

# Tracing the factors responsible for arsenic enrichment in groundwater of the middle Gangetic Plain, India: a source identification perspective

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**Abstract** Arsenic contamination in groundwater is of increasing concern because of its high toxicity and widespread occurrence. This study is an effort to trace the factors responsible for arsenic enrichment in groundwater of the middle Gangetic Plain of India through major ion chemistry, arsenic speciation, sediment grain-size analyses, and multivariate statistical techniques. The study focuses on the distinction between the contributions of natural weathering and anthropogenic inputs of arsenic with its spatial distribution and seasonal variations in the plain of the state Bihar of India. Thirty-six groundwater and one sediment core samples were collected in the pre-monsoon and post-monsoon seasons. Various graphical plots and statistical analysis were carried out using chemical data to enable hydrochemical evaluation of the aquifer system based on the ionic constituents, water types, hydrochemical facies, and factors controlling groundwater quality. Results suggest that the

groundwater is characterized by slightly alkaline pH with moderate to strong reducing nature. The general trend of various ions was found to be  $\text{Ca}^{2+} > \text{Na}^+ > \text{Mg}^{2+} > \text{K}^+ > \text{NH}_4^+$ ; and  $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{PO}_4^{3-} > \text{F}^-$  in both seasons. Spatial and temporal variations showed a slightly higher arsenic concentration in the pre-monsoon period (118  $\mu\text{g/L}$ ) than in the post-monsoon period (114  $\mu\text{g/L}$ ). Results of correlation analyses indicate that arsenic contamination is strongly associated with high concentrations of Fe,  $\text{PO}_4^{3-}$ , and  $\text{NH}_4^+$  but relatively low Mn concentrations. Further, the enrichment of arsenic is more prevalent in the proximity of the Ganges River, indicating that fluvial input is the main source of arsenic. Grain size analyses of sediment core samples revealed clay (fine-grained) strata between 4.5 and 7.5 m deep that govern the vertical distribution of arsenic. The weathering of carbonate and silicate minerals along with surface-groundwater interactions, ion exchange, and anthropogenic activities seem to be the processes governing groundwater contamination, including with arsenic. Although the percentage of wells exceeding the permissible limit (50  $\mu\text{g/L}$ ) was less (47%) than that reported in Bangladesh and West Bengal, the percentage contribution of toxic As(III) to total arsenic concentration is quite high (66%). This study is vital considering that groundwater is the exclusive source of drinking water in the region and not only makes situation alarming but also calls for immediate attention.

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## Introduction

The World Health Organization (WHO) has repeatedly insisted that the single major factor adversely influencing the general health and life expectancy of a population is the lack of ready access to clean drinking water (Kumar et al. 2007). Because of severe problems of water stress and deterioration of water quality, there is high concern about groundwater quality all over the world. The quality of groundwater depends on the composition of recharging water, the mineralogy and reactivity of the geological formations in aquifers, the impact of human activities, and environmental conditions that may affect the geochemical mobility of certain constituents (Kumar et al. 2006). These geochemical processes are responsible for seasonal and spatial variations in groundwater chemistry (Matthess 1982; Kumar et al. 2009a). Groundwater evolves chemically by interacting with aquifer minerals or by internal mixing among different groundwater along flow paths in the subsurface (Domenico 1972). The presence of trace elements in groundwater is an important issue because it affects possible uses of water. Arsenic contamination of groundwater, in particular, is of increasing concern because of the high toxicity and widespread occurrence of this element (Jain and Ali 2000; Smedley and Kinniburgh 2002).

Further, exposure to elevated levels of arsenic (As) in groundwater has become a global concern in recent years (Meliker et al. 2008). Most estimates of arsenic pollution have focussed on the predominance of arsenic poisoning in the groundwater of West Bengal (India) and Bangladesh (Bhattacharya et al. 1997; Ben et al. 2003; Ahmed et al. 2004), which has been thought to be limited to the Ganges Delta (the lower Gangetic Plain). Early surveys have been conducted on arsenic contamination in groundwaters of West Bengal (Saha 1984; Mazumder et al. 1998) and groundwater in the Union Territory of Chandigarh and its surroundings in the northwestern upper Ganga

Plain and recent findings in the Tarai areas of Nepal (Chakraborti et al. 2003). Several authors have suggested that the reductive dissolution of Fe(III) oxyhydroxides under strongly reducing conditions of the young alluvial sediments is the cause for As mobilization (Ahmed et al. 2004; Bhattacharya et al. 1997; Harvey et al. 2002; McArthur et al. 2004; Nickson et al. 1998; Nickson et al. 2000). The reduction is driven by microbial degradation of sedimentary organic matter, which is a redox-dependent process consuming dissolved  $O_2$  and  $NO_3$  (Nickson et al. 2000).

Gradually, researchers have shifted their focus to vertical profiling of arsenic contamination (Datta and Subramanian 1994; Acharyya and Shah 2004; Acharyya 2004, Nickson et al. 2007; Shah 2008). It was an important step towards precise understanding of vertical mobilization of As and role of geology of the area. However, most of the studies have correlated sediment colour with As concentrations (Brömssen et al. 2007, 2008). Very few attempts have been made to correlate grain-size distribution with As contamination. Further, it is recognized that the environmental impact and mobility of an element is not purely controlled by total concentration but the speciation of the element. For example in the case of arsenic, As(III) is more toxic than As(V). Therefore, inclusion of spatial distribution of different species becomes imperative for complete discussion of arsenic contamination.

Furthermore, the arsenic contamination in the alluvium plain of the Gangetic Plain may cause secondary effects when it enters the food chain. The arsenic contamination may result from a combination of natural and anthropogenic processes, for example weathering reactions, biological activity, mining activity, combustion of fossil fuels, use of arsenical pesticides, herbicides, and crop desiccants, and use of arsenic as an additive to livestock, particularly for poultry feed (Huq et al. 2001). Therefore, it is essential to distinguish between the contributions by natural weathering and anthropogenic inputs. Globally, contaminated drinking water is the chief source of chronic human intoxication (Gebel 2000; Smith et al. 2000) and may result in skin ailments, such as hyper-pigmentation and kurtosis, and progress to cancer and, ultimately, death. The current drinking water quality guideline for arsenic is  $10 \mu\text{g/L}$  (WHO 1993). The current standard for arsenic in drinking

water in both Bangladesh and India is 50 µg/L. Moreover, the upper, middle, and lower Gangetic Plains are the most densely populated areas of India. Moreover, in these parts of India dependence on groundwater has increased tremendously, which intensifies the likely health problem from groundwater contamination. Thus, there is an immediate need for an inventory which can take account of the amount of arsenic actually present in the area and number of people actually exposed to it.

In the work discussed in this paper an effort was therefore made to reveal the spatial distribution and predict the seasonal changes in the levels of various contaminants, to quantify the occurrence of different arsenic species, and to understand the mechanism controlling the occurrence of arsenic and to distinguish between the contributions made by natural weathering and anthropogenic inputs. An effort has been also made to correlate grain-size distribution of sediment with depth-wise variation of As concentration.

