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Distribution and sources of arsenic contaminated groundwater in parts of Thatta district, Sindh

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Abstract

Problem of natural arsenic (As) in groundwater is of growing concern to the health of people worldwide because of its carcinogenic properties. Arsenic contamination in groundwater affects the Indus alluvial and deltaic plains in Punjab and Sindh including Thatta district, where people are suffering from arsenic ingested diseases. Groundwater samples were collected from deltaic areas of Ghulamullah and Gujjo in Thatta district, where large section of population depends on groundwater for drinking and irrigation purpose. These water samples were analyzed to determine their arsenic contents, physicochemical and microbiological characteristics. Arsenic concentrations in groundwaters of Ghullamullah and Gujjo are in the range of 10-200 µg/L and 10-20 µg/L respectively. Most of As contaminated wells have also been found sewage impacted, which may be due to reducing conditions in the aquifers created by microbial degradation which favor As mobilization into groundwater. To determine the mobilization mechanism and source of arsenic, 11 soil samples were also collected from near the well sites and analyzed for their mineralogical and elemental composition using XRD and AAS techniques. Average concentrations of As in the soil of Ghulamullah and Gujjo areas are 73 and 65 µg/kg respectively. Higher As concentrations in Ghulamullah groundwater are due to dominance of clayey soil and presence of arsenic rich minerals particularly muscovite and phloghopite than in Gujjo area**.** The data reveal that the dominant hydrofacies in the study area are Na-Cl type indicating the impact of recent or ancient sea water intrusion. Arsenic distribution in shallow aquifers of Thatta district is patchy and seems to be controlled by degradation of organic matter by natural and anthropogenic sources.

Keywords: Arsenic; Pollution; Groundwater; Thatta; Sindh.

1. Introduction

Arsenic related problems have been encountered recently in several countries including Bangladesh, India, Nepal, Myanmar, Mexico, Pakistan, Vietnam, Cambodia, China and Turkey (Mukherjee et al., 2006). Arsenic is of increasing concern due to its potential carcinogenic properties (Lin et al., 2010). Most of the aquifers that are polluted by arsenic host anoxic groundwater and occur in alluvial, especially deltaic settings (McArthur et al., 2010). In Pakistan, groundwater quality is badly affected by salinity, arsenic, fluoride and microbial pollution, which further worsens in low lying, deltaic and flood plains of Sindh (Husain et al., 2012). In these areas agriculture dominated areas people mostly depend on groundwater for drinking and irrigation purpose due to scarcity of water because of low annual precipitation and reduced flow of Indus river.

Previous work on arsenic contaminated groundwater is confined to arsenic testing kit survey in Punjab and Sindh by Kahlown et al. (2004) showing that 16% and 36% population was exposed to >50 µg/L and >10 µg/L arsenic contaminated water respectively. A few studies on arsenic pollution in groundwater and its sources have been carried out in parts of Lahore, Kasur and Muzaffargarh districts (Farooqi et al., 2007; Nickson et al., 2005). Preliminary studies on As distribution in groundwater show that elevated As concentrations (10-600) exceeding WHO limit of 10 µg/L occur in groundwaters of Thatta, Tando Mohammad Khan, Matiari, Khairpur districts and other parts of Sindh (WHO, 2011; Husain et al., 2012; Naseem, 2012; Majidano et al., 2010; Husain, 2009; Arain et al., 2007). As a result people are suffering from various mild to moderate arsenicosis.

However, detailed studies regarding the arsenic distribution in groundwater, its sources and impact on health were still lacking like other As affected countries were still lacking. Present study has been carried out 1) to quantify the arsenic and pathogens concentrations in the groundwater of Ghulamullah and Gujjo areas of Thatta district 2) to understand the source and mechanism controlling the mobilization of As in groundwater.

