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Arsenic occurrence in groundwater of Kathmandu Valley, Nepal

S.K. Chapagain[a]*, S. Shrestha[b], T. Nakamura[a], V.P. Pandey[a], F. Kazama[a]

[a]*Department of Ecosocial System Engineering, University of Yamanashi, 4-3-11 Takeda, Kofu, Yamanashi, 400-8511, Japan*
Tel. +81 55 220 8193; email: saroj@yamanashi.ac.jp; g05dea01@yamanashi.ac.jp; g07dea03@yamanashi.ac.jp, kfutaba@yamnashi.ac.jp
[b]*Institute for Global Environmental Strategies, 2108-11 Kamiyamaguchi, Hayama, Kanagawa 240-0115, Japan*
email: shrestha@iges.or.jp

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ABSTRACT

This study was carried out to assess arsenic occurrence in groundwater of Kathmandu Valley, Nepal, with focus on spatial and seasonal variation, total and dissolved arsenic concentration and the arsenic release process. Several deep and shallow groundwater samples from northern and central groundwater districts were collected during the pre-monsoon and monsoon seasons, and analyzed for major physicochemical parameters. The arsenic concentration was $<10\ \mu$g/L (i.e. WHO standard) in shallow groundwater, whereas in deep groundwater, 52% of samples exceeded the WHO standard. The arsenic varies spatially with higher concentration towards the central groundwater district whereas seasonal variation is insignificant. Arsenic is mainly released into the groundwater from the natural source under the reductive process. The central groundwater has a higher reducing condition than the northern groundwater districts. The concentrations of total and dissolved arsenic are not significantly different.

Keywords: Arsenic; Groundwater; Kathmandu Valley

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1. Introduction

Arsenic contamination in groundwater is a serious problem around the world and the problem is more severe in countries like Bangladesh, India, Vietnam, Argentina, Cambodia, China, Thailand, parts of the Americas [1,2]. In Nepal, study on arsenic was started in 1999 from the southern plain region of the country called the Terai belt. It has revealed the contamination of groundwater with arsenic in many parts of the country [3]. Groundwater is an important water resource in Kathmandu Valley, Nepal. It shares 50% of the total water supply in the valley [4]. In recent years, there has been a marked expansion in groundwater demand due to population and industrial growth. The huge dependency on groundwater and adverse health effects (such as dermal changes, cardiovascular, reproductive, mutagenic and carcinogenic; [5]) from the arsenic exposure have drawn high attention to study of arsenic contamination in groundwater of the valley.

Though some earlier studies highlighted arsenic contamination in groundwater of the valley and release of arsenic into groundwater, its interaction with groundwater parameters is not yet clearly known. Arsenic concentration in such earlier studies is expressed in the form of total arsenic only, and occurrence of particulate arsenic is so far unknown. Since information about particulate arsenic has significant to undertake the effective arsenic removal treatment from drinking water [6], attempts are made to study both total and particulate

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*Corresponding author.

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arsenic and their spatial variation in groundwater of the Kathmandu Valley. Furthermore, seasonal variation in arsenic is also studied to address the concern of possible dilution effects of monsoon rainfall on arsenic concentration (as reported by Maharajan et. al. [7]), considering the seasonal variation in rainfall pattern (more than 80% of rainfall during July–September) in the study area.