**Study area**

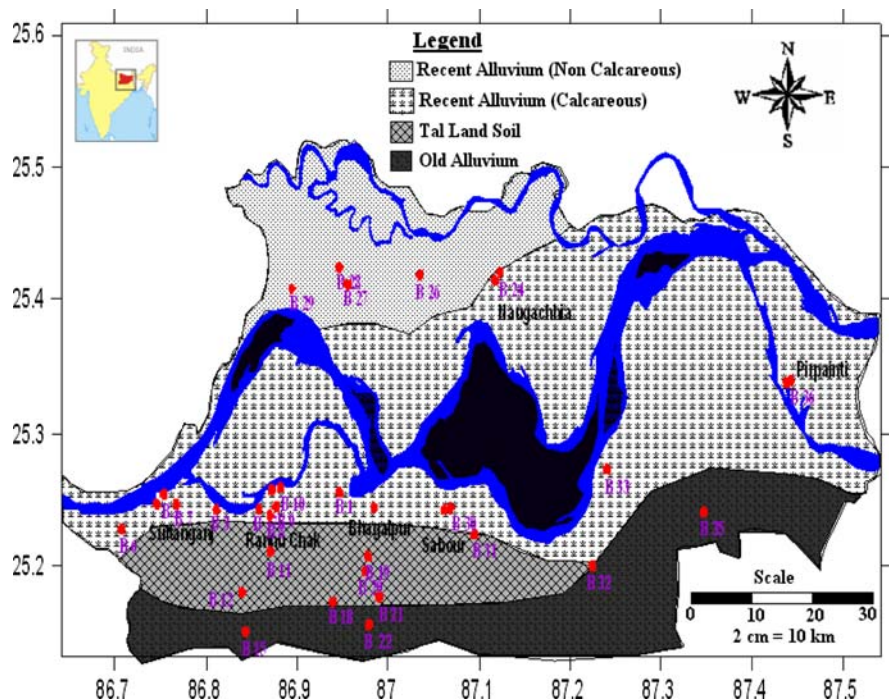
Bhagalpur district is located between 25°13.29' and 25°608' N and from 86°64.08' to 87°542' E (Fig. 1).

The district is a peneplain, intersected by numerous streams. Surface levels varied because of high banks of the Ganga, Koshi (Ghugri), Chanari, and Chandan. The geomorphology of the area is a monotonously flat and featureless plain with gradient towards the river Ganges. The southwestern monsoon brings much-needed rainfall and nearly 70% of the rainfall occurs during the months of July, August, and September. There is significant seasonal variation in temperature with mean annual temperature of 26°C. The major part of the district is characterised by an arid soil moisture regime according to the criteria laid in Soil Taxonomy (Brady and Weil 2002).

**Hydrogeology**

Geomorphologically the area is represented by alluvial deposits of Quaternary age. The area has been divided into four different zones: recent alluvium (non-calcareous), recent alluvium (calcareous), Tal land soils, and old alluvium (Fig. 1). The Gangetic Plain has huge amounts of river-deposited sediments which accumulate on the flood and deltaic plain of Bengal. They generally consist of Pleistocene and Holocene formations. However, the central Gangetic Plain has a unique geological geo-morphological

**Fig. 1** General geomorphology of the Bhagalpur district, including sampling locations



setting with aquifers ranging from Quaternary Himalayan alluvial fans and plains to the pre-Cenozoic (Precambrian to Cretaceous) Indian cratonic, igneous, and meta-sediment shield provinces. XRD studies on soil samples of As-safe older alluvial and As-contaminated newer alluvium from the Middle Gangetic Plain reveals an mineralogical assemblage of quartz, muscovite, chlorite, kaolinite feldspar, amphibole, and goethite (Shah 2008). The groundwater occurs in Holocene sandy sediments and forms extensive unconfined to leaky confined aquifers, i.e., the area has both confined and unconfined aquifers. Water level fluctuates with seasonal recharge and discharge. The depth of wells varies from 6 to 13 m bgl. The depth to water level has been observed to vary from 3 to 8 m bgl during the pre-monsoon season and from 2 to 4 m bgl during the post-monsoon season. The multiple aquifer system of this region has variable hydraulic conductivity. The deeper aquifers i.e., semi-confined aquifers are prolific aquifers and may be a future water resource supplies for the region with good hydraulic characteristics. The district has two means of irrigation—canals and tube wells (groundwater).

## Materials and methods

Sampling wells were selected in such a way that they represent different geological formations and land-use patterns at varying topography of the area. Thirty-six groundwater samples were collected during May and November 2007 (Fig. 1) in order to observe the impact of the monsoon season on arsenic and other contaminant status of the area. Groundwater samples were collected in clean polyethylene bottles. For bore well and hand pumps, the water samples were collected after pumping the water for 5–10 min. For open wells, water samples were collected 30 cm below the water level using a depth sampler.

In-situ measurements mainly EC, PH, and ORP were measured by use of a portable Orion Thermo water analysing kit (Model Beverly, MA, USA; 01915). Total arsenic was determined with the help of a Digital Arsenator (Wagtech, UK) and arsenic speciation was performed in the field with disposable cartridges (Metal Soft Center, PA, USA) which absorb As(V), but allow As(III) to pass through. Further, these samples were stored below 4°C in a

portable ice-box to minimize chemical alteration. The collected groundwater samples were classified for anion and cation analysis. Further, groundwater samples were filtered through 0.45- $\mu\text{m}$  Millipore filter paper and acidified with 2 M  $\text{HNO}_3$  (Ultra pure; Merck) for cation analysis.  $\text{HBO}_3$  acid was used as preservative for nitrate analysis (Kumar et al. 2009b).

Fe and Mn were analysed in the laboratory by use of an atomic absorption spectrophotometer (Shimadzu AA-6800). Concentrations of total arsenic were cross-checked on acidified samples using graphite-furnace (GF) AAS (Shimadzu AA-6800) in absorption mode using chemical standards; the detection limit was 2  $\mu\text{g/L}$ . Major cation analysis ( $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ ) was carried out by use of an EEL flame photometer (APHA 1995). The concentration of  $\text{HCO}_3^-$  was measured by acid titration. Other anions  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{PO}_4^{3-}$  were analysed by use of a Dionex DX-120 ion chromatograph. Other characteristics, for example  $\text{SiO}_2$  and  $\text{NH}_4^+$  were analysed with a Jenway model 6505 spectrophotometer. High-purity reagents (Merck) and Milli-Q water (Model Milli-Q, Biocel) were used for all the analysis.

Analytical precision was checked by normalized inorganic charge balance (NICB) (Huh et al. 1998; Kumar et al. 2006). This is defined as  $[(\text{Tz}^+ - \text{Tz}^-)/(\text{Tz}^+ + \text{Tz}^-)]$  and represents the fractional difference between total cations and anions. In general, samples showed a charge imbalance mainly in favour of positive charge. The observed charge balance support the quality of the data points, which is better than  $\pm 5\%$  except for some samples.

## Sediment grain-size analysis

With the help of local drillers one borehole was drilled to confirm different lithological units and its relationship with arsenic enrichment. During drilling, samples were collected on the basis of visible change in sediment colour and texture. The change was rapid initially and thus samples were collected every 1.5 m; later the depth interval between two samples collected was 3 m. Thus, thirteen ( $n = 13$ ) samples were obtained from the single core sediment. Only washed and disturbed sediments could be sampled as a result of the hand percussion technique. Washed sediments were collected in a bucket and allowed to settle before being transferred on to a bamboo carpet. Later,

the sediment samples were allowed to drain (but not dry), before putting them into plastic bags (Brömssen et al. 2007). Samples were air-dried in the laboratory. Stones and plant fragments were removed by passing the dried samples through a 2-mm sieve. The sieved samples were powdered and finally passed through ASTM standard sieves. Size fractions of the bed sediments down to 37 μm were separated and fractionated using the Astenburg cylinder method based on Stoke’s law (Griffiths 1967).

### Statistical analysis

The matrix of hydrogeochemical data obtained was subjected to multivariate analytical techniques. These techniques help to simplify and organize large data sets in order to make useful generalizations. At first data was subjected to correlation analysis using Spearman’s rank coefficient which is based on the

ranking of the data and not their absolute value. Thereafter, factor analysis was performed for effective display of complex relationships among many samples (Kumar et al. 2009c). These analyses were performed using Statistical Package for Social Sciences (SPSS) software package (Version 10.5).

## Result and discussion

### General groundwater chemistry

A statistical summary of the analytical results (minimum, maximum, mean, and the standard deviation) for each water-quality characteristic analysed is given in Table 1.

Result for pH values at different lithology showed that the groundwater was alkaline in nature (Kumar et al. 2007). pH, EC, and Cl<sup>-</sup> values were higher in

**Table 1** Statistical summary of hydro-geochemical parameters of the groundwater

Characteristic	Unit	Minimum		Maximum		Average		SD	
		Pre-monsoon	Post-monsoon	Pre-monsoon	Post-monsoon	Pre-monsoon	Post-monsoon	Pre-monsoon	Post-monsoon
Ph		7.8	7.66	8.3	8.16	8.13	7.98	0.13	0.13
ORP	mV	-134	-125	169	135	8.97	7.91	95.19	86.68
EC	μs/cm	250	217	980	967	599	591	175	174
TDS	mg/L	191.25	185	710	685	455	441	133	128
Na <sup>+</sup>	mg/L	5.76	5.16	41.91	37.58	18.66	17	8.54	8.13
K <sup>+</sup>	mg/L	0.44	0.43	3.22	3.16	1.45	1.39	0.70	0.66
Ca <sup>2+</sup>	mg/L	20.7	18.87	126	161	64.81	69.4	30.28	37.91
Mg <sup>2+</sup>	mg/L	7.83	8.36	18.2	19.43	10.67	11.5	2.21	2.28
HCO <sub>3</sub> <sup>-</sup>	mg/L	15.2	23.74	214.5	266	105	129	42.19	57.68
F <sup>-</sup>	mg/L	0.08	0.06	4.94	2.48	0.76	0.63	0.86	0.50
Cl <sup>-</sup>	mg/L	6.56	6.08	219.93	154	69.67	55.52	50.78	35.89
NO <sub>3</sub> <sup>-</sup>	mg/L	1.01	1.12	39	46.7	23.71	22.66	11.59	13.33
SO <sub>4</sub> <sup>2-</sup>	mg/L	3.84	3.61	72.8	68.4	32.93	27.68	20.19	16.65
PO <sub>4</sub> <sup>3-</sup>	mg/L	2.66	2.36	6.37	5.65	4.05	3.59	0.95	0.84
H <sub>4</sub> SiO <sub>4</sub>	mg/L	14	17.27	49.33	55.55	30.01	35.1	8.15	9.24
NH <sub>4</sub> <sup>+</sup>	mg/L	0.59	0.54	3.11	2.83	1.38	1.26	0.76	0.70
Fe	mg/L	0.66	0.43	7.62	6.84	3.19	2.82	2.23	1.98
Mn	mg/L	0.01	0.01	1.79	1.75	0.66	0.63	0.56	0.55
As (tot)	μg/L	19.1	18.46	118	113.5	51.23	48.97	27.64	25.76
As <sup>3+</sup>	μg/L	10.9	11.63	81.1	75.9	34.43	32.73	21.04	18.04
As <sup>5+</sup>	μg/L	7.1	5.32	55.2	47.89	16.81	16.24	10.28	11.49

the pre-monsoon season whereas  $\text{HCO}_3^-$  values were higher in the post-monsoon season. On the other hand, no significant seasonal variations were observed for  $\text{SO}_4^{2-}$  (Table 1). Higher values of pH and EC in the pre-monsoon season are the combined effect of the high concentration of dissolved solids and/or high ionic strength of the groundwater, local variation in soil type, multiple aquifer system, and agricultural activities in the area. Higher  $\text{HCO}_3^-$  concentrations in the post-monsoon period are because of weathering of carbonaceous sandstones by rain water followed by subsequent precipitation of  $\text{HCO}_3^-$  along with other cations.

The higher and lower concentrations of  $\text{Cl}^-$  in the pre-monsoon and post monsoon seasons, respectively, may be because of the input from sewage effluents in the village areas and dilution by rain water in post monsoon season (Todd 1959). The groundwater seems to have secondary salinity, as indicated by high  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  concentrations. The presence of salts in the unsaturated top zone of the groundwater suggests that the flushing rate of the aquifers may be slow because of high  $\text{Cl}^-$  concentrations trapped in clayey lenses, which may be gradually diffusing into the aquifer.

The average concentration of  $\text{NO}_3^-$  was found to be as high as 23 mg/L (Table 1), which indicates the influence of agricultural activities, fertilizer use, and microbial mineralization on the groundwater. Further, the higher concentration of nitrate may also be a result of the presence of *E. coli*, *Staphylococcus aureus*, *Proteus vulgaris*, *Salmonella typhi*, and *Pseudomonas aeruginosa* in the faecal matter, as reported by Saha and Kumar (2006). This seems reasonable as there is a severe lack of a proper sanitation system in the district. Further, animal waste sources are also a significant contributor of nitrate to groundwater, especially within mixed land-use. Some parts of the study area is occupied by industrial and urban land-use, therefore some nitrate leaching from landfill sites and industrial effluents cannot be neglected. The underlying cause of  $\text{PO}_4^{3-}$  in the groundwater of the Bhagalpur district indicates input of fertilizers in farmlands to enhance paddy and wheat productivity and the dilution effect in the rainy season. In Bhagalpur,  $\text{F}^-$  varied significantly from 0.08 to 4.94 mg/L in the pre-monsoon season and 0.06 to 2.48 mg/L in the post-monsoon season

(Table 1), which is an indicator of weathering of mica containing mineral like biotite. Pollution and health aspects of this have already been reported by Chaurasia et al. (2007). Finally, among anions the average value trend found was  $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{PO}_4^{3-} > \text{F}^-$  in both seasons.

The dominant cation was  $\text{Ca}^{2+}$  followed by  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ , and  $\text{NH}_4^+$ . There was little seasonal variation in most of the cations, except a few locations where seasonal variations were significant (Table 1). In the post-monsoon season, high concentrations of  $\text{Ca}^{2+}$  may be because of weathering of carbonate mainly from gypsum, plagioclase and feldspar minerals, which are abundant in the floodplain regions (Bhattacharya et al. 1997). However, the results for  $\text{Na}^+$  and  $\text{K}^+$  followed the opposite pattern with high concentrations in the pre-monsoon season (Table 1). This implies that the contribution of cations via alumino-silicate weathering is low in comparison with carbonate weathering. The average ratio trend of  $\text{Ca}^{2+} + \text{Mg}^{2+}/\text{Na}^+ + \text{K}^+$  varied from 4.65 in the pre-monsoon season to 5.45 in the post-monsoon season, indicating the dominance of carbonate rock weathering in the groundwater of Bhagalpur. Further, the concentration of  $\text{H}_4\text{SiO}_4$  varied from 14 to 49 mg/L in the pre-monsoon season and from 17 to 56 mg/L in the post-monsoon season (Table 1). The slight increment in concentration indicates the possibility of alumino-silicate weathering in the rainy season.

Total arsenic concentrations in the groundwater varied from 19.1 to 118  $\mu\text{g/L}$  in the pre-monsoon season and from 18.5 to 113.5  $\mu\text{g/L}$  in post-monsoon season (Table 1). The concentration of Fe varied from 0.66 to 7.62 mg/L in the pre-monsoon season and from 0.43 to 6.84 mg/L in the post-monsoon season whereas Mn values were very low in both seasons (Table 1). This high concentration of Fe and low concentration of Mn is indicative of a reducing environment in the groundwater environment of the area. The concentration of  $\text{NH}_4^+$  varied from 0.59 to 3.11 mg/L in the pre-monsoon season and from 0.54 to 2.83 mg/L in the post-monsoon season (Table 1).  $\text{NH}_4^+$  is a good indicator of contamination from inadequate sanitation facilities. The finding supports an earlier argument pertaining to higher  $\text{NO}_3^-$  and further shows that some dilution occurs in the post-monsoon season.