2. Materials and methods

2.1. Geology of the study area

The study area comprising of Gujjo and Ghulamullah union councils of Thatta district lies between 24°44′46.02″N; 67°55′27.61″E at a distance of about 100 km from Karachi (Fig. 1).The climate is semi arid and irrigation in the area depends on both surface (Indus river canals) and groundwater. It is a part of Lower Indus alluvial and deltaic plaincovered by thick alluvial deltaic sediments which host the aquifers in the area (Giosan et al., 2006). Thatta district is a part of Indus delta and has been formed primarily by deposition of Late Holocene (7000-10,000 years BP) sediments carried by Indus River from Himalaya which host aquifers in the area (Giosan et al., 2006; Clift, 2002). The deltaic soil constitutes fine grained sediments, rich in organic matter containing high amount of arsenic, which is supposed to become part of aquifers by various geochemical processes (McArthur et al., 2001; Nickson et al., 2005).

Indus river has changed its course throughout the ages (Inam et al., 2007; Holmes and Western, 1969; Snelgrove, 1967). The abandoned organic matter rich courses (meander scars, oxbow lakes and levee remnants) of Indus river have been silted up and are under cultivation. This region includes alluvial plains trenched with river channels and river terraces (Siddiqui, 2007; Kazmi, 1984; Farshori, 1972).These channels are traceable from Qambar, Dadu and Hala to TandoAllayar and beyond (Kazmi and Jan, 1997; Holmes, 1968; WPWAPDA, 1966), which are hot spots of arsenic contaminated groundwaters in the study area and other As affected areas of Sindh.

