Journal of Hazardous Materials 171 (2009) 189–199

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Nitrate contamination in groundwater of some rural areas of Rajasthan, India

Surindra Suthar[a,*], Preeti Bishnoi[b], Sushma Singh[c], Pravin K. Mutiyar[a], Arvind K. Nema[a], Nagraj S. Patil[a]

[a] Department of Civil Engineering, Indian Institute of Technology Delhi, Hauz Khas, New Delhi 110016, India
[b] P.G. Department of Zoology, B.R.G. Govt. Girls PG College, Sri Ganganagar 335001, Rajasthan, India
[c] Department of Chemistry, N.M. PG College, Hanumangarh Town 335513, Rajasthan, India

ARTICLE INFO

Article history:
Received 19 May 2009
Received in revised form 25 May 2009
Accepted 25 May 2009
Available online 6 June 2009

Keywords:
Drinking water quality
N-fertilizer
Nitrate leaching
Animal manure
Pit latrines

ABSTRACT

Efforts were made to evaluate the level of nitrate in some agro-economy based rural habitations of northern Rajasthan, India. A total of 64 groundwater samples from 21 different villages/sub-villages of district Sri Ganganagar, India were collected and analyzed for nitrate (as NO_3^-), sulphate (as SO_4^{2-}) and few other parameters. NO_3^- level in groundwater was 7.10–82.0 mg l^{-1} for individual samples. But average NO_3^- for total samples was 60.6 ± 33.6 (SD) mg l^{-1}, which indicates the non-suitability of groundwater for drinking purposes, if BIS permissible limit (22.6 mg l^{-1}) is considered as reference level. SO_4^{2-} ranged form 28.6 to 660.3 mg l^{-1} in this area. The regression analysis indicates the difference sources for NO_3^- and SO_4^{2-} contamination in different regions rather than a common source. The point and non-point sources of NO_3^- and SO_4^{2-} in groundwater of this region may be N-fertilizer, sewerage, animal waste, organic manure, geology of sub-surface soil layers, pit latrines, etc. Results thus indicated that groundwater of this part of the State is severely polluted due to anthropogenic activities. The continuous consumption of such water may pose serious health hazardous in local residents.

1. Introduction

The chronic impact of chemical contamination of groundwater is more dreadful particularly in rural areas of developing world [1] where groundwater is the main assessable source for potable water. Groundwater can have some dissolved forms of chemicals, which may be unacceptable due to their chronic health effects, taste and aesthetic reasons [2]. Undesirable chemicals in groundwater may cause very serious health problems, whether the chemicals are naturally occurring or derived from source of pollution. The World Health Organization (WHO), a premier and most prestigious international health organization has published a guideline on the chemical safety of drinking water. According to the guideline there are two main criteria for identifying specific chemicals of concern to public health: high probability of consumer exposure from drinking water and, significant hazard to health [3]. Several chemicals, whether occurring naturally or due to anthropogenic activities, present different level of health hazardous in humans.

Nitrate contamination in groundwater is a common problem in many part of the world arising from diffuse reasons, e.g. intensive agriculture, unsewered sanitation in densely populated areas, or from point sources such as irrigation of land by sewage effluents. Nevertheless, the heavy use of nitrogenous fertilizers in cropping system is the largest contributor to anthropogenic nitrogen in groundwater worldwide. Nitrogenous fertilizer rapidly converts into NO_3^- form in soils, which is readily available to plants, but is highly soluble and hence easily leachable to deep soil layers. When quantity of nitrogen added to the soil exceeds the amount that the plants can use, the excess NO_3^- does not get much adsorbed by soil particles, leaches out from the root zone by water percolating through the soil profile and ultimately accumulates into the groundwater [4]. Since NO_3^- is the part of nitrogen cycle in nature and it represents the most oxidized chemical form of nitrogen found in the natural systems. Also, it is an essential part of building blocks of living organism, i.e. protein, genetic materials (DNA and RNA), vitamins, hormones and enzymes [5]. But human health consequences of exposure to high nitrate levels are of great concern. Greater NO_3^- intake reduces the oxygen-carrying capacity in the blood by binding to hemoglobin, causing a condition referred to as methemoglobinemia or "blue baby syndrome," which may cause mortality by asphyxiation especially in newly born infants. However, infants less than six months of age are at highest risk due to the presence of bacteria in their digestive systems that speed the binding process. Recent studies have revealed that nitrate can be endogenously reduced to nitrate, which can then undergo nitrosation reactions in the stomach with amines and amines to form a variety of *N*-nitroso compounds (NOC) [6], which are mainly carcinogens [7]. The continuous consumption of water containing high

* Corresponding author. Tel.: +91 9460121182; fax: +91 1126586111.
E-mail address: sutharss_soilbiology@yahoo.co.in (S. Suthar).

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doi:10.1016/j.jhazmat.2009.05.111

Table 1
Nitrate (as NO_3^-) contamination in some regions of India.

Sampling/study site	State	Range	Ref.
Ludhiana district	Punjab	0.31–13.3 mg l^{-1}	Bijay-Singh et al. [8]
Kanpur district (n = 297)	Uttar Pradesh	1.0–166 mg l^{-1}	Sankararamakrishnan et al. [2]
Hooghly district (n = 412)	West Bengal	0.01–4.56 μg ml^{-1}	Kundu et al. [4]
Nadia district (n = 342)	West Bengal	0.01–5.97 μg ml^{-1}	Kundu and Mandal [21]
Anantapur district (n = 48)	Andhra Pradesh	3.0–684 mg l^{-1}	Reddy et al. [5]
New Delhi (n = 95)	Delhi	0.04–98.3 mg l^{-1}	Data et al. [14]
Jaipur district (n = 5)	Rajasthan	26–459 mg l^{-1}	Gupta et al. [13]

Table 2
N based fertilizer consumption in Rajasthan.

N-based fertilizer	Tonnes			
	1996–1997	1997–1998	1998–1999	1999–2000
Urea	1057.38	1132.41	1007.69	1031.50
Ammonium sulphate	1.95	3.44	3.35	2.17
Calcium ammonium nitrate (CAN)	14.29	13.83	12.69	13.28
Di-ammonium phosphate (DAP)	246.77	322.75	330.79	453.18

Table 3
Livestock population and its contribution in total NPK production in Sri Ganganagar district.

Livestock	Total population[a]	Tones nutrient $year^{-1}$ [b]		
		N	P	K
Cattle	70,1805	35,090.25	10	7,018,050
Pigs	3,865	46.38	4	15,460
Sheep	33,8962	3,389.62	2	677,924
Goats	268,853	2,688.53	2	537,706
Horses	1,058	47.61	8	8,464
Poultry	132,113	792.678	0.19	25,101.47
Total[c]	1,622,516			

[a] Census 2001 [16].
[b] Based on nutrient production rate as calculated by Sheldrick et al. [29].
[c] Including all other livestock populations.

nitrate may cause several health hazardous in animals, e.g. gastrointestinal cancer, Alzheimer disease, vascular dementia, absorptive, secretive functional disorders of the intestinal mucosa, multiple sclerosis, Non-Hodgkin's lymphoma, hypertrophy of thyroid, etc.

In developing countries like India NO_3^- enrichment in groundwater has been appearing as a major threat in few intