



Spatial Distribution of Arsenic in Ground Water from Nuggihalli Mining Area and its Surroundings, Southern Karnataka, India

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ABSTRACT

Arsenic (As) is one of the most toxic elements in the nature. The main aim of this study investigates the occurrence and distribution of arsenic in groundwater from Nuggihalli mining area and its surroundings, Southern of Karnataka, India. The ground water samples from the study area are analyzed for arsenic and other quality parameters (pH, TDS, Ca, Mg, Na, K, Cl, HCO₃, NO₃, SO₄, B, Fe, Mn). The elevated levels of arsenic concentrations are found in the range from 0.002 to 0.51 mg/L, which demonstrated that almost 79% of examined water samples exhibit arsenic concentration higher than the maximum concentration limit of WHO specifications (0.01mg/L) for drinking. Pierson's correlation analysis and principal component analysis were used to find out possible relationships among the examined parameters and ground water samples. Arsenic is highly correlated with manganese and iron suggesting common geogenic origin of these elements. No significant correlation observed between As and Ca or SO₄, suggesting that the contamination source of As is different in the wells. Spatial distribution was also explained to identify the areas with higher arsenic concentration in groundwater.

Keywords: Arsenic, Groundwater, Occurrence and distribution, Pierson's correlation, Principal Component Analysis

INTRODUCTION

Arsenic (As) is an element of great concern as a “known” human carcinogen based on occupational and drinking-water exposures. Widespread occurrence of geogenic arsenic in groundwater in the worldwide [1] is concern in the developing countries (Bangladesh, India and China) and industrialized countries (USA). The natural occurrence of arsenic in the aquatic environment is usually associated with sedimentary rocks of marine origin, weathered volcanic rocks, geothermal areas, and fossil fuels. Most of the arsenic derived from anthropogenic sources is released as a by-product of mining, metal refining processes, the burning of fossil fuels, and agricultural use [2,3]. Almost 25% of groundwater from alluvial aquifers contains high concentration of arsenic, the consumption of which threatens the health of millions of people [4-6]. Recent reports suspected that more than 100 million people worldwide ingest excessive amount of arsenic through drinking water contaminated from natural geogenic sources [7]. Geochemical weathering of subsurface soil by natural process has caused an unacceptable level of dissolved arsenic in groundwater in many regions of the Indian subcontinent [8].

Increased concentrations of arsenic have been determined in many areas all over the world such as in South East Asia [9-12], in Central, South and North America [13-15] and in Australia [16]. In addition, elevated arsenic concentrations have also found in various European countries [17]. Many areas in Greece are reported elevated arsenic concentrations in groundwater probably due to natural enrichment from geothermal activities and/or pollution [18, 19]. In order to minimize the possible risk from arsenic a parametric value of 0.01mg As/L has been set for water intended for human consumption according to Directive 98/83/EC [20]. Further, USEPA also decided to move forward in implementing the same standard for drinking water [21], which is also the guideline value recommended by the World Health Organization [22]. On the other hand, higher levels of arsenic tend to be found in ground water than in surface water bodies, and typically groundwater is the source for drinking water for the people in rural areas.

In the present study, the distribution of arsenic in groundwater from Nuggihalli mining area and its surroundings located in Hasan district of Karnataka State, India. Nuggihalli and its surrounding areas are exclusively dependent on groundwater from bore wells for their agricultural and drinking purposes. Thus, the investigation of quality of groundwater in this area has vital importance. This research is an integrated approach on the occurrence and distribution of arsenic in groundwater and their correlation with other water quality parameters in order to delineate the sources of contamination.

The major objectives of this study includes (i) establishment of occurrence and spatial distribution of the arsenic to identify the sources of contamination and (ii) to find out possible relationships among the arsenic and other physicochemical parameters through correlation and principal component analysis.

Description of the study area

Nuggihalli mining areas are situated in Hasan district of Karnataka state in India. The study area located between the latitudes of 12° 55'00" to 13° 05' 00" N and longitudes of 76°25' 00" to 76°35'00" E (Fig. 1) and covers an area of 400sq.km. The area has many abandoned chromite mining areas, a few active mines with little extraction of chromium ore. Besides, the area is under rapid developments with greater extent of agricultural activity and habitants. It is well known that agricultural activities required large quantities of water resources, wherein the study area is completely dependent on groundwater from bore wells for agriculture. Naturally it is

expected the overexploitation of groundwater led to lower the water table and quality is also deteriorated.