Graphical representation of hydrochemical data

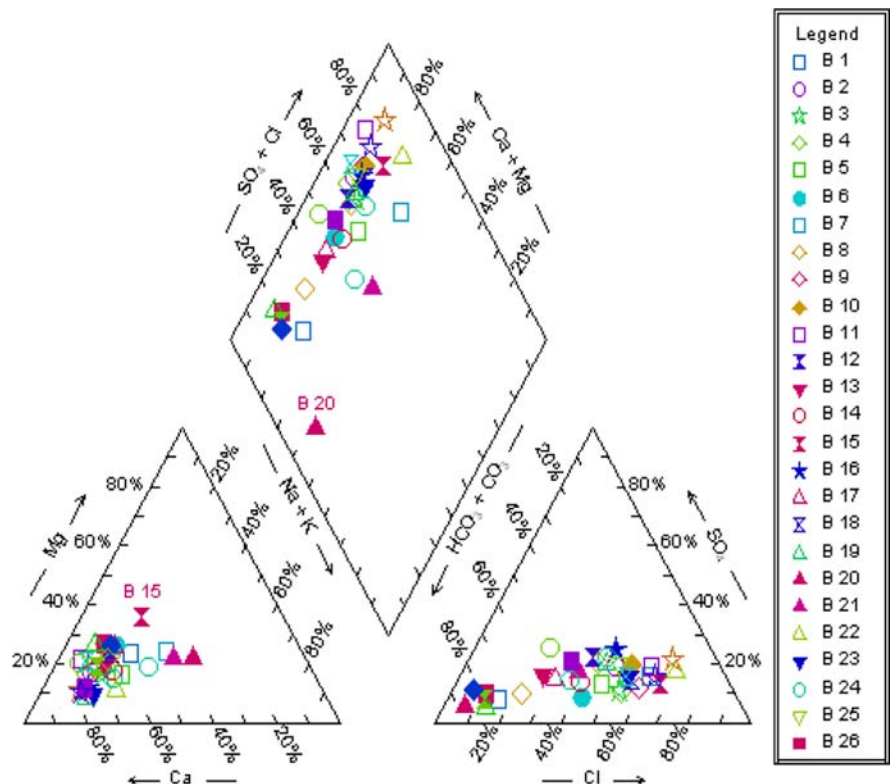
In this study, water-quality data were analysed by use of Piper diagrams to gain better insight into the hydro-geochemical processes operating in the hydro-geochemical processes operating in the groundwater environment of the central Gangetic Plain that resulted in the spatial and temporal variation. The major cations and anions for the analysed water were plotted on a Piper diagram (Figs. 2, 3). It was found that not only were the water samples of Ca-Cl (about 62%) and Ca-HCO<sub>3</sub> (about 36%) type in the pre-monsoon period but also remained so in the post-monsoon period, with slight changes in their percentage share of each water type of Ca-HCO<sub>3</sub> (~59%) and Ca-Cl (~40%). This indicates the presence of carbonaceous sandstone in the aquifers and weathering of carbonate minerals in the post-monsoon period whereas in the pre-monsoon period the result favours salt precipitation. In most places hardness is of the CaCO<sub>3</sub> type with some locations saturated with calcite.

Statistical analysis

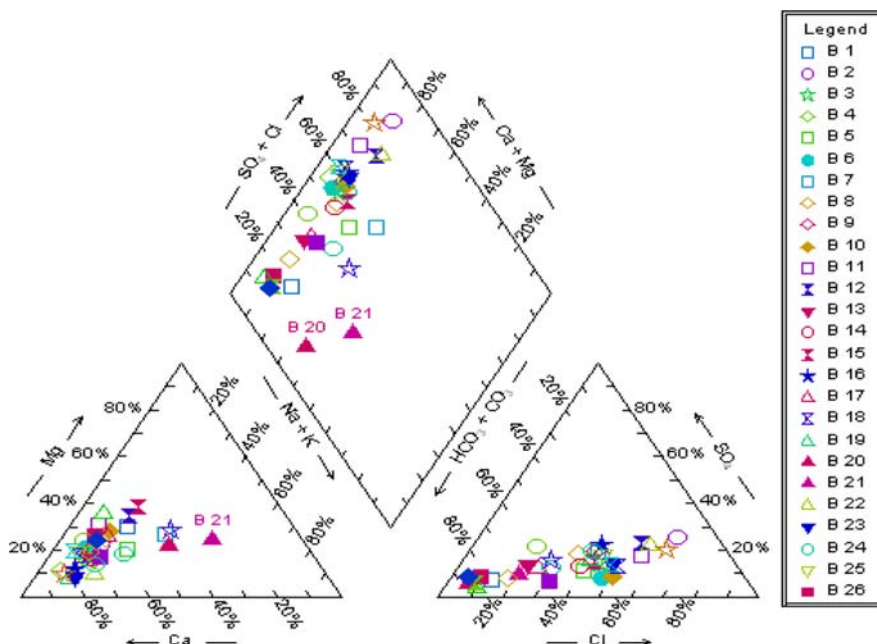
Correlation analysis

The correlation pattern for the pre-monsoon and post-monsoon periods are given in Tables 2 and 3, respectively; these give a clear picture of hydro-chemical processes occurring in the study area. There was an inverse correlation between arsenic and Eh (redox potential), i.e., the value of Eh increases with decreasing arsenic concentration, which mean concentration of arsenic increases with increasing redox status of any environment. Further, significant positive correlation between As and Fe, NH<sub>4</sub><sup>+</sup>, and PO<sub>4</sub><sup>3-</sup> were also noticed along with negative correlation between As and Mn, which substantiate the strong reducing character of this environment. There is a good correlation between Na<sup>+</sup> and K<sup>+</sup> which indicates that the source for both elements could be the same. There are good correlations between TDS and Ca<sup>2+</sup>, between TDS and Cl<sup>-</sup>, and between TDS and SO<sub>4</sub><sup>2-</sup>, which indicate that carbonate weathering

**Fig. 2** Piper diagram of groundwater in the pre-monsoon season with legend of 26 sampling locations



**Fig. 3** Piper diagram of groundwater in the post-monsoon season with legend of 26 sampling locations



with anthropogenic sources, mainly fertilizers, control the geochemistry of the groundwater in the pre-monsoon period. Further, good correlations between  $\text{Ca}^{2+}$  and  $\text{Cl}^-$  and between  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  are readily apparent, which indicates that both gypsum (absorbed in clay) and limestone are acting as a source of calcium. During the post-monsoon season, an additional correlation arises between TDS and  $\text{HCO}_3^-$ , which further supports the existence of carbonate weathering in the area. Other correlations of different strength, for example the correlation between  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  and between  $\text{Cl}^-$  and  $\text{NO}_3^-$ , may be attributed to secondary leaching.

#### Factor analysis

Factor analysis of hydrochemical properties of the groundwater samples identified five major components (factor) in each season (Tables 4 and 5). The number of significant factors within the dataset was determined by including only components with eigenvalues  $>1.0$ . The total variability explained by the identified five factors in the pre-monsoon and post-monsoon seasons were 80.38 and 81.77%, respectively. The degree of association between each variable and factor is shown by their respective loading for each component. The pre-monsoon (Table 4) and post monsoon (Table 5) results

substantiate the fact that there is an insignificant seasonal variation in the governing hydro-geochemical processes that control the groundwater quality in the area, as is evident from the loading of each variable on each factor and the communality of each variable. Although there is a slight difference in the loading of each variable in each factor in each season, the numbers of pairs of variables in each component remain same throughout the year with very few exceptions.

In general, factor 1 exhibits high loading, i.e., strong geochemical associations between  $\text{PO}_4^{3-}$ , Fe, As, and As(III) and inverse association with Depth, ORP and Mn in both seasons. Such loading for the most important factor i.e., component 1 is a strong indicator of a reducing environment which seems to be the main reason for arsenic enrichment in the groundwater. Factor 2 represents the association of EC and TDS with some major ions which indicates percolation of salts and halite deposits. Factor 3 is accounted for Na and K that indicate feldspar weathering. Thus, factors 2 and 3 account for geochemical processes which are less important than the existence of the reducing environment expressed by factor 1. Factor 4 is attributed to anthropogenic activities, as is evident from the high loading for Ph with  $\text{NO}_3^-$ , which arise from biological mineralization and agricultural activities. Further, component 4



**Table 2** Correlation matrix of chemical constituents of groundwater of Bhagalpur for the pre-monsoon season

	Ph	ORP	EC	TDS	Na <sup>+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	F <sup>-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	PO <sub>4</sub> <sup>3-</sup>	H <sub>4</sub> SiO <sub>4</sub>	NH <sub>4</sub> <sup>+</sup>	Fe	Mn	As (tot)	As (3+)	As (5+)														
ORP	-0.18																																		
EC	-0.26	0.27																																	
TDS	-0.23	0.28	0.76																																
Na <sup>+</sup>	-0.30	0.28	0.56	0.56																															
K <sup>+</sup>	-0.26	0.30	0.55	0.55	0.98																														
Ca <sup>2+</sup>	-0.19	0.06	0.78	0.78	0.36	0.31																													
Mg <sup>2+</sup>	0.08	-0.48	-0.13	-0.11	-0.11	-0.13	0.15																												
HCO <sub>3</sub> <sup>-</sup>	-0.05	0.30	0.48	0.48	0.42	0.45	0.10	-0.10																											
F <sup>-</sup>	0.09	0.34	0.16	0.16	0.46	0.59	-0.22	-0.11	0.40																										
Cl <sup>-</sup>	-0.33	0.16	0.70	0.70	0.50	0.48	0.78	0.04	-0.07	0.04																									
NO <sub>3</sub> <sup>-</sup>	-0.26	-0.04	0.13	0.12	0.07	0.01	0.42	0.17	-0.34	-0.28	0.37																								
SO <sub>4</sub> <sup>2-</sup>	-0.14	0.22	0.73	0.73	0.49	0.49	0.85	0.06	0.09	0.09	0.81	0.27																							
PO <sub>4</sub> <sup>3-</sup>	0.28	-0.80	-0.12	-0.12	-0.11	-0.10	-0.04	0.43	-0.23	-0.08	0.02	-0.07	-0.04																						
H <sub>4</sub> SiO <sub>4</sub>	-0.18	0.34	0.42	0.42	0.33	0.31	0.39	0.16	0.24	0.08	0.26	0.38	0.42	-0.28																					
NH <sub>4</sub> <sup>+</sup>	0.33	-0.78	-0.13	-0.12	-0.29	-0.31	0.02	0.26	-0.22	-0.31	-0.10	-0.12	-0.11	0.70	-0.29																				
Fe	0.21	-0.90	-0.27	-0.28	-0.30	-0.32	-0.09	0.30	-0.32	-0.26	-0.11	0.11	-0.21	0.81	-0.39	0.79																			
Mn	-0.20	0.91	0.25	0.26	0.28	0.30	0.07	-0.33	0.35	0.34	0.17	-0.09	0.22	-0.74	0.31	-0.72	-0.89																		
As (tot)	0.28	-0.86	-0.22	-0.22	-0.22	-0.25	-0.03	0.37	-0.28	-0.24	-0.05	0.02	-0.16	0.83	-0.41	0.82	0.93	-0.83																	
As (3+)	0.20	-0.86	-0.17	-0.18	-0.16	-0.19	-0.03	0.37	-0.28	-0.20	-0.04	0.01	-0.13	0.85	-0.34	0.80	0.93	-0.81	0.95																
As (5+)	0.34	-0.55	-0.23	-0.22	-0.27	-0.28	-0.02	0.24	-0.19	-0.22	-0.06	0.03	-0.15	0.50	-0.41	0.57	0.59	-0.57	0.75	0.49															

**Table 3** Correlation matrix of chemical constituents of groundwater of Bhagalpur for the post-monsoon season

	Ph	ORP	EC	TDS	Na <sup>+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	F <sup>-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	PO <sub>4</sub> <sup>3-</sup>	H <sub>4</sub> SiO <sub>4</sub>	NH <sub>4</sub> <sup>+</sup>	Fe	Mn	As (tot)	As (3+)	As (3+)	
ORP	-0.24																					
EC	-0.35	0.27																				
TDS	-0.33	0.28	0.87																			
Na <sup>+</sup>	-0.29	0.32	0.54	0.54																		
K <sup>+</sup>	-0.28	0.29	0.52	0.52	0.92																	
Ca <sup>2+</sup>	-0.23	0.03	0.70	0.71	0.27	0.30																
Mg <sup>2+</sup>	0.23	-0.55	-0.21	-0.20	-0.18	-0.16	-0.06															
HCO <sub>3</sub> <sup>-</sup>	-0.01	0.29	0.51	0.51	0.45	0.42	0.13	-0.13														
F <sup>-</sup>	0.02	0.38	0.15	0.16	0.60	0.60	-0.08	-0.14	0.34													
Cl <sup>-</sup>	-0.31	0.05	0.66	0.67	0.37	0.39	0.85	-0.06	-0.08	-0.02												
NO <sub>3</sub> <sup>-</sup>	-0.32	-0.11	0.49	0.50	0.12	0.13	0.65	0.13	-0.14	-0.12	0.61											
SO <sub>4</sub> <sup>2-</sup>	-0.26	0.14	0.62	0.62	0.41	0.42	0.77	-0.02	-0.05	0.04	0.78	0.54										
PO <sub>4</sub> <sup>3-</sup>	0.39	-0.80	-0.13	-0.12	-0.10	-0.07	0.07	0.52	-0.17	-0.14	0.10	0.06	-0.03									
H <sub>4</sub> SiO <sub>4</sub>	-0.17	0.64	0.41	0.41	0.37	0.36	0.41	-0.27	0.19	0.25	0.41	0.25	0.46	-0.39								
NH <sub>4</sub> <sup>+</sup>	0.37	-0.78	-0.12	-0.11	-0.31	-0.28	0.10	0.36	-0.16	-0.33	0.05	-0.02	-0.05	0.70	-0.55							
Fe	0.26	-0.92	-0.25	-0.25	-0.34	-0.31	0.02	0.44	-0.33	-0.30	0.06	0.14	-0.10	0.82	-0.63	0.79						
Mn	-0.25	0.91	0.21	0.22	0.30	0.27	-0.09	-0.42	0.34	0.38	-0.09	-0.18	0.06	-0.74	0.56	-0.72	-0.89					
As (tot)	0.32	-0.87	-0.22	-0.21	-0.26	-0.23	0.03	0.46	-0.29	-0.23	0.08	0.10	-0.11	0.82	-0.56	0.83	0.91	-0.83				
As (3+)	0.27	-0.84	-0.18	-0.18	-0.28	-0.25	0.06	0.48	-0.24	-0.26	0.07	0.15	-0.15	0.84	-0.55	0.75	0.92	-0.83	0.92			
As (5+)	0.30	-0.62	-0.20	-0.20	-0.14	-0.12	-0.02	0.28	-0.27	-0.11	0.07	0.00	-0.02	0.53	-0.40	0.68	0.64	-0.57	0.79	0.45		

**Table 4** Multivariate factor analysis score for the pre-monsoon period

Variables	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5
Depth	−0.74	0.13	0.23	0.01	−0.07
Ph	0.24	−0.16	−0.21	0.65	0.08
ORP	−0.92	0.14	0.12	0.05	−0.13
EC	−0.15	0.91	0.24	0.11	0.00
TDS	−0.15	0.91	0.24	0.13	0.00
Na <sup>+</sup>	−0.15	0.43	0.81	−0.16	0.01
K <sup>+</sup>	−0.16	0.40	0.87	−0.10	−0.01
Ca <sup>2+</sup>	0.01	0.94	−0.11	−0.14	0.16
Mg <sup>2+</sup>	0.39	−0.03	−0.03	−0.01	0.76
HCO <sub>3</sub> <sup>−</sup>	−0.27	0.23	0.46	0.59	0.07
F <sup>−</sup>	−0.20	−0.09	0.77	0.26	0.00
Cl <sup>−</sup>	0.00	0.82	0.16	−0.37	−0.04
NO <sub>3</sub> <sup>−</sup>	0.01	0.26	−0.21	−0.68	0.37
SO <sub>4</sub> <sup>2−</sup>	−0.09	0.86	0.13	−0.12	0.13
PO <sub>4</sub> <sup>3−</sup>	0.89	−0.02	0.10	0.06	0.10
H <sub>4</sub> SiO <sub>4</sub>	−0.38	0.36	0.10	−0.07	0.69
NH <sub>4</sub> <sup>+</sup>	0.84	0.04	−0.22	0.21	−0.04
Fe	0.95	−0.12	−0.12	−0.08	−0.01
Mn	−0.89	0.13	0.15	0.09	−0.06
As (tot)	0.97	−0.05	−0.08	0.02	−0.03
As (3+)	0.95	−0.05	0.01	−0.04	0.02
As (5+)	0.66	−0.03	−0.23	0.14	−0.13
Eigen value	7.41	4.66	2.70	1.62	1.30
Percentage of variance	33.67	21.18	12.26	7.37	5.92
Cumulative percentage	33.67	54.84	67.10	74.47	80.38