2. Materials and methods

2.1. Study area

The Kathmandu Valley is situated in the central part of Nepal (Fig. 1) with an average altitude of 1300 m above sea level (masl). The watershed boundary of the valley covers almost 664 km^2[4], which is drained by the Bagmati River to the south of the watershed. The groundwater system of Kathmandu Valley can be seen as a closed groundwater basin, with several more or less connected and interrelated aquifers. The basin has two major aquifers (i.e. shallow and deep aquifers) and which are separated by the aquitard; and the thickness of the aquitard decreases towards the margin of the basin. Based on the hydrogeology, the deep part of the groundwater basin has been classified into three districts, namely northern, central and southern groundwater districts [8]. The groundwater characteristics are described based on groundwater districts in this study. The northern groundwater district (NGWD) is characterized as a potential recharge zone in the valley. The deposits of the district are composed of unconsolidated, highly permeable materials consisting of micaceous quartz, sand and gravel, about 60 m thick, interbedded with several impermeable fine layers. The central groundwater district (CGWD) is characterized as thick layer of black clay deposit, named as Kalimati formation. The Kalimati formation is overlaid by fluvial originated fine to medium sand, silt intercalated clay and fine gravels. The dominance of black clay layer has appeared as barrier in groundwater recharge in the central groundwater. The southern groundwater district (SGWD) has also the thick clay formation and low permeable basal gravel and parts of the zone (eastern area of the SGWD) are covered with sand and gravel deposits which are potential for groundwater recharge [8,9].

2.2. Sampling and analysis

Forty-two deep groundwater and thirteen shallow groundwater wells were sampled during the pre-monsoon (April) and monsoon (September) seasons in 2007. The sampling was carried out mainly from major groundwater extracting areas in the valley (Fig. 1). Samples were collected after removing the stagnate water, and stabilizing the groundwater parameters (i.e. pH, ORP, EC). The sampling bottles (polyethylene) were rinsed three times before being filled, and the collected samples were kept immediately in the ice bag, which were further filtered (0.45 μm), labelled and stored. Sampling days was usually sunny; with mean air temperature of 28.4ºC and 26.7ºC during the pre-monsoon and monsoon seasons respectively.

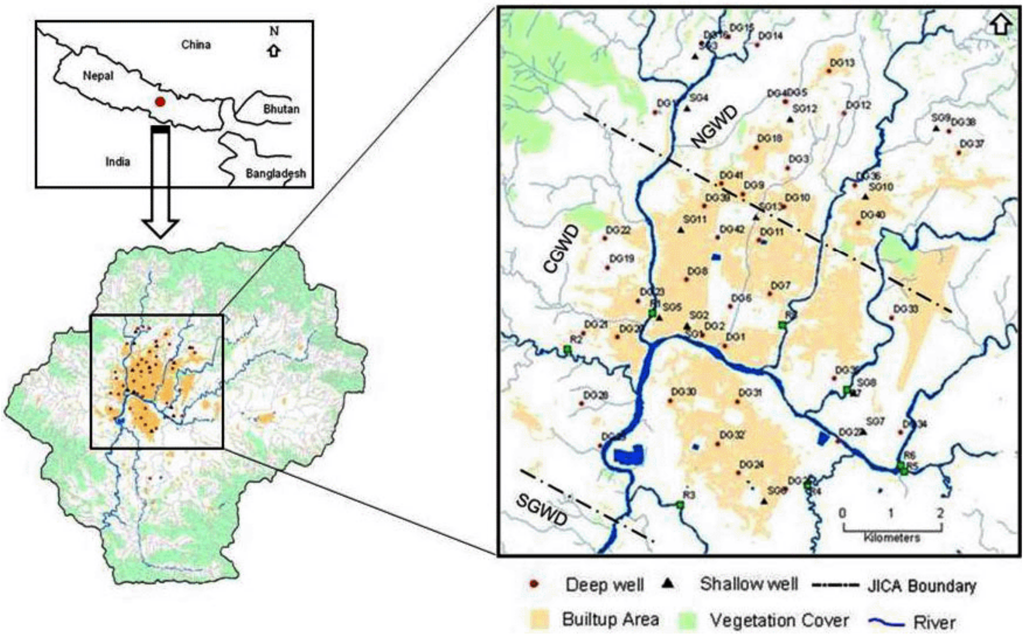

Fig. 1. Location map of study area (Kathmandu Valley) and sampling stations.

