyhydroxides phase (Guo et al., 2013). Since there is low SO_4^{2-} concentration, Fe does not precipitate as pyrite. However, it might precipitate as siderite. Studies suggest groundwater with high HCO_3^- and high Fe, show siderite formation, which leads to limited availability of Fe in groundwater. There is a poor but positive correlation among As and Fe (0.28), Mn (0.11) and Al (0.31). Similar findings have been reported where the poor correlation between As and

Table 4: Correlation among various parameters measured for the samples collected across the Sharda river basin

	As	*EC*	Mn^{+}	Fe^{3+}	Na^{+}	K^{+}	Ca^{2+}	Mg^{2+}	Cl^{-}	SO_4^{2-}	NO_3^{-}	HCO_3^{-}	PO_4^{3}-
As	1												
EC	-0.15	1											
Mn^{+}	0.11	0.39^{*}	1										
Fe^{3+}	0.28	-0.02	0.31^{*}	1									
Na^{+}	-0.47	0.39^{*}	0.28	-0.27	1								
K^{+}	0.04	0.48^{**}	**0.89****	0.31^{*}	0.36^{*}	1							
Ca^{2+}	-0.29	**0.65****	0.43^{**}	0.04	0.31^{*}	**0.49****	1						
Mg^{2+}	0.19	0.10	0.29	0.31^{*}	-0.13	0.18	0.28	1					
Cl^{-}	-0.39^{*}	**0.55****	0.20	-0.17	**0.72****	0.34^{*}	**0.53****	-0.13	1				
SO_4^{2-}	-0.39^{*}	0.38^{*}	0.31^{*}	-0.10	0.42^{*}	0.34^{*}	**0.62****	-0.08	**0.73****	1			
NO_3^{-}	-0.21	0.37^{*}	0.23	0.01	-0.02	0.08	0.40^{*}	**0.52****	0.00	0.13	1		
HCO_3^{-}	**0.55****	-0.18	0.27	0.25	-0.12	0.32^{*}	-0.17	0.26	-0.09	-0.21	-0.22	1	
PO_4^{3}-	0.10	0.35^{*}	**0.97****	0.28	0.25	**0.89****	0.40^{*}	0.27	0.17	0.24	0.19	0.29	1