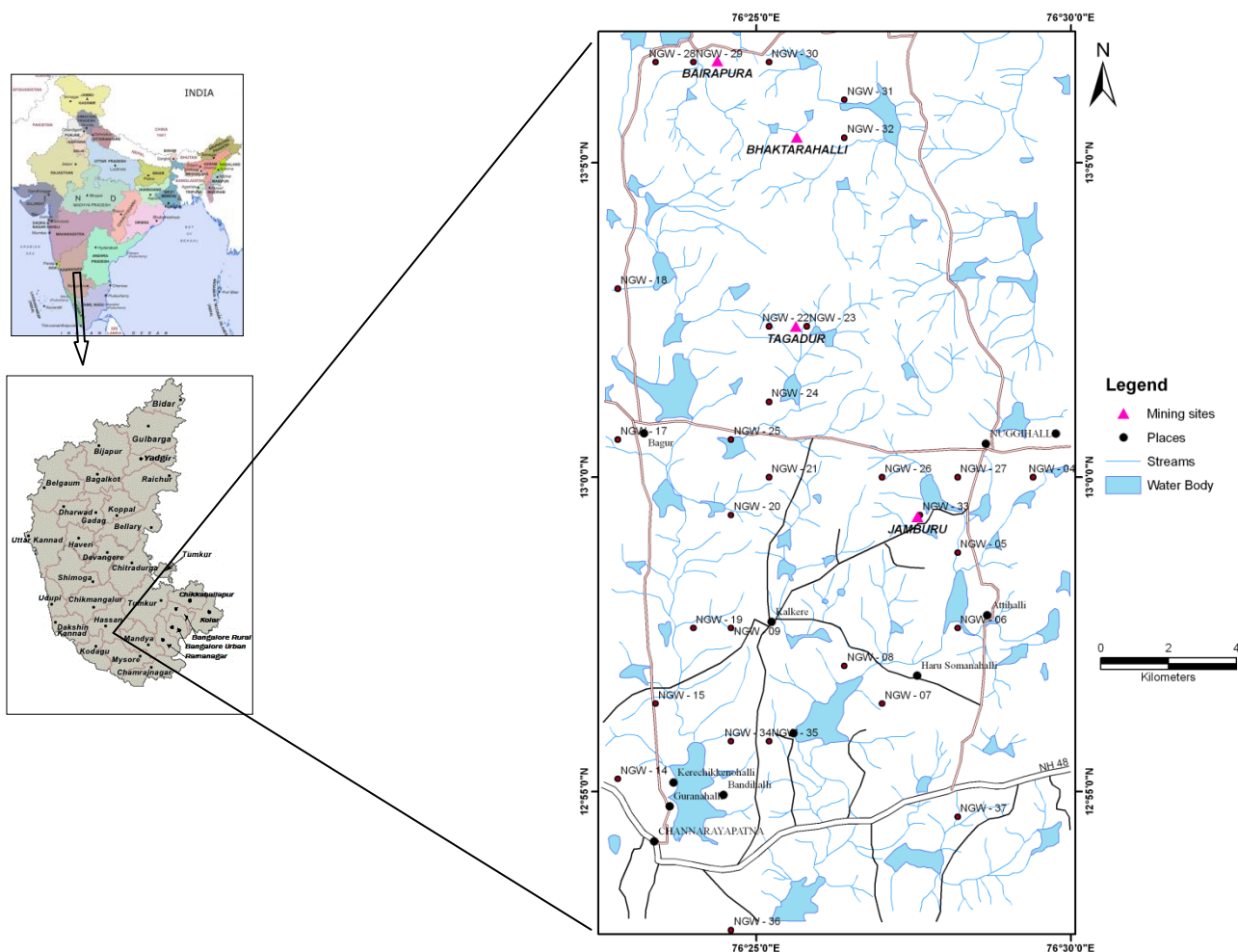


Fig. 1 Study area map with sample location

The main aquifer formed in the study is due to weathered and fractured granites, granitic gneiss and chromite bearing ultramafic rocks. No studies were reported on the occurrence and distribution of arsenic, its correlation with other physicochemical parameters; the quality of groundwater in Nuggihalli and its surrounding areas which has vital importance.