Electrical conductivity (EC), salinity, pH, and water temperature were measured on-site using a portable tester (Hanna Combo Tester, HI 98129). The ORP was recorded on-site using ORP tester (Oakton Waterproof ORPTestr BNC 10). Those testers were used to calibrate once in a sampling day using the respective standards The major cations (Na^+,K^+,Ca^{2+},Mg^{2+}, NH_4^+-N) and anions (SO_4^{2-}, Cl^-, F^-, PO_4^{3-}-P, NO_3^--N) were determined by ion chromatography (761 Compact IC, Metrohm). The dissolved organic carbon (DOC) and inorganic carbon (IC) were determined by total carbon analyzer (TOC-VCSU, Shimadzu). Bicarbonate (HCO_3^-) was indirectly calculated based on pH, IC and temperature of groundwater sample. Heavy metal (cupper, cadmium, zinc, nickel, chromium, lead, iron, manganese) and silica were analyzed by inductively coupled plasma spectrometer (ICP). Arsenic was determined by using atomic absorption spectrometry with hydride generation (AAS-HG). Arsenic was measured from unfiltered and 0.45 μm filtered samples, which are termed as total and dissolved arsenic respectively. Stable isotope (^2H and ^{18}O) values of groundwater were determined by an isotope ratio mass spectrometer (IRMS) (Micromass, Prism Mass, UK) after equilibration with H_2 gas for ^2H and CO_2 gas for ^{18}O. The filtered sample (0.45 μm) was used for the laboratory analysis except total arsenic, and the statistical analysis was performed using Microsoft Excel and SPSS.

Table 1
Summary of physicochemical parameters of groundwater

(a) Deep groundwater (n=42)

Parameter	Unit	Mean	Median	Min.	Max.	Std. dev.
Temperature	°C	25.1	25.4	19.7	28.0	1.8
pH		6.6	6.6	6.1	7.3	0.2
ORP	mV	−82	−86	−135	30	30
EC	μS/cm	586	495	111	1722	379
DOC	mg/L	5.5	4.4	0.1	16.4	4.4
IC	mg/L	77.0	62.0	12.4	219.3	53.2
NH_4^+-N	mg/L	23.3	14.4	0.0	119.8	27.3
Na^+	mg/L	36.2	27.9	2.7	93.9	23.3
K^+	mg/L	2.5	2.1	0.2	6.4	1.8
Mg^{2+}	mg/L	8.4	6.1	2.2	31.3	6.6
Ca^{2+}	mg/L	33.7	28.7	7.2	77.3	21.7
Cl^-	mg/L	2.3	0.9	0.2	32.0	5.2
PO_4^{3-}-P	mg/L	2.5	1.9	0.0	8.7	2.6
SO_4^{2-}	mg/L	1.2	0.1	0.0	31.0	4.9
HCO_3^-	mg/L	314	242	36	941	217
NO_3-N	mg/L	0.0	0.0	0.0	0.1	0.0
F^-	mg/L	0.3	0.2	0.0	2.3	0.4
Fe	mg/L	2.9	2.0	0.0	10.7	3.0
Mn	mg/L	0.4	0.3	0.0	1.7	0.3
Si	mg/L	28.5	30.7	4.6	40.1	7.6

(b) Shallow groundwater (n=13)

Parameter	Unit	Mean	Median	Min.	Max.	Std. dev.
Temperature	°C	21.1	20.7	19.5	24.8	1.5
pH		6.3	6.3	6.0	6.8	0.2
ORP	mV	9	−42	−73	130	80
EC	μS/cm	621	659	269	1169	263
DOC	mg/L	2.1	1.5	0.5	4.6	1.3
IC	mg/L	50.1	54.7	20.4	78.1	19.6
NH_4^+-N	mg/L	5.3	4.7	0.0	12.3	4.4
Na^+	mg/L	35.8	39.4	7.7	79.0	19.2
K^+	mg/L	13.3	10.2	1.4	49.7	12.9
Mg^{2+}	mg/L	12.6	12.5	2.8	22.0	6.9
Ca^{2+}	mg/L	38.8	39.8	7.5	71.4	19.4
Cl^-	mg/L	47.1	48.0	11.8	82.7	24.3
PO_4^{3-}-P	mg/L	0.1	0.001	0.001	0.4	0.2
SO_4^{2-}	mg/L	22.0	12.2	0.0	60.0	20.3
HCO_3^-	mg/L	177	174	68	302	84
NO_3-N	mg/L	1.4	0.01	0.001	5.3	2.1
F^-	mg/L	0.2	0.1	0.1	0.4	0.1
Fe	mg/L	4.8	3.8	0.1	17.1	5.8
Mn	mg/L	0.5	0.5	0.0	1.4	0.4
Si	mg/L	14.5	15.5	6.6	26.6	5.4