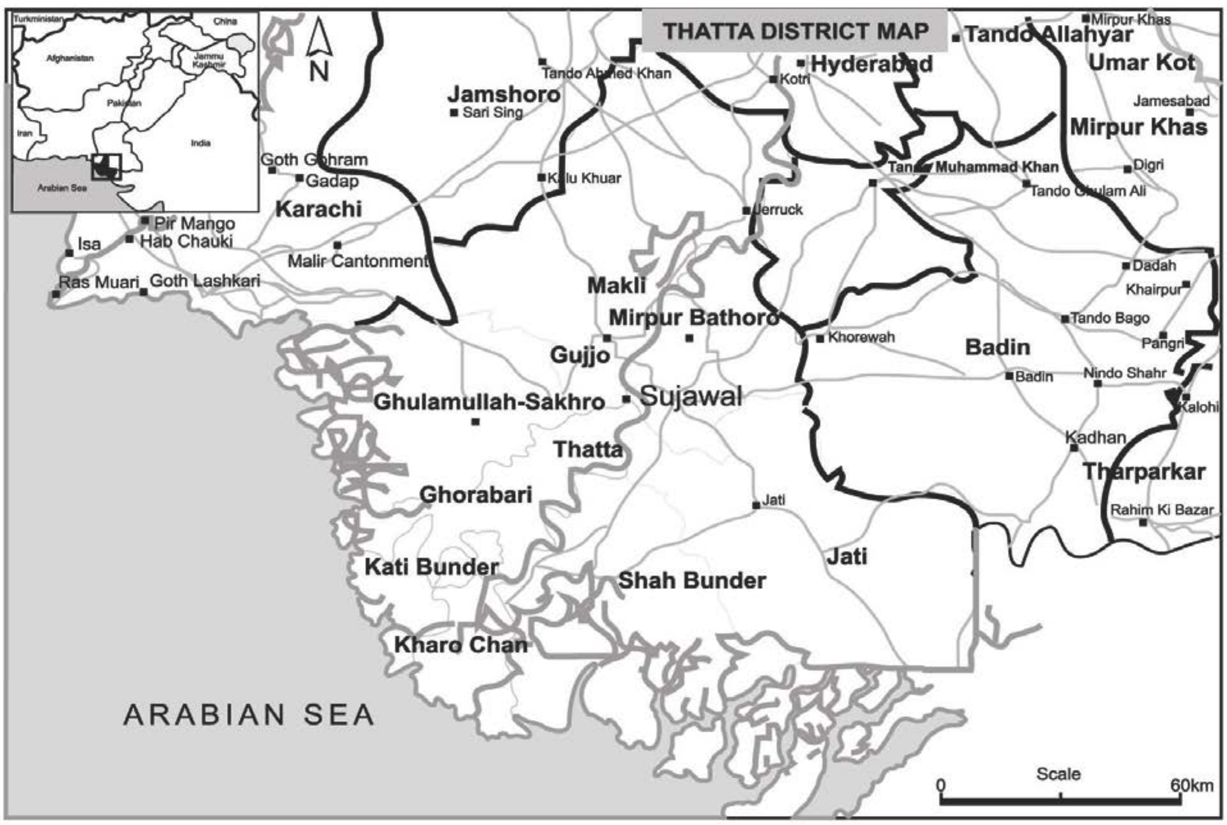

Fig. 1. Location map of the study area (modified after Ahsan et al., 2006).

2.2. Sampling and analytical procedures

Thirty seven groundwater samples, collected from hand pump wells and tube wells of Gujjo (23) and Ghulamullah (14) Union Councils of Thatta district were analyzed for various physico-chemical characteristics and fecal organics. These water samples were collected from shallow wells (5-30 m) in polystyrene bottles of 0.5 liter for nitrate determination and 1.5 liter for the analysis of physico-chemical parameters. The water samples for microbiological analysis were collected in sterile plastic bottles of 200 ml capacity, kept in icebox and transported to the laboratory within recommended time period. In the field, pH and temperature were measured using portable pH meter (HANNA, model, HI92210N, Italy). Arsenic content in the field was determined using arsenic testing kit (Merck Germany, measuring range 0.01-0.5 mg/L). Groundwater samples preserved in the boric acid were analyzed to determine the nitrate-nitrogen concentration. In the laboratory Total Dissolved Solids, HCO_3 SO_4, Na^+, K^+, Ca^+, Mg^+ and Cl^- contents were analyzed by using APHA standard analytical procedures. Microbiological analysis for Total Coliforms and E. Coli was carried out by Multiple Tube Fermentation Technique (MPN).

Eleven representative soil samples from Gujjo (5) and Ghulamullah (6) were collected from the aquifer sites at 30 cm below the surface with the help of hand shovel. Physical characteristics of soil samples were also noted. These soil samples were subjected to mineralogical study by XRD and analyzed for total (aqua-regia extractable) arsenic and iron contents. The As and Fe contents were determined by Perkin-Elmer A Analyst 600 Graphite Furnace Atomic Absorption Spectrophotometer.

3. Results and discussion

3.1. Physico-chemical characteristics of groundwater

The pH values of Ghulamullah groundwaters are 6.7-8.1 (average, 7.5) and total dissolved solids concentrations are 267-1619 mg/L (average, 839 mg/L). Higher TDS contents in the groundwater may partly be due to semi-arid climate and the effect of sea water intrusion, indicated by elevated concentrations of Na (average, 145 mg/L) and Cl (average, 218 mg/L) ion (Table 1). Whereas, high Na (average, 145 mg/L) and low Ca (average, 62 mg/L) contents in the aqueous phase could be due to increased carbonate ion concentration that favors the solubility of alkaline elements from silicic rocks and precipitation of Ca as carbonate mineral under high alkalinity respectively (Nickson et al., 2005; Lambrakis and Kallergis, 2005). While, low contents of Ca (average, 62 mg/L) and Mg (average, 47 mg/L), K (average, 8 mg/L), HCO_3 (average, 260 mg/L) and SO_4 (average, 96 mg/L) may be due to mixing of fresh water with sea water because of installation of hand pump wells along the banks of canals and water ponds (Table 1).

Major hydrofacies identified in Ghulamullah are Na-Cl–Mg-Cl (marine) type followed by Mg-HCO_3 (mixed water) and Ca-HCO_3 (fresh water). The pH values of Gujjo groundwaters are 6.6-7.7 (average, 7.3) and Total Dissolved Solids concentrations are 356-20800 mg/L (average, 905 mg/L). Higher contents of Mg (94 mg/L), HCO_3 (340 mg/L), Cl (905 mg/L), Na (517 mg/L) and SO_4 (250 mg/L) in Gujjo wells may be due to semiarid climate characterized by excess evaporation over rainfall, leading to the accumulation of salts derived from rainfall and irrigation water (Farooqi et al., 2007) or due to ancient seawater trapped during deposition (Panhwar, 1969).

This area being part of Indus deltaic plain constitutes unconsolidated Holocene sediments rich in organic matter, silicate and carbonate minerals and these ions are released in shallow (5-24m) alluvial aquifers through dissolution by organic matter degradation (Naseem, 2012; Kahlown, et al., 2004; Malik, 2000; Glover, 1959). In Gujjo groundwater, Na-Cl (marine) type hydrofacies is dominant, followed by Mg-HCO_3-Na-HCO_3 (mixed water type) and freshwater type Ca-HCO_3 (Table 2). It suggests hydro-chemical evolution of groundwater from Ca-HCO_3>Mg-HCO_3>Na-HCO_3, indicating that fresh water is trapped by seawater and cation exchange occurs within the aquifer material (Jiang et al., 2009).

Table 1. Physico-chemical and microbiological characteristics of Ghulamullah groundwater.

S #	Sample ID	pH	TDS mg/L	Ca mg/L	Mg mg/L	K mg/L	Na mg/L	Cl mg/L	HCO_3 mg/L	SO_4 mg/L	Fe mg/L	As µg/L	Fecal & presumptive Coliform	E-Coli	Water Type
1	Gh-31	7.4	1403	78	62	18	280	510	300	80	0.01	40	8	-Ve	Na-Cl
2	Gh-32	7.4	578	48	41	4	72	117	205	80	0.2	200	0	-Ve	Mg-Cl
3	Gh-33	7.1	834	88	44	12	106	195	260	112	0.91	20	6	-Ve	Na-Cl
4	Gh-34	7.4	578	48	41	4	72	117	205	80	0.2	50	0	-Ve	Mg-Cl
5	Gh-35	7.2	602	52	48	4	64	99	280	46	0.02	0	13	-Ve	Mg-HCO_3
6	36Gh-	8.1	267	40	36	4	50	96	165	45	0.16	10	11	-Ve	Mg-Cl
7	Gh-37	7.5	1619	120	75	12	283	539	360	130	0.38	200	0	-Ve	Na-Cl
8	Gh-38	7.9	668	18	62	5	125	163	190	74	0.02	0	0	-Ve	Na-Cl
9	Gh-39	7.3	474	56	27	4	53	68	220	48	0.03	0	0	-Ve	Ca-HCO_3
10	Gh-40	7.8	1109	72	55	11	197	277	310	144	0.83	50	13	-Ve	Na-Cl
11	Gh-41	7.8	783	56	34	5	144	177	260	177	0.04	20	13	-Ve	Na-Cl
12	Gh-42	7.8	562	48	44	4	62	99	250	41	0.02	0	21	-Ve	Mg-HCO_3
13	Gh-43	6.7	1078	68	44	18	219	270	300	140	0.05	60	13	-Ve	Na-Cl
14	Gh-44	7.9	1189	72	44	3	256	330	320	129	0.02	10	0	-Ve	Na-Cl
	Avg.	7.5	839	62	47	7.7	218	218	259	95	0.21	47	7		

Table 2. Physico-chemical and microbiological characteristics of Gujjo groundwater.