MATERIALS AND METHODS

Chemicals and materials

All analytical standards for ICP-OES (multi element standard), Ion-chromatograph standards (Na, K, Mg, Ca, Cl, NO₃ and SO₄) both are obtained from Merck, Germany. Suprapur nitric acid and sulfuric acid (Merck, Germany) were used for eluent preparation for use in Ion Chromatographic separation. Other chemicals were analytical grade only. Milli-Q water (Millipore) with resistivity of 18MΩ is used for entire samples preparation and analysis.

Sampling of groundwater

Groundwater samples were randomly collected from 33 bore wells in Nuggihalli mining and its surrounding areas (Fig.1) in order to distribute the sampling locations in the study area. Throughout the survey a global positioning system (GPS) was used to locate the sampling locations. Polypropylene bottles (100 ml), which were soaked in a nitric acid bath and then washed with Milli-Q water, were used for water sampling. The samples were collected in duplicate at each sampling location. The basic parameters like pH and Total Dissolved Solids (TDS) were measured in the field using hand held pH and TDS meters. The collected water samples were stored at 4°C before analysis.

Water analysis

Water samples were analyzed at Environmental Geochemistry Laboratories in CSIR-National Geophysical Research Institute, Hyderabad, India. Immediately after receiving the samples in the laboratory, from one set of the water samples two aliquots were acidified with HCl for As and HNO₃ for heavy metal analysis. These aliquots are used for quantitative determination of arsenic using continuous flow hydride generation [23] whereas other metals analyzed using EPA200.7 method by inductively coupled plasma optical emission spectrophotometer (ICP-OES, Optima 4300 DV). However, the other set of water samples (not acidified) were filtered through 0.2µm cellulose nitrate membrane filters (Whatman, Germany), and were analyzed for Ca, Mg, Na, K, Cl, SO₄, NO₃ using EPA 300.1 method by Ion Chromatography (882 compact IC, Metrohm). Titrimetric method was employed for analysis of bicarbonates in the water samples.

Quality control samples include replicates and field blanks. Replicate samples were collected immediately after the routine samples in the field using the same collection method and equipment. All difference measured in concentrations between replicate pairs were well within the precision of the method for all measured parameters. Analysis of blank samples did not show any inherent bias in the method of analysis for the trace elements and major ions. The accuracy of methods has been checked by standard solutions (NIST, USA). All statistical analyses were performed with SPSS, Version7.0 software. Pearson's correlation coefficient was used to measure the strength of the associations between arsenic concentrations and other water quality parameters.

RESULTS AND DISCUSSION

Water quality parameters

The quality characteristics (physicochemical parameters) of groundwater in the area of Nuggihalli and its surroundings in southern Karnataka, India, are shown in Table 1. Summary of statistical data of each parameter is presented in Table 2. The ranges are influenced due to some extreme values measured in Bairapura and Chikkondanahalli areas. The pH values ranged from slightly neutral to slightly alkaline (6.8–8.2), with a mean value of 7.2. Total dissolved solids (TDS) were relatively high, ranging from 220 to 1940mg/L, with mean value 883mg/L. The higher values of Total dissolved solids (TDS), were observed at the areas of Jamburu, Bairapura and Bhaktarahalli suggesting influence mining activity. Bicarbonate was the dominant anion in groundwater, followed by chloride, sulphate or nitrate. Potassium and sodium were the dominant cations in ground waters; Magnesium exhibited significant ion in some wells. Calcium and bicarbonate were also recorded as dominant ions in the majority of groundwater samples in the study area.

Table 1 Concentrations of arsenic and other water quality parameters of the study area