(Note: Min: minimum, Max: maximum, Std. dev.: standard deviation).

3. Results and discussion

3.1. Physicochemical parameter

The summary of physicochemical parameters of groundwater samples is presented in Table 1. The results of pre-monsoon samples are discussed in the following text. The pH of groundwater was nearly neutral, ranging from 6.1–7.3. EC in deep groundwater varied from 111 to 1722 μS/cm with a mean value of 586 μS/cm. It was a relatively higher value (mean 621 μS/cm) in shallow groundwater, with a range of 269–1169 μS/cm and the higher level is more likely due to local contamination. The major anions were dominated by HCO_3^- followed by PO_4^{3-}-P and Cl^- and lower concentrations of SO_4^{2-} and NO_3^--N in deep groundwater, but the dominance of HCO_3^- was followed by Cl^- and SO_4^{2-} and NO_3^--N and lower concentrations of PO_4^{3-}-P in shallow groundwater. Deep groundwater was enriched in DOC (mean 5.5 mg/L), whereas the value was lower (mean 2.1 mg/L) in shallow groundwater. The higher organic matter content in the sediment may have increased the DOC level in deep groundwater. All deep groundwater (except one well) showed a negative ORP value, which apparently reveals the reducing condition of the groundwater. It was also demonstrated by lower concentrations of NO_3^--N (mean

0.02 mg/L), SO_4^{2-} (mean 1.2 mg/L) and a higher concentration (mean 23.3 mg/L) of NH_4^+-N. The reducing condition is possibly due to microbial degradation of organic matter. Moreover, higher concentrations of NO_3^--N (mean 1.4 mg/L) and SO_4^{2-} (mean 22.0 mg/L) indicate a relatively lower reducing condition in shallow groundwater.

3.2. *Arsenic occurrence in groundwater*

Arsenic concentration varied significantly between shallow and deep groundwater. The mean arsenic concentrations were 1 µg/L and *17 µg/L* in shallow and deep groundwater respectively. In deep groundwater, nearly 52% of samples exceeded arsenic concentration of 10 µg/L i.e., WHO standard. The mobility of arsenic depends on redox levels of groundwater whereas a reducing condition (i.e. lower redox level) is considered favourable to arsenic mobility. Similarly, arsenic concentration increases with higher phosphate and bicarbonate due to the competition of these ions with arsenic for the sorption sites. Higher mobility of arsenic is also reported in the presence of dissolved organic anions; which is usually derived from DOC [10]. Therefore, the lower concentration of arsenic in shallow groundwater is possibly related to the higher redox level and lower concentrations of competitive ions. On the contrary, reducing groundwater condition and high levels of PO_4^{3-}-P, HCO_3^- and DOC are possibly liable for higher arsenic concentration in deep groundwater of the study area.

This study revealed a wide range of arsenic (<1–73 µg/L) occurrence in deep groundwater. However, an earlier study reported the concentration up to 265 µg/L [11]. This discrepancy is likely due to the difference in sampling locations, and it has further suggested the wider spatial variation. This study reveals that higher concentrations of arsenic in the central groundwater district (Fig. 2). In addition to arsenic, higher concentrations of other parameters (like DOC, PO_4^{3-}-P, NH_4^+-N, Fe, Mn) were also observed in the central groundwater district (Table 2).