S. #	Sample ID	pH	TDS mg/L	Ca mg/L	Mg mg/L	K mg/L	Na mg/L	Cl mg/L	HCO_3 mg/L	SO_4 mg/L	Fe mg/L	As µg/L	Fecal & presumtive Coliform	E-Coli	Water Type
1	Gu-8	7.1	986	112	51	10	117	185	380	121	0.02	5	0	-Ve	Ca-HCO_3
2	Gu-9	7.4	933	78	45	14	148	185	340	112	0.01	0	8	-Ve	Na-Cl
3	Gu-10	6.9	2202	176	73	15	425	812	380	184	0.02	0	0	-Ve	Na-Cl
4	Gu-11	7.3	445	40	36	3	44	68	210	46	0.11	0	23	-Ve	Mg-HCO_3
5	G-u12	7	479	64	29	4	44	75	205	61	1.05	0	11	-Ve	Ca-HCO_3
6	G-u13	7.6	356	46	21	3	34	57	155	42	0.07	20	0	-Ve	Ca-HCO_3
7	G-u14	7.5	474	44	34	5	52	93	205	28	0.03	0	37	+Ve	Mg-HCO_3
8	G-u15	7.5	796	68	53	7	126	153	320	74	0.04	0	18	-Ve	Na-HCO_3
9	G-u16	7.7	2380	140	73	9	546	908	300	264	0.02	10	0	-Ve	Na-Cl
10	G-u17	7.5	2246	132	100	30	440	844	400	146	0.06	0	0	-Ve	Na-Cl
11	G-u18	7.4	783	68	56	5	91	145	320	68	0.12	5	38	-Ve	Mg-HCO_3
12	G-u19	6.8	15168	560	450	76	3640	6702	850	1240	1.17	0	0	-Ve	Na-Cl
13	G-u20	7.7	599	52	41	4	70	124	250	35	2.62	0	44	+Ve	Mg-HCO_3
14	G-u21	7.4	838	60	56	16	119	199	320	48	0.04	0	33	-Ve	Na-Cl
15	G-u22	6.6	504	40	34	5	68	121	170	44	0.03	0	48	+Ve	Na-Cl
16	G-u23	7.5	957	72	54	9	154	291	270	64	0.02	0	26	-Ve	Na-Cl
17	G-u24	7.3	664	48	39	5	106	167	200	68	0.02	5	44	-Ve	Na-Cl
18	G-u25	7.4	716	52	44	6	112	171	240	69	0.12	0	36	-Ve	Na-Cl
19	G-u26	7.7	815	52	41	14	138	188	280	66	0.08	10	29	-Ve	Na-Cl
20	G-u27	7.2	669	56	53	3	54	92	350	41	0.13	0	30	-Ve	Mg-HCO_3
21	G-u28	7.4	1786	104	85	26	318	500	460	210	0.09	0	33	-Ve	Na-Cl
22	G-u29	6.8	20800	880	632	69	4986	8635	950	2680	0.05	0	0	-Ve	Na-Cl
23	G-u30	7.6	565	44	51	5	54	89	280	28	0.08	0	59	+Ve	Mg-HCO_3
	Avg.	7.3	2442	130	94	15	517	904.5	340.6	249.5	0.26	2.39	23.5		

3.1a. Arsenic and microbial contamination of groundwater

In Ghulamullah groundwater arsenic content ranges between 10-200 µg/Lin 10 out of total 14 shallow wells in the area, seven As contaminated wells were also found sewage impacted (Table 1). While, in Gujjo groundwaters, only three wells (out of total 23) were found contaminated with As in the range of 10-20 µg/L. Though, 14 out of total 23 groundwater samples in the study are sewage impacted, but only one of these wells are also As contaminated (Table 2). Low As content in Gujjo groundwater may be due to negative correlation with Ca (Table 3). This can be explained by the dissolution of calcite in the aquifers of the area. Arsenate adsorbs onto calcite and the adsorption increases with decreasing concentration of carbonate. Presence of solid calcite slightly decreases the aqueous concentrations of arsenic (Swartz et al., 2004). It shows that the mobility of arsenic is dependent on redox conditions, with high arsenic mobility in anoxic carbonate aquifers and low mobility in oxic carbonate aquifers (Zheng et al., 2004).

The bacterial contamination of groundwater in the study area may be due to unlined sanitation, open pit toilet systems, waste water ponds, open air excretion and free roaming cattle which may also trigger the mobilization of arsenic in groundwater by creating local redox zonation in the aquifers (McArthur et al., 2001; Husain, 2009; Cole et al., 2005; Nickson et al., 2005). Elevated levels of arsenic in Ghulamullah groundwaters may be due to weak positive correlation of As with HCO_3(r=0.26) indicating that ion exchange and substitution with bicarbonate may enhance the mobility of As in aqueous phase (Table 3) (Anawar et al., 2004). Strong positive correlation of HCO_3^- with Ca (r = 0.82) and Na (r = 0.81) reflects that substantial amount of HCO_3^- is contributed by the dissolution of carbonate and silicate mineralsby water-rock interaction (Jeevanandam et al., 2007). Depth of aquifers in the study area ranges between 4.5 to 24 meters, which are vulnerable for anthropogenic pollution like sewage and agricultural runoff, which create favorable conditions for the mobilization of arsenic in groundwater, while in deeper wells (>70 m) As release takes place through geogenic processes (Nickson et al., 2005).