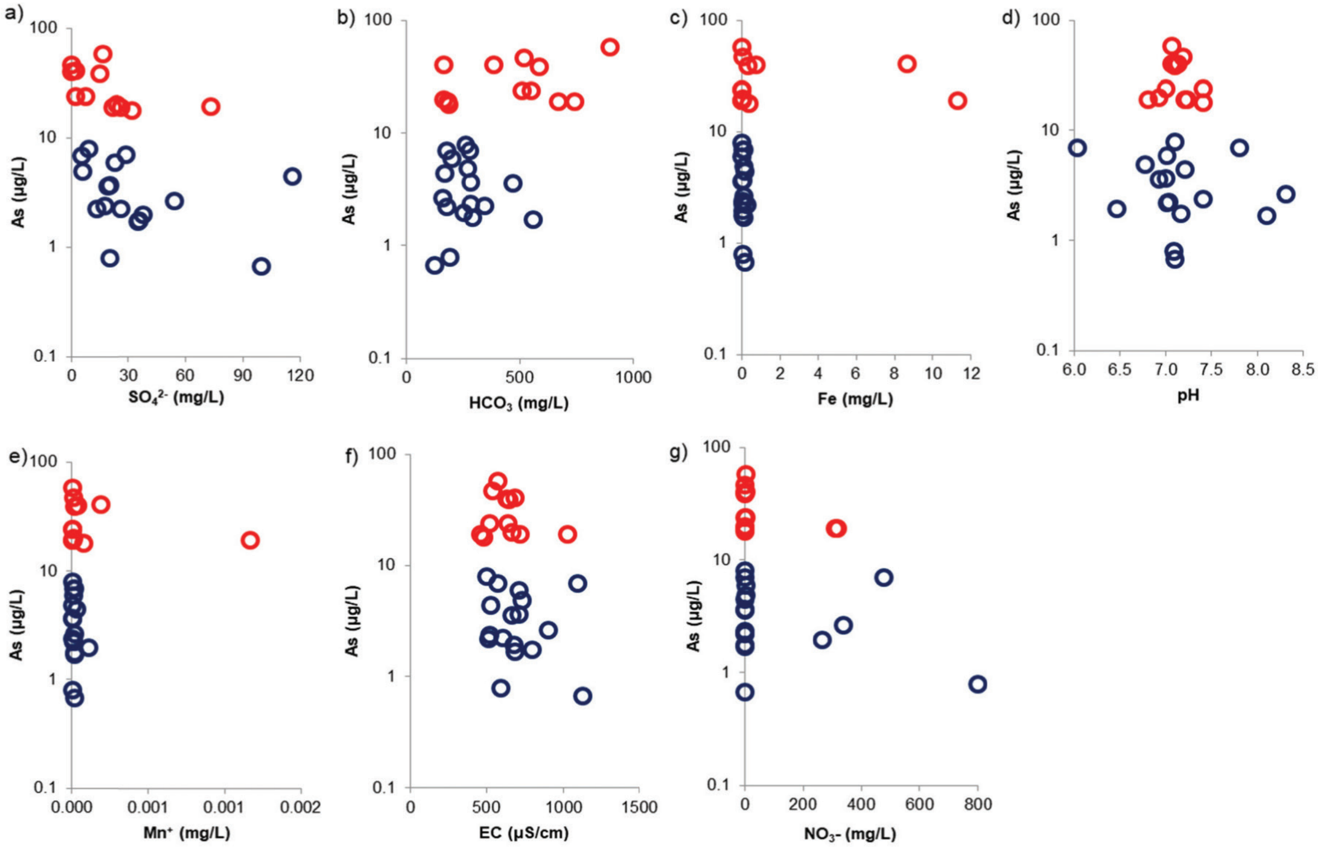

Figure 9: Scatter plot between As (log plot) and other water parameters, blue circle denotes As < 10 µg/L and the red circle indicates As > 10 µg/L.

Fe have been found (Nickson et al., 2000; Mahanta et al., 2015). Thus, it can be stated that the process of reductive dissolution of Fe-oxyhydroxides is found as the major mechanism for As release in groundwater in the Sharda river basin. The study area, which is also part of the active floodplain, where microbial oxidation of organic matter takes place. This might lead to Fe-oxyhydroxides reduction and high HCO_3^- formation due to silicate and carbonate weathering (Saha and Sahu, 2016).

In the cultivation of rice, the flooded irrigation method is the major agricultural practice in the region. Rice cultivation can lead to the formation of an impermeable layer of water between land surface and atmosphere. Studies suggest that this might be responsible for high organic matter content, which is oxidised by microbes and leads to reducing conditions in shallow aquifers (van Geen et al., 2014b; Williams et al., 2006). It can be seen from Figure 1 that most of the high As samples belong to the younger alluvium deposits. The studies also suggest dissolved As remains to be high in areas with restricted local recharge by the surface cover of low permeability (van Geen et al., 2014b). Over the past 30 years, there has been a shift in cropping patterns resulting in an increase in cultivated agricultural area to 62.08%. Rice cultivation has increased from 2002-03 to 2014-15, i.e. 180163 to 182548 hectares increase in area as compared to yield increase from 2.38 to 17.96 tonnes/hectare over the same land holdings (Indiastat, 2014-2015). This overburden on land leads to over pumping of groundwater for irrigation purposes which might have led to As dissolution in groundwater. Therefore, the handpumps installed over newer alluvial deposits might be more prone to As contamination.

Policy Implication: Switching to a Safe Well

The study indicates that the Sharda River basin is under the threat of As contamination. Blanket testing can be prioritised by the government or non-governmental organisations. Results from this study can be used as a guide for government and policymakers to downscale their sites of action and provide interventions in the affected regions belonging to newer alluvium. Further, these regions can be targeted for proper mitigation measures and access to As free drinking water can be ascertained. Since the spatial distribution of As is spatially heterogeneous; the use of reliable As field kits can be incorporated to maximise testing of handpumps along with quality control. Alongside, well switching is a significant mitigation option in As affected areas until a long term sustainable solution is established. However, the viability of well switching is based on blanket testing of the handpumps. In Bihar, indulging handpump owners into a social network leading to safe drinking water have been tried to address this problem. This resulted in people's ability to differentiate safe/unsafe handpumps and share among neighbours (Barnwal et al., 2017). This is an example of regular interventions after the baseline survey resulted in 30.5% higher switching to a safer well. Similar patterns of switching results of 26-41% were obtained in Bangladesh by community participation. The costs and logistics of treating and supplying water are considerably higher and most of the time have been prohibitive due to logistics, its operation and maintenance issues.