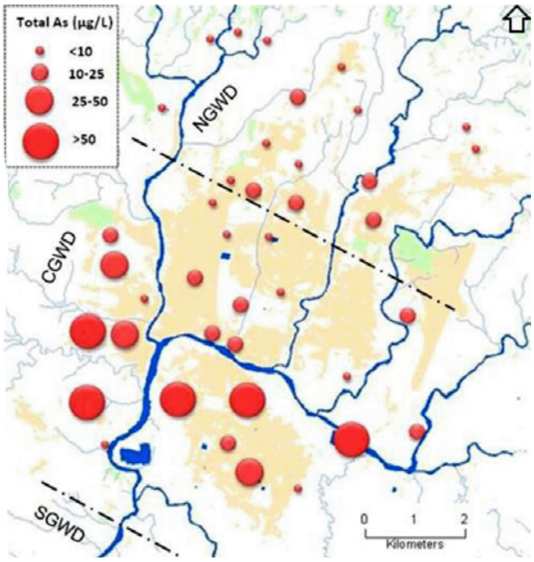

Fig. 2. Spatial distribution of arsenic in groundwater.

Table 2
Arsenic and major water quality parameters based on the groundwater district

(a) Northern groundwater district (NGWD)

Parameter	Unit	Mean	Median	Min.	Max.	Std. dev.
As (total)	µg/L	7.7	6.8	<1	18.0	4.7
Temperature	°C	24.8	25.1	19.7	27.2	1.7
pH		6.6	6.6	6.1	7.1	0.2
ORP	mV	−68	−69	−135	30	35
EC	µS/cm	309	279	111	663	156
DOC	mg/L	2.4	2.1	0.1	9.5	2.3
IC	mg/L	38.9	35.1	12.4	75.9	17.2
NH_4^+-N	mg/L	6.4	2.7	0.0	29.8	8.0
PO_4^{3-}-P	mg/L	1.4	0.6	0.0	8.7	2.2
SO_4^{2-}	mg/L	2.9	0.1	0.0	31.0	8.1
HCO_3^-	mg/L	159	143	36	339	70
Fe	mg/L	2.5	1.9	0.0	9.0	2.4
Mn	mg/L	0.3	0.2	0.0	0.5	0.1
Si	mg/L	29.0	30.8	4.6	40.1	7.9

(b) Central groundwater district (CGWD)

Parameter	Unit	Mean	Median	Min.	Max.	Std. dev.
As (total)	µg/L	24.8	13.6	2.2	73.1	23.4
Temperature	°C	25.5	25.7	21.2	28.0	1.6
pH		6.6	6.6	6.4	7.3	0.2
ORP	mV	−93	−98	−123	−35	22
EC	µS/cm	800	763	283	1722	356
DOC	mg/L	7.8	7.1	0.8	16.4	4.2
IC	mg/L	106.4	99.7	41.7	219.3	52.1
NH_4^+-N	mg/L	36.3	28.5	2.2	119.8	29.6
PO_4^{3-}-P	mg/L	4.0	3.8	0.4	8.1	2.3
SO_4^{2-}	mg/L	0.2	0.1	0.0	2.3	0.6
HCO_3^-	mg/L	434	411	177	941	213
Fe	mg/L	2.8	2.2	0.0	10.7	3.0
Mn	mg/L	0.5	0.4	0.1	1.7	0.4
Si	mg/L	28.5	31.1	12.7	38.0	7.5

Note: Min: minimum, Max: maximum, Std. dev.: standard deviation.

Groundwater quality is influenced by the geological formation of the aquifers, recharge source and anthropogenic activities [12], and physicochemical properties of sediments greatly influence arsenic concentration [13]. The groundwater districts vary in the sediment characteristics; a progressive increase in the finer particles and trace elements is reported towards the central part of the sediments from the northern part in the valley [14]. Therefore, the major variation in groundwater quality between central and northern groundwater district (Table 2) is more likely due to geochemical heterogeneity of the sediment.