Well NO	pH	TDS (mg/L)	As (mg/L)	Na (mg/L)	K (mg/L)	Mg (mg/L)	Ca (mg/L)	Cl (mg/L)	NO ₃ (mg/L)	SO ₄ (mg/L)	HCO ₃ (mg/L)	Fe (mg/L)	Mn (mg/L)	B (mg/L)
NGW-4	7.4	530	0.16	72.97	6.40	21.35	9.07	18.88	18.20	14.96	398.25	0.07	0.07	0.02
NGW-5	7.7	880	0.11	52.82	3.24	101.26	6.12	135.35	23.14	39.02	389.82	0.02	0.11	0.05
NGW-6	7	1120	0.14	90.59	79.31	24.16	21.09	240.24	42.26	53.64	309.56	0.03	0.22	0.11
NGW-7	7.3	1240	0.24	114.98	9.08	62.21	36.98	278.61	98.77	138.51	444.73	0.03	0.37	0.07
NGW-8	7.1	1940	0.00	268.90	127.36	54.04	15.80	498.38	119.22	219.06	581.36	0.02	0.08	0.10
NGW-9	7.4	670	0.16	91.01	7.45	30.36	9.06	84.78	3.52	38.53	468.76	0.04	0.17	0.06
NGW- 14	7.2	700	0.05	72.07	8.74	25.89	20.92	110.10	50.35	56.17	336.52	0.05	0.09	0.07
NGW-15	7.3	900	0.17	139.74	5.23	38.52	0.05	146.35	61.39	41.17	496.90	0.07	0.12	0.06
NGW- 16	7.2	980	0.15	123.24	5.17	47.39	4.86	123.83	39.87	61.42	571.82	0.05	0.12	0.08
NGW- 17	7.1	1250	0.18	140.97	33.71	30.47	32.63	268.42	59.51	89.62	460.32	0.12	0.07	0.12
NGW- 18	7.3	570	0.13	42.24	4.26	18.73	37.93	90.73	2.18	3.72	381.62	0.02	0.18	0.04
NGW- 19	7.2	740	0.04	79.56	8.87	37.75	8.76	90.62	22.41	29.56	452.83	0.01	0.09	0.06
NGW- 20	7	1120	0.05	107.48	4.65	56.92	25.94	212.16	2.66	55.33	622.00	0.01	0.10	0.06
NGW- 21	7.2	680	0.06	100.24	4.61	24.87	8.38	68.14	20.39	27.70	298.06	0.02	0.12	0.04
NGW- 22	7.2	830	0.15	104.63	2.02	47.09	0.03	121.93	21.12	35.88	306.72	0.04	0.14	0.05
NGW- 23	7.1	630	0.05	45.56	4.56	18.34	57.53	85.95	31.61	52.26	196.21	0.02	0.09	0.04
NGW- 24	7.2	330	0.44	39.37	1.25	10.79	37.61	14.98	10.67	10.61	255.64	0.19	0.44	0.05
NGW- 25	7.2	520	0.37	47.51	4.62	15.24	63.20	87.56	23.36	45.17	235.92	0.13	0.09	0.06
NGW- 26	8.1	1000	0.06	34.77	4.43	138.12	12.96	155.75	93.17	14.35	557.98	0.10	0.04	0.05
NGW- 27	7.1	880	0.27	62.58	1.68	49.59	19.04	72.92	14.76	40.88	316.54	0.12	0.31	0.07
NGW- 28	7	1610	0.44	155.84	3.47	64.10	73.00	407.24	65.99	191.90	226.72	0.17	0.34	0.23
NGW- 29	7.2	980	0.18	83.24	1.44	62.22	10.82	172.33	18.63	79.22	282.31	0.06	0.17	0.08
NGW- 30	7.2	500	0.35	81.39	2.39	18.27	34.05	35.06	13.56	15.67	347.35	0.12	0.11	0.06
NGW- 31	7.3	550	0.27	67.36	9.13	18.91	14.79	33.28	2.12	21.80	268.95	0.08	0.39	0.08
NGW- 32	6.8	1800	0.07	227.12	2.76	25.52	99.66	511.75	172.69	158.83	155.43	0.07	0.07	0.12
NGW- 33	7	1160	0.43	118.24	150.35	29.28	0.06	192.99	28.82	75.41	423.08	0.09	0.46	0.16
NGW- 34	7.5	820	0.04	90.27	10.02	48.44	0.03	96.72	18.12	48.06	346.87	0.03	0.03	0.07
NGW- 35	7.2	1630	0.43	161.43	301.96	30.34	55.28	321.73	66.35	75.45	670.18	0.16	0.50	0.22
NGW- 36	7.6	220	0.51	18.43	3.20	7.14	27.47	12.89	3.95	5.65	162.86	0.16	0.62	0.05
NGW- 37	6.9	510	0.05	49.62	4.96	9.27	54.85	61.75	34.93	52.30	181.50	0.03	0.05	0.06

The concentration of nitrate ranged from 2.1 to 172.7 mg NO₃/L was detected. The higher concentrations were observed in Jamburu and Bhaktarahalli area. Extremely high concentration of nitrate (172.7 mg NO₃/L) was found in Bhaktarahalli area, suggesting direct runoff from surrounding agricultural activities, On the other hand, this well also exhibited high values of total dissolved solids, chloride, calcium, sodium, sulphate and boron probably due to this location is active mining area leading leaching of various ions.