shows a relationship of EC and TDS with HCO<sub>3</sub>, which indicates weathering of carbonaceous material. The only property that shows significant seasonal variation is H<sub>4</sub>SiO<sub>4</sub>, which accounts for significant loading in factor 5 in the pre-monsoon period but switched to factor 2 in the post-monsoon period with similar loading. This indicates that with rainfall and elevated groundwater levels silicate weathering becomes dominant in the area (Kumar et al. 2009a). Thus, factor analysis indicates the multiple sources and processes controlling the overall groundwater quality in the middle Gangetic floodplain.

#### Arsenic and its speciation

The distribution pattern of arsenic in the pre-monsoon and post-monsoon seasons is shown in Figs. 4 and 5, respectively. It was found that arsenic contamination is more prevalent in the vicinity of the floodplain of the river Ganges and its tributary

the Koshi (Ghugri) river. Most of these regions are located in the northern and northwestern part of Bhagalpur district, viz. Sabour (94 µg/L), Sultanganj (87 µg/L), and Ranuchak (118 µg/L) (Figs. 4, 5). In general, 47% of sampling location exceed the 50 µg/L permissible limit which shows the magnitude of arsenic contamination in the area. However, in many areas of the Lower Meghna Estuary, Bangladesh, more than 80% of wells (Ravenscroft et al. 2005) and 93% of wells in Hajiganj Upazila of southeast Bangladesh exceed the permissible limit (Jakariya et al. 1998). The arsenic concentration is, therefore, still not too high in comparison with reported values for West Bengal and Bangladesh, which indicates nascent stage of arsenic enrichment. These observations suggest that the high concentrations of arsenic in the lower catchment of the Ganges River can be attributed to the existence of multiple sources and the likelihood of related mechanisms of mobilization/enrichment across the entire Central Gangetic

**Table 5** Multivariate factor analysis score for the post-monsoon period

Variables	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5
Depth	-0.74	-0.01	0.32	0.01	-0.07
Ph	0.30	-0.27	-0.08	0.03	0.85
ORP	-0.93	0.08	0.17	0.08	0.09
EC	-0.15	0.72	0.20	0.59	-0.13
TDS	-0.15	0.73	0.20	0.58	-0.11
Na <sup>+</sup>	-0.17	0.29	0.88	0.22	-0.14
K <sup>+</sup>	-0.14	0.30	0.88	0.19	-0.13
Ca <sup>2+</sup>	0.06	0.92	-0.02	0.18	0.02
Mg <sup>2+</sup>	0.52	-0.03	0.00	-0.18	0.13
HCO <sub>3</sub> <sup>-</sup>	-0.21	-0.04	0.30	0.87	0.10
F <sup>-</sup>	-0.21	-0.10	0.81	0.06	0.16
Cl <sup>-</sup>	0.08	0.92	0.14	-0.04	-0.08
NO <sub>3</sub> <sup>-</sup>	0.11	0.75	-0.08	-0.10	-0.28
SO <sub>4</sub> <sup>2-</sup>	-0.07	0.88	0.18	-0.10	0.03
PO <sub>4</sub> <sup>3-</sup>	0.88	0.04	0.07	-0.02	0.18
H <sub>4</sub> SiO <sub>4</sub>	-0.62	0.52	0.20	-0.07	0.29
NH <sub>4</sub> <sup>+</sup>	0.84	0.02	-0.21	0.12	0.18
Fe	0.95	-0.02	-0.17	-0.10	-0.05
Mn	-0.89	-0.05	0.21	0.10	0.07
As (tot)	0.96	-0.01	-0.05	-0.10	0.06
As (3+)	0.92	-0.01	-0.13	-0.01	-0.03
As (5+)	0.71	0.00	0.07	-0.21	0.19
Eigen value	7.80	4.61	2.74	1.70	1.14
Percentage of variance	35.46	20.95	12.45	7.74	5.18
Cumulative percentage	35.46	56.41	68.86	76.60	81.78

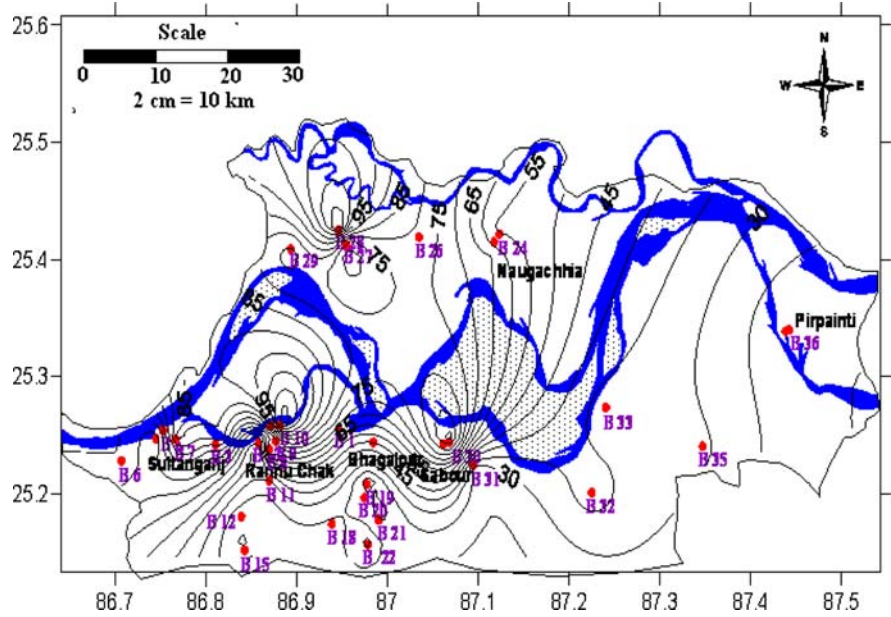
Plain. It has been reported by Ravenscroft et al. (2005) that average arsenic contents of riverbed samples are 2.03 mg/kg in the Ganges, 2.79 mg/kg in the Brahmaputra, and 3.49 mg/kg in the Meghna. Therefore, the high As concentration in the groundwater is mainly because of infiltration of river water through contaminated river bed sediment. This observation must be further supported by the regional groundwater flow direction based on topography or hydrographs. Although groundwater helps to sustain base flow of the river in some places, perennial Himalayan rivers, for example the Ganges and the Yamuna are likely to feed groundwater (Kumar et al. 2009a).