Table 2 reveals that the central groundwater is under a stronger reducing condition than the northern groundwater district. It was revealed by lower values of ORP, SO_4^{2-} and a higher concentration of NH_4^+-N in the central groundwater district. The condition is more likely due to microbial degradation of organic matter; because the sediment of the central part in the study area was reported as rich in organic matter content [15]. The enrichment of organic matter is also revealed by the higher DOC value of the central groundwater district (Table 2b). Unlike the northern groundwater, the sulfate content in the central groundwater district is much lower; which is possibly due to the transformation of sulfate into sulfide under the reduced groundwater condition. Moreover, the central groundwater district reveals high mineralization wherein EC ranges 283–1722 μS/cm.

The relationship of the arsenic and groundwater parameter was examined by calculating the Spearman's correlation coefficients (Table 3). Arsenic was significantly (at 5% *p*-level) positively correlated with PO_4^{3-}-P, HCO_3^-, and DOC, which was in close agreement with the earlier finding [10,16]. It suggests that competition between anions (namely, PO_4^{3-}, HCO_3^-, anion released from DOC) and arsenic for the sorption sites have influenced the arsenic mobility in the groundwater. On the contrary, a negative correlation (significant at 5% *p*-level) between arsenic and ORP suggests increased arsenic activity with the reducing groundwater condition. Therefore, the higher concentration of arsenic in the central deep groundwater could be attributed to the following: geologically enriched clay sediment, elevated level of DOC, PO_4^{3-}-P, HCO_3^- and reducing groundwater environment.

The relationship of arsenic with selected groundwater parameters is plotted in Fig. 3. In general, arsenic concentration increases with increasing concentration of DOC, PO_4^{3-}-P, NH_4^+-N, Mn; but the relationships are not distinct at higher levels. Anawar et al. [17] suggests that a reductive process is responsible for arsenic concentration, especially when the concentration of nitrate and sulfate are lower; and high concentrations of dissolved iron and ammonium ions. Following this, it is likely that reductive process is more dominant to release arsenic into the groundwater of the valley. The degradation of organic matter might have increased the reducing condition of groundwater and typically, Fe/Mn oxy-hydroxides could be the source of arsenic and lead to desorption of arsenic, iron and manganese under the reducing condition (see [12]). Arsenic showed positive relationship with Mn, however lack of such relationship with iron (Table 3). This could be due to some of the samples with very high iron content had low to moderate arsenic content (see [18]).

3.3. Seasonal arsenic occurrence

To examine possible effect of monsoon rainfall on arsenic, we analyzed the seasonal (pre-monsoon and monsoon) concentrations of arsenic and stable isotope (^{2}H and ^{18}O) in groundwater. The concentration of arsenic in groundwater between the seasons is shown in Fig. 4, which reveals a close similarity of arsenic concentration between pre-monsoon and monsoon seasons. Further-

Table 3
Spearman's correlation coefficients (n=42)

	As	PH	ORP	DOC	NH_4^+-N	NO_3^--N	PO_4^{3-}-P	SO_4^{2-}	HCO_3^-	Fe	Mn	EC	Na^+	K^+	Si
As	1														
PH	0.24	1													
ORP	-0.36*	-0.13	1												
DOC	0.42*	-0.35*	-0.27	1											
NH_4^+-N	0.48*	-0.30	-0.31*	0.95*	1										
NO_3^--N	-0.10	-0.15	0.17	0.30	0.37*	1									
PO_4^{3-}P	0.39*	-0.35*	-0.21	0.74*	0.68*	0.02	1								
SO_4^{2-}	-0.29	-0.03	0.16	-0.02	0.04	0.54*	-0.14	1							
HCO_3^-	0.54*	-0.11	-0.44*	0.90*	0.92*	0.28	0.64*	0.03	1						
Fe	-0.27	-0.54*	-0.09	0.70	0.09	0.11	-0.07	0.01	-0.04	1					
Mn	0.19	-0.32*	0.07	0.39*	0.39*	0.21	0.23	-0.03	0.31*	0.59	1				
EC	0.46*	-0.14	-0.42	0.86*	0.92*	0.41*	0.57*	0.14	0.95*	0.02	0.33*	1			
Na^+	0.59*	-0.14	-0.30	0.73*	0.63*	-0.03	0.68*	-0.15	0.73*	-0.17	0.26	0.65*	1		
K^+	0.46*	-0.21	-0.35*	0.88*	0.90*	0.40*	0.56*	0.10	0.92*	0.03	0.40*	0.91*	0.67*	1	
Si	0.05	-0.44*	0.16	0.36*	0.23	-0.11	0.29	-0.17	0.14	0.46*	0.67*	0.12	0.32*	0.16	1