Table 2 Summary statistics of arsenic and other water quality parameters

Parameter	Units	Min	Max	Median	Mean	S.D
pH		6.800	8.100	7.200	7.236	0.246
TDS	(mg/L)	220.000	1940.000	830.000	883.333	22.926
As	(mg/L)	0.002	0.510	0.154	0.182	0.146
Ca	(mg/L)	0.029	99.658	19.641	26.085	24.152
Na	(mg/L)	18.429	268.901	83.240	95.416	56.104
K	(mg/L)	1.245	301.963	4.845	25.116	60.584
Mg	(mg/L)	7.139	138.123	29.280	37.090	27.294
HCO ₃	(mg/L)	155.425	670.177	346.871	370.464	37.191
Cl	(mg/L)	12.887	511.754	110.104	152.463	32.315
NO ₃	(mg/L)	2.119	172.690	23.355	37.713	37.896
SO ₄	(mg/L)	3.719	219.061	41.173	55.928	51.830
Fe	(mg/L)	0.008	0.186	0.047	0.069	0.051
Mn	(mg/L)	0.032	0.622	0.112	0.180	0.157
B	(mg/L)	0.016	0.229	0.062	0.076	0.048

The concentration of sulphate ranged from 3.7 to 219.1 mg/L the higher concentrations were observed in Jamburu mining area. Sulphate in the study area is found to be in the permissible concentration limit of 45 mg/L as per WHO drinking water standards. It varies between 3.72 and 219.06 mg/L with an average of 55.93 mg/L. The high concentration of sulphate in drinking water is toxic and cause to laxative effective. The concentrations of iron (0.069 mg/L), manganese (0.18 mg/L) and boron (0.076 mg/L) were observed below permissible limit.

Arsenic concentrations

The variation of arsenic concentrations in groundwater is an important consideration for monitoring, regulation, and treatment of arsenic as well as for environmental and health studies in regional scale. The present study demonstrated that almost 79% of the examined wells

exhibited arsenic concentrations higher than the WHO permissible limit of 0.010 mg/L for drinking (Table 1). The concentration of arsenic ranged from 0.002 to 0.51 mg/L (Table 2). The spatial distribution of arsenic in groundwater of Nuggihalli and its surrounding areas, Southern of Karnataka is illustrated in Fig.2. The highest concentrations of arsenic were found in wells numbers 35 and 36 (0.43 and 0.51 mg/L, respectively). As well as these wells also having high concentrations of potassium, sodium, total dissolved solids, bicarbonate, chloride, iron and boron. On the contrary, the minimum concentrations of nitrates were determined in these wells. Pearson's correlation coefficients have been calculated to examine possible relationships among the measured parameters (Table 3). Arsenic concentrations are also plotted against selected physicochemical parameters and metals (Fig.3). Arsenic exhibited good significant correlation ($R^2=0.84$) with iron and manganese indicating common geogenic origin of these elements. It is also proved with the reported literature [24], that the correlation between arsenic and iron or manganese is often observed in groundwater because, under certain conditions, the presence of Fe/Mn oxyhydroxides could lead to desorption of arsenic, iron and manganese. Nevertheless, it is observed that the wells with high arsenic content exhibited also high boron concentrations suggesting common origin of these elements, which is also reported [25]. These results further suggesting that these relations of common geogenic origin and condition may enhance the