Less than 50 µg/L of arsenic concentration in the central portion of the study area is possibly because of the accumulation of coarser sediment along Holocene course of the river Ganges (Ravenscroft et al. 2005). This indicates how depositional

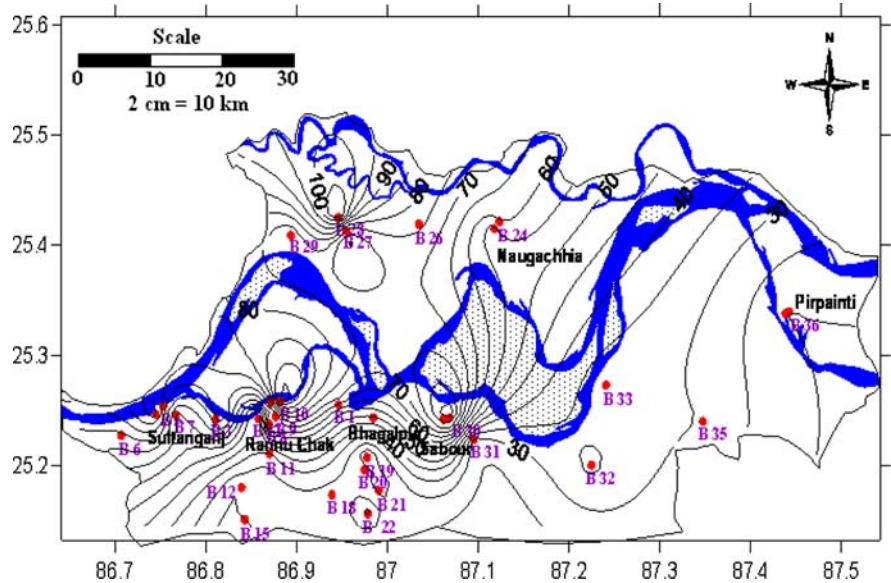
environment and geological age are important factors in controlling arsenic mobilization. After speciation analysis it has been observed that As(III) varies from 11 to 81 µg/L in the pre-monsoon season and from 12 to 76 µg/L in the post-monsoon season. The average value of As(III) was found to be equal to 66% of the average value of the total As concentration; this is the most important issue in the area, because As(III) is prime source of lethality. Although the percentage of wells exceeding the permissible limit (50 µg/L) is less than in Bangladesh and West Bengal, the percentage contribution of As(III) to the total arsenic concentration is quite high. It has also been found that As(III) has the same correlation as total As.

The depth distribution of arsenic reveals a strong correlation between occurrence of arsenic and the depth of hand pumps or tube wells (Fig. 6). In general, the highest arsenic concentration and the spatial distribution and significant temporal variation

**Fig. 4** Depiction of the spatial distribution of arsenic concentration ( $\mu\text{g/L}$ ) in the pre-monsoon season by contouring



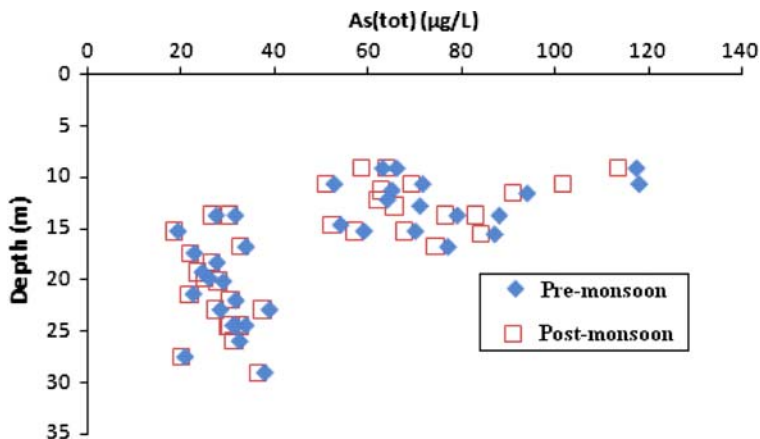
**Fig. 5** Contour map of arsenic concentration ( $\mu\text{g/L}$ ) in the post-monsoon season



were mainly found in the case of shallow aquifer viz. 10–20 m below ground level. The arsenic concentration decreases rapidly below about 40 m. Similar observations are reported by Karim et al. 1997. This condition requires strong redox conditions to drive arsenic mobility along the well depth (McArthur et al. 2001). Moreover, between 10 and 20 m well depths, the high concentration of arsenic is associated with high concentrations of  $\text{Fe}$ ,  $\text{PO}_4^{3-}$ , and  $\text{NH}_4^+$  which

favours development of anoxic conditions during microbial oxidation of sedimentary organic matter along with some anthropogenic output, i.e., the source might be both natural and artificial. Low  $\text{NO}_3^-$  concentration reveals that the  $\text{NO}_3^-$  is the thermodynamically favourable oxygen donor for microbial degradation of dissolved organic materials in the shallow aquifers (HP). It can thus be concluded that “oxyhydroxide reduction theory” is responsible

**Fig. 6** Scatter diagram for depth (meter) and total As conc. ( $\mu\text{g/L}$ ) showing the vertical profile of arsenic contamination



for release of arsenic in the aquifer of Bhagalpur. It has been reported by Acharyya and Shah (2004) that although the concentration of dissolved Fe in groundwater is generally low ( $<1\text{ mg/L}$ ) in the Ganga Alluvial Plain, biogeochemical conditions are generally unfavourable for triggering release of As to groundwater. Therefore, in Bhagalpur, where the Fe content of groundwater reaches 7.62 and 6.84 mg/L in the pre and post-monsoon periods, respectively, conditions suitable for mobilization of As to groundwater may be a local effect.

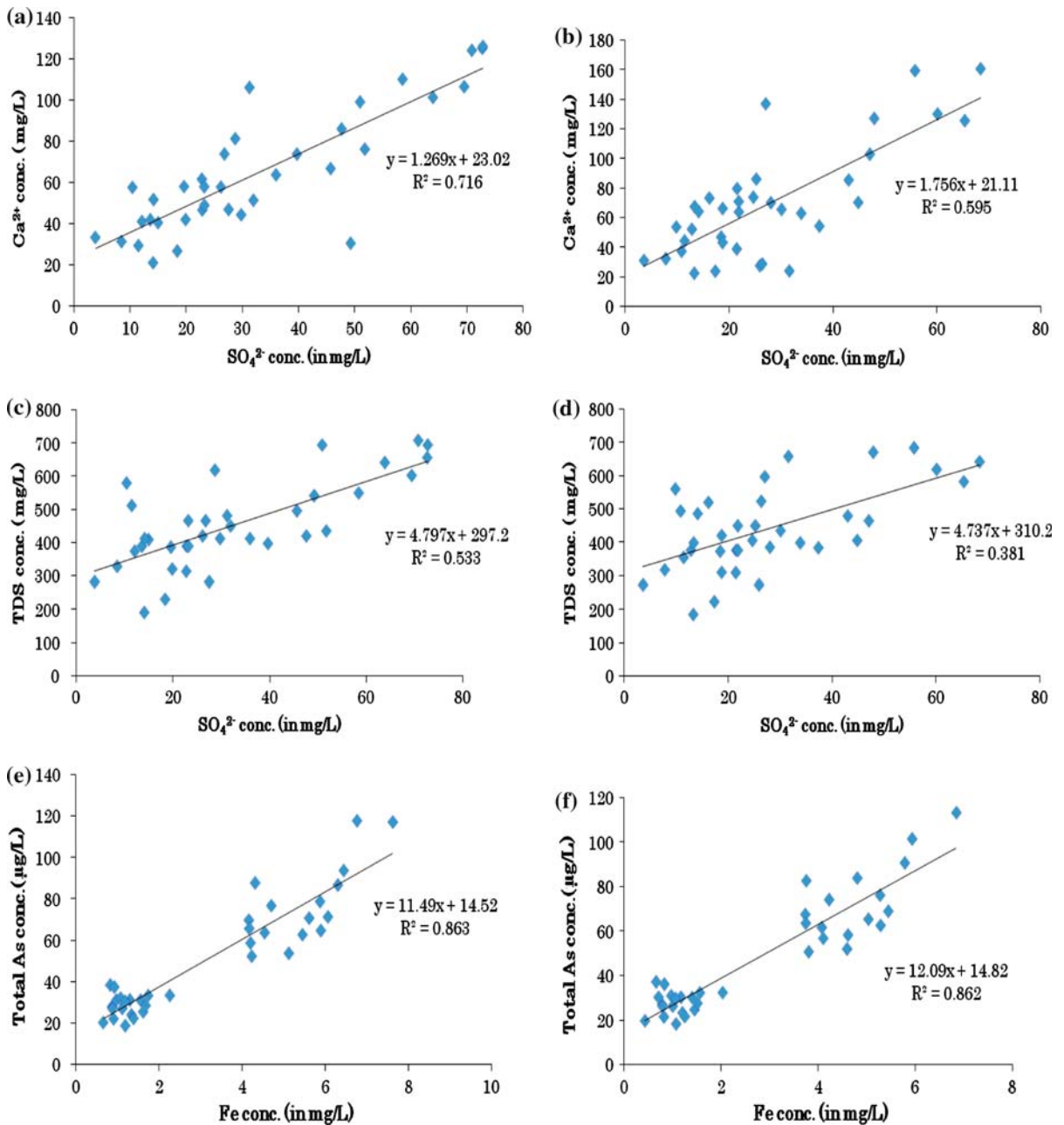
From different scatter diagrams relationships between Ca and  $\text{SO}_4^{2-}$ , between TDS and  $\text{SO}_4^{2-}$ , and between As and Fe were observed (Figs. 7a, c, e, respectively, for pre-monsoon and Figs. 7b, d, f for post-monsoon). A good correlation ( $>0.5$ ) between  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  (Figs. 7a, b) and TDS (Figs. 7c, d) also indicates that the acid produced by oxidation of pyrites is being neutralized by carbonate formed during the weathering process, and arsenic is consequently released by sorbed hydrated Fe-oxide. High correlation between arsenic and phosphate shows the mobilization of arsenic may be partially governed by anthropogenic agricultural activities. This assumption is supported by high As–Fe correlation. The Fe–As relationship under redox conditions is also proved by the paleo-channels and shifting behavior of river Ganga. The shifting behavior may lead to variation in hydrological budget, which further triggers the conversion of oxidized arsenic to the reduced form (a more mobile and toxic species).