For most cases in the Gangetic plains, the local population uses traditional cleaning and filtering due to lack of alternatives for maintaining a clean drinking water supply for themselves (Ahmad et al., 2003). Alternatively, the use of groundwater received from As-free deep handpumps can be used. Researchers have conducted studies in parts of UP and Bihar, in the middle Gangetic plain, and have emphasised on the usage of deep aquifers for drinking water supply (Ramanathan, 2015; Saha et al., 2011; Singh, 2015). These alternatives are viable options that may potentially influence the human ability to access As-free drinking water and reduce health risks. Awareness of the current quantitative and qualitative status of groundwater in the location of high As-risk zones located in the newer alluvium in conjunction with socio-economic factors, therefore, can be crucial to determining the potential exposure of people due to As.

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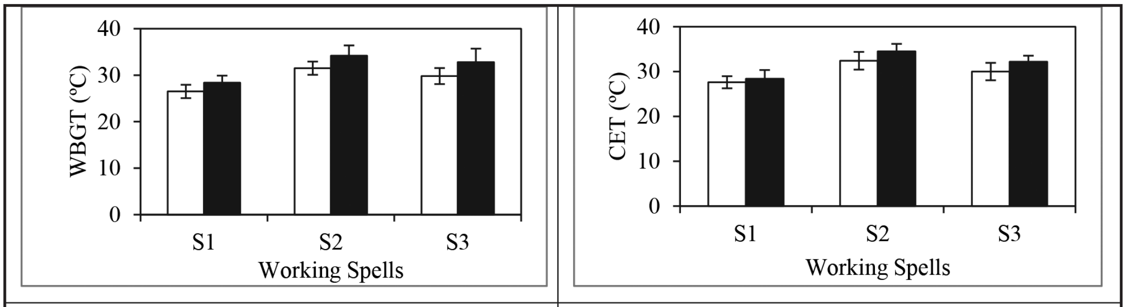
Errata

Please read Table 2 and Figure 1 on page 61 of Volume 6 Number 1 issue of *Journal of Climate Change* as given below.

Table 2: Physical and physiological parameters of the study participants

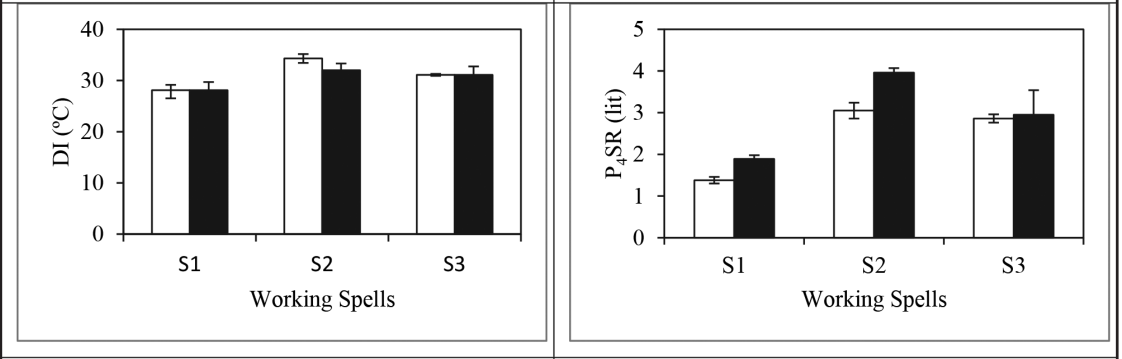
Variables	*Values*
Stature (cm)	158 ± 4.85
BW (kg)	55.4 ± 3.31
BMI	21.0 ± 4.55
$HR_{Pre\text{-}work}$(beats.min^{-1})	70.0 ± 3.58
$SBP_{Pre\text{-}work}$ (mm Hg)	119.0 ± 10.80
$DBP_{Pre\text{-}work}$ (mm Hg)	74.0 ± 9.38

Data presented as AM ±SD



a) WBGT

b) CET



c) MDI

d) P4SR

□RG- A, ■ RG - B

Figure 1: Environmental condition in terms of WBGT, CET, MDI and P_4SR.