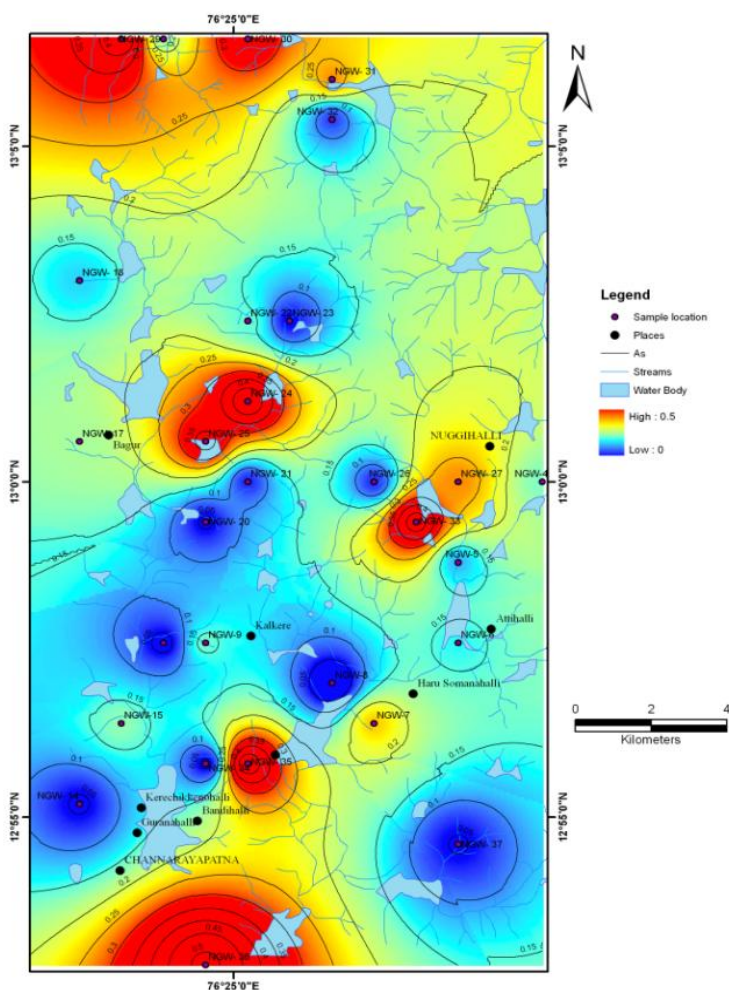


Fig.2. Spatial distribution of Arsenic in the study area

mobility of arsenic in the groundwater. Arsenic concentration was negatively correlated with Mg, Na, HCO_3 and NO_3 demonstrates, might be related to geological and geochemical properties in the Nuggihalli mining and its surrounding area. No significant correlation between As and Ca or SO_4 were found, suggesting that the contamination source of As and Ca or SO_4 was different in the wells. The present investigations on pH showed no correlation with arsenic concentrations in groundwater as the pH range is at neutral (~ 7.2). However, other investigators reported a