*Significant at 5% *p*-level.

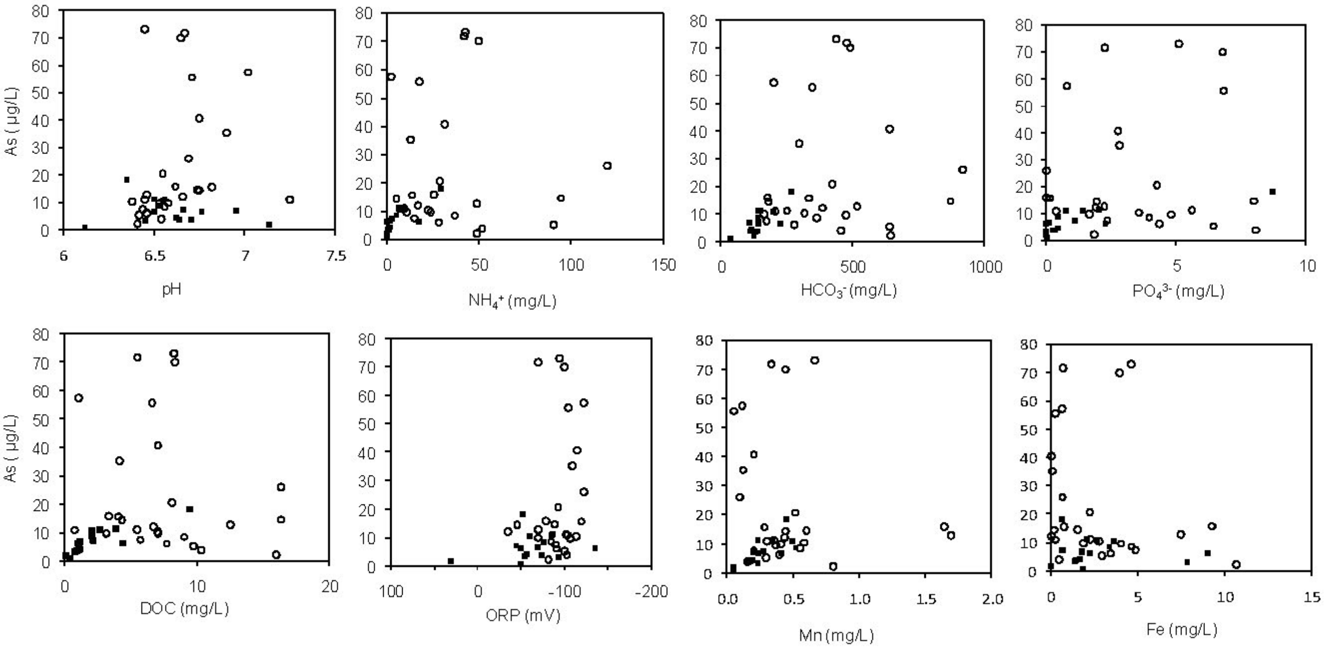

Fig. 3. Scatter plots of arsenic with selected groundwater papramenters. ○, CGWD; ■, NGWD.

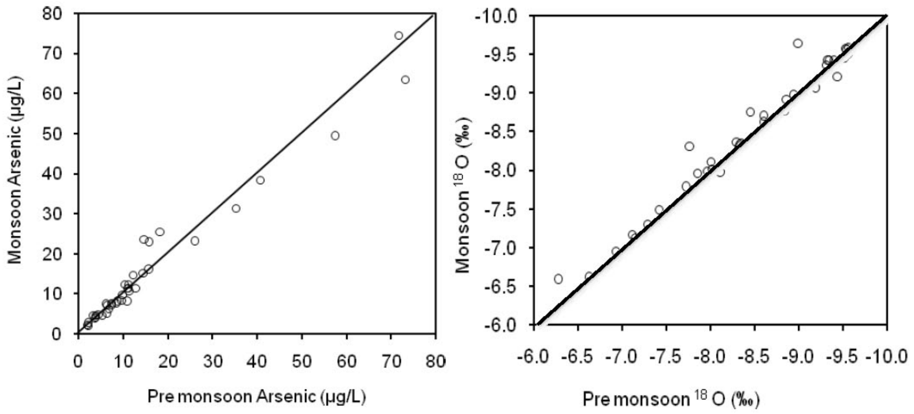

Fig. 4. Scatter plots of (a) arsenic concentration and (b) $\delta^{18}O$ concentration in deep groundwater during the pre-monsoon and monsoon season. (Line in the figure represents a 1:1 relationship).

more, there is no significant (at 5% *p*-level) difference in the mean arsenic concentration. However, the reports on the seasonal variation of arsenic concentration in other parts of the world are inconsistent. Some studies have found the significant variation in arsenic concentration among the seasons but not in some other studies [19,20]. Similar to arsenic, seasonal variation of stable oxygen isotope ($\delta^{18}O$) composition in groundwater was insignificant.

The central groundwater district is considered as poorly recharging due to the presence of a thick black clay layer whereas the northern groundwater district is regarded as the major recharge area in the valley [8]. Moreover, this study showed insignificant changes of isotopic composition in groundwater between the seasons. It suggests the immediate effect of the monsoon rainfall in arsenic concentration is less apparent. Though we found no clear change in arsenic concentration between the seasons, it is likely to change over time due to changes in the redox condition, pumping rate, or other factors. Hence, a follow-up study would be important for addressing the trend more precisely.

3.4. Total and dissolved arsenic concentration

Total and dissolved arsenic concentrations were measured based on unfiltered and 0.45 μm filtered samples, whereas difference was considered as particulate arsenic concentration. The ratio between arsenic concentrations of unfiltered to 0.45 μm filtered samples was close to 1, which indicates a lack of significant difference in concentration between these forms. A similar result has been reported from groundwater in southeast Michigan, USA, by Kim et al. [21]. On the contrary, a study carried out in Inner Mongolia showed a significant difference between dissolved and total arsenic concentration where 35% of the total arsenic retained as the particulate arsenic, which did not pass through 0.45 μm filters [6]. However, the distribution of particulate and dissolved arsenic is said to vary on geographic location (rock type) and chemical properties of groundwater [21].

4. Conclusions

Arsenic concentration varies significantly between the shallow and deep groundwater in the valley. Higher concentration is mainly observed in deep groundwater. Fifty-two percent of deep groundwater samples exceeded the WHO standard for arsenic in drinking water (i.e. 10 μg/L). The arsenic also varies in spatial scale, with a relatively higher concentration in the central groundwater district. Moreover, unusual lower arsenic and sulfate contents in parts of the central groundwater district may indicate the possibility of arsenic-sulfide substances formation, which, however, needs a detailed study. The difference between total and dissolved arsenic concentration is observed insignificant. Regarding arsenic release into the groundwater, it is mainly from the natural source under the reductive process. The central groundwater has higher reducing condition than the northern groundwater districts. Finally, the insignificant seasonal variation in arsenic and stable isotope values reveals dilution effect of monsoon rainfall on arsenic concentration is less apparent.

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