The Bhagalpur district experiences frequent flooding by the south-west monsoon and has a channelized

natural water system, so the groundwater interacts with the surface water, which accelerates weathering and other hydrogeochemical processes and consequently triggers the sediment load and accumulation of arsenic in these aquifers. Therefore, study must take geochemical investigation of surface sediments and core sediments into account to assess mineralogical control over the arsenic mobility at different depths.

#### Aquifer sediment dispositions

The sequence of aquifer sediment was determined by grain-size analysis of core samples obtained at different depths. Data for grain-size analysis of disturbed core samples is shown in Table 6 and a scatter diagram showing relationship between core sediment properties with depth is shown in (Fig. 8). These results reveal that a thick layer of clay extends between the depths 4.57 and 7.62 m (i.e., 15–25 feet) from surface. Such a layer is likely to contain biotite and other dark coloured ferromagnesian and opaque minerals in relatively high percentages. This clay layer has been exposed to drained and oxidized conditions that subsequently act as an impervious barrier in the aquifer system. This is also because of differences in the composition of alluvium, as evident from grain-size analysis. Further, depth of clay occurrence as obtained by grain-size analysis can be easily correlated with the depth profile of As contamination (Fig. 6). Thus, grain-size analysis confirms the “oxyhydroxide reduction theory” postulated earlier.



**Fig. 7** Different scatter diagrams showing the relationship between calcium and sulphate in the pre-monsoon (a) and post-monsoon (b) periods, between TDS and sulphate in the pre-

monsoon (c) and post-monsoon (d) periods, and between arsenic and iron in the pre-monsoon (e) and post-monsoon (f) seasons

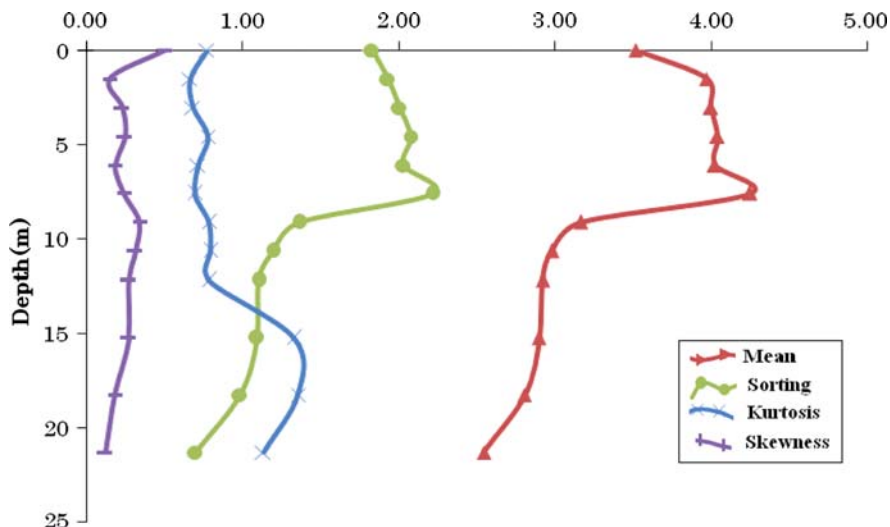
**Conclusions and recommendations**

The groundwater quality of the study area has a primary problem of arsenic followed by nitrate contamination which needs immediate attention. The cation and anion ratios reflect the seasonal

variability in the weathering pattern of carbonate and silicate minerals, which is consistent with the aquifer depths. However, there is insignificant seasonal variation in the governing hydro-geochemical processes that control the groundwater quality in the area. The groundwater in the area is being affected

**Table 6** Grain-size analysis of disturbed core samples

Grain size ( $\mu\text{m}$ )	Depth in meters											
	0 (Top soil)	1.5	3.0	4.5	6.0	7.5	9.0	10.5	12.0	15.0	18.0	21.0
	Weight in grams											
250	38.83	33.21	35.98	32.34	30.86	25.35	34.30	40.18	38.63	40.10	34.41	34.93
125	12.65	10.20	12.70	14.60	11.34	13.40	18.72	24.83	35.16	34.54	48.61	53.82
63	10.02	8.90	6.72	8.23	10.65	11.00	14.95	13.40	7.41	7.06	4.16	2.13
50	7.00	7.34	8.74	11.62	8.43	8.87	10.68	6.03	6.87	5.23	3.82	1.79
30	7.02	7.65	5.22	5.90	7.08	7.70	9.35	6.25	4.94	4.99	2.55	1.22
20	6.24	10.26	8.64	4.24	9.34	6.01	4.70	3.40	2.84	3.28	2.05	1.03
10	8.43	9.53	7.34	6.68	9.50	7.63	3.08	2.45	1.46	1.10	1.26	1.02
5	4.47	7.71	9.56	8.26	6.25	9.22	1.87	1.47	1.01	1.03	1.01	1.01
2	3.75	3.28	4.22	6.15	6.20	8.54	1.43	1.12	1.00	1.01	1.00	1.00
Recovered weight	98.42	98.07	99.13	98.02	99.67	97.72	99.07	99.14	99.31	98.34	98.88	97.95
Initial weight	100	100	100	100	100	100	100	100	100	100	100	100

**Fig. 8** Scatter diagram showing relationship between core sediment properties and depth

both by complex hydrogeochemical processes and by anthropogenic activity, mainly intensive agricultural practices. The heterogeneous distributions of arsenic (elemental signature) in aquifers were found to be concentrated in the vicinity of the river flood plain. This behavior of arsenic is governed by redox reactions at shallow depth which support the dissolution of arsenic from arseniferous iron oxyhydroxides. Sediment grain-size analysis has indicated the presence of clay at shallow depth which coincided with the highest As contamination. This higher As contamination at a particular depth because of clay seems to be well known by a local driller. However,

just avoiding that particular depth for tapping groundwater for drinking purposes is not likely to solve the problem because of possible further mobilization of As. Therefore, quantified regulation of groundwater withdrawal based on aquifer properties, for example transmissivity (T), storativity (S), and hydraulic conductivity (K), must be put in place for sustainable drinking water management. Further geochemical investigations using X-ray diffraction and geochemical software, for example PhreeqC, to assess mineralogical control over arsenic mobility at different depths of groundwater in the central Gangetic Plain can be the subject of a future study.



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