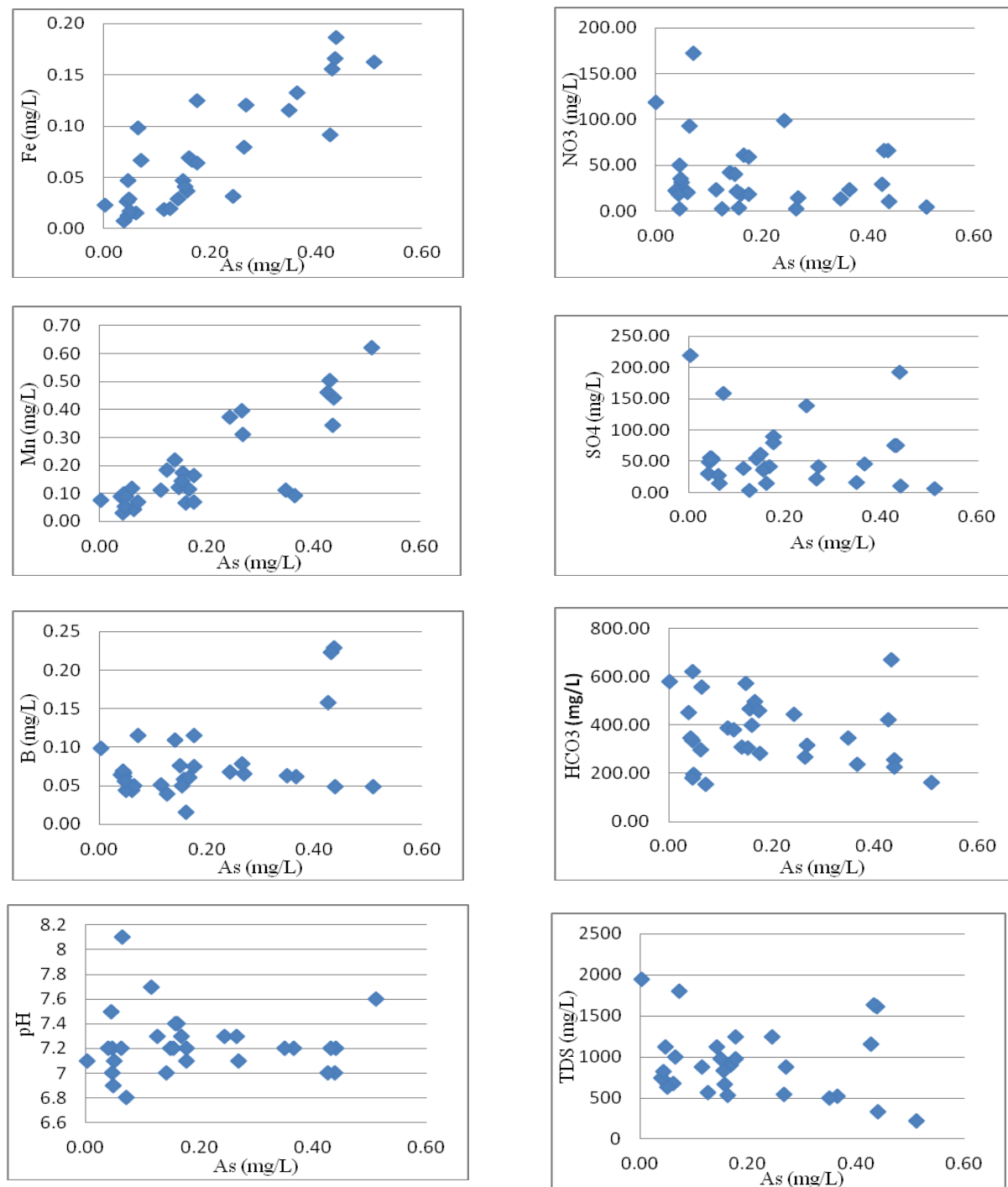


Fig. 3 Correlation between As and Fe, Mn, B, pH, NO_3 , SO_4 , HCO_3 or TDS

positive correlation between arsenic and pH probably due to desorption processes at higher pH values [26]. Further, the sample locations at Chikkondanahalli and Bairapura wells with the highest arsenic concentrations exhibited also showed no influence of pH, it might be the locations were near to the chromite mining areas.

Principal component analysis

Principal Component Analysis (PCA) is a powerful recognition technique that attempts to explain the variance of a large set of inter correlated variables with a smaller set of independent variables-principal components. PCA has been frequently employed on large and complex water quality datasets obtained from monitoring studies surface or ground waters. PCA was used to compare the compositional patterns between the examined waters samples and to identify the factors influencing each one.

The treatment of dataset with PCA, after Varimax rotation, showed three factors interpreting the 77.3% of variance (Table 4). The first PC, accounting for 36.8% of the variance in the whole data set was correlated with B, Cl, Na, K, NO₃ and SO₄. The second PC, accounting for 25.9% of the total variance is strongly correlated positively with As, Fe, Mn and K while negative correlation with Cl, HCO₃, Mg, Na, NO₃ and SO₄. The third PC 14.5% is strongly correlated with HCO₃ and K. These observations suggested that PC2 is good correlation between As and Fe or Mn which is also proved through Pearson's correlation analysis.

APPLICATIONS

Spatial distribution of arsenic in groundwater and elevated As levels presented. Further, relationship between As and Fe or Mn levels, as well as other water quality parameters are demonstrated.

This study has application in understanding the correlation between Arsenic and Fe/Mn oxyhydroxides and other quality parameters. Government or non-government agencies can take care for supply of protected water to the public in this area in order to protect their health and its associated risks.

CONCLUSION

Groundwater samples were collected from bore wells from Nuggihalli mining area and its surroundings were analyzed for arsenic and other quality parameters (pH, EC, Ca, Mg, Na, K, Cl, HCO₃, NO₃, SO₄, B, Fe, Mn). Arsenic showed high spatial variation ranged from 0.002 to 0.51 mg/L. Almost 79% of the examined wells exceeded the WHO limit of 0.010 mg/L proposed for water intended for human consumption. Bairapura, Tagadur, Jamburu and Chikkondanahalli wells exhibited extremely high arsenic concentrations (0.1–0.5 mg/L) and high potassium (200-300 mg/L). Arsenic showed strong correlation with Mn and Fe. Iron and manganese are probably the key factors in regulating arsenic concentration in groundwater. PCA has been employed and three components of groundwater with different physicochemical characteristics are observed.

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