3.2. Arsenic distribution in soil

Soil in the study area is dominated by light brown clay followed by silty and sandy clay. Arsenic content in the Ghulamullah soil is relatively higher (45-90: mean, 73µg/kg) than in Gujjo soil (44-78µg/kg, mean: 66 µg/kg) (Table 4). Similarly, Fe content in Ghulamullah soil (71-103 mean: 98 µg/kg) is also higher than in Gujjo soil (22-92, mean: 72µg/kg).In both union councils, clayey soil contains more arsenic (average, 79 µg/kg) than silty (average, 66 µg/kg) and sandy soil (44 µg/kg).

Arsenic concentration in sediments is related to grain size and generally ranges from 1 mg/kg in sandy sediments to 20 mg/kg in clay layers (Harvey et al., 2002). Arsenic is concentrated in clay size fraction (< 2µm) and clayey sediments/soils more than sandy soils (Fitz, 2002). Subsurface soil samples in Ghulamullah contain more clay dominated soil than in Gujjo area (Table 4). Quartz, feldspar and arsenic can also be sorbed in discrete phases of Fe oxyhydroxide entrapped in argillaceous and organic rich Early-mid Holocene deltaic and floodplain sediments (Acharyya and Shah, 2007).

The XRD data show that in Ghulamullah subsurface soil, various As containing minerals-quartz, calcite, albite, muscovite and phlogopite, dolomite, chlorite-serpentines, hydrated halloysite and anorthite occur (Table 4). Arsenic contents range between <0.1-8 mg/kg in these minerals (Pichler et al., 1999). However, many of As-rich minerals including phyllosilicates (muscovite and phlogopite), dolomite and anorthite are lacking in the soil samples of Gujjo area resulting in lower As concentrations in the groundwater of Gujjo (10-20µg/L) than in Ghulamullah (10-200µg/L) wells. Phyllosilicate plays an important role in As-fixing (Ahmed, 2004). Chakraborty et al. (2007) reported that silt-sized micas provide an effective adsorption sites for arsenate and arsenite, while, Fe oxyhydroxides are important carriers of arsenic in the aquifers (Ungaro et al., 2008).

Table 3. Correlation coefficients among As, HCO_3 and major constituents in study area.

Constituents	pH	TDS	Ca	Mg	K	Na	Cl	HCO_3	SO_4	Fe
Ghulamullah (Arsenic)	-0.20	0.40	0.40	0.40	0.22	0.30	0.41	0.26	0.25	0.20
Bicarbonate	-0.25	0.89	0.82	0.54	0.54	0.81	0.78	1	0.26	0.21
Gujjo Area (Arsenic)	0.37	-0.15	-0.14	-0.18	-0.20	-0.14	-0.14	-0.25	-0.12	-0.17
Bicarbonate	-0.45	0.94	0.94	0.94	0.95	0.93	0.93	1	0.89	0.07

Table 4. Mineralogical and arsenic-iron contents in soil of the study area.

Area	Sample No.	Mineral	Sediment characteristics	Fe (mg/kg)	As (μg/kg)
Ghulamullah	10	Quartz, Calcite, Albite, calcian, Phlogopite-1	Light brown silty clay.	98	81
	11	Quartz, Albite, calcian, Muscovite-2	Medium brown, clay with plant roots.	116	81
	12	Quartz, Albite ordered, Calcite, magnesian	Light brown, Silty clay.	71	45
	13	Quartz, Phlogopite-1, Calcite, magnesian, Albite, calcian,, Hydrated Halloysite	Brown clay	103	90
	14	Quartz, Calcite, Phlogopite-1, Anorthite,	Medium brown, clay.	103	76
	15	Quartz, Calcite, Anorthite, ordered, chlorite-serpentines, dolomite, Li bearing muscovite	Light brown silty clay.	96	63
Gujjo	1	Quartz, albite, calcian, Calcite	Light brown, Sandy clay muscovite particles.	22	44
	2	Quartz, Zinnwaldite, Calcite, magnesian, Chlorite-serpentine, Albite, calcian	Silty clay, light brown. Contain plant debris.	91	76
	3	Quartz, Calcite, Albite,	Silty clay with muscovite particles.	64	62
	4	Quartz, Calcite, Albite,	Dark brown clay.	90	69
	5	Quartz, Calcite, Albite, calcian	Light brown silty clay, muscovite particles.	92	78

3.3. Mechanism of arsenic mobilization in the groundwater

Most of the As contaminated groundwater across the globe are of Quaternary age (Smedley, 2002). Similarly, in the study area As is mainly associated with Holocene fine grained organic rich sediments dominated by sandy and silty clay. The arsenic contaminated wells have a spatial association with abandoned channels, oxbow lakes and swamps (Acharyya and Shah, 2007; Mukherjee and Bhattachraya, 2001). Most of the aquifers in the study area are located in these ancient channels.

Chemical analysis and X-ray diffraction of these organic rich detrital fractions show the abundance of mica dominated by phlogopite and muscovite along with anorthite and dolomite in Ghulamullah clay dominated soil, which explains higher As contents in the soil and groundwater of Ghulamullah than in Gujjo area. The fine sized micas and clays provide effective sites for adsorption of arsenic (Chakraborty et al., 2007).

Biogenic reductive dissolution of Fe-oxyhydroxides is another important mechanism that put arsenic into groundwater of the alluvial aquifer under reducing conditions (Von Brömssen et al., 2007; Ahmed et al., 2004; McArthur et al., 2001; Nickson et al., 1998; Bhattacharya et al., 1997). The unlined sanitation and sewage contamination in the study area has also resulted in the degradation of organic matter by micro-organisms in the Holocene aquifers of the study area, causing heterogeneous local reducing conditions in the aquifers and generating high concentration of HCO_3^- and dissolved organic matter in the groundwater (Hasan et al.,2007; Bhattacharya and Mukherjee, 2002). Positive relationship between As and sewage contamination of groundwater in the study area suggests microbial processes in these organic rich alluvial aquifer sediments, which create a favorable anoxic environment and permit reduction of iron oxyhydroxides (FeOOH) and release of sorbed arsenic to solution (Bhattacharya et al., 1997; Nickson et al., 1998, 2000, McArthur et al., 2001). The distribution pattern of arsenic in groundwater is patchy with the observation of many hotspots along various geomorphological features and bank of Indus river resembling with As enrichment along West Bengal Hoogly river and its abandoned channels (Naseem, 2012; Husain, 2009; Nath et al., 2009; Charlet et al., 2007; Bhattacharya et al., 1997).

4. Conclusions

Groundwater in Thatta district is highly saline, due to semiarid climate, excessive evapotranspiration, connate water and recent seawater intrusion affecting soil and groundwater quality of this area. Depth of the wells in the area is very shallow, which led to the microbiological contamination of groundwater through unlined sanitation pit toilets and surface runoff. Elevated concentrations of arsenic in the soil and groundwater of the study area are mainly associated with abandoned courses of Indus river, and confined to Holocene fine grained, silty, clayey organic rich sediments. Biogenic reductive dissolution of Fe-oxyhydroxides release arsenic from these sediments in the groundwater. Moreover, in Ghulamullah area, high concentration of arsenic as compared to Gujjo area is due to soil texture and mineralogy and prevalence of anoxic conditions in Holocene aquifers. The bacterial contamination of groundwater is causing heterogeneous local reducing conditions in the aquifers which may trigger the mobilization of arsenic in groundwater. Thus, both geogenic and anthropogenic factors have contributed to the arsenic contamination of soil and groundwater in the study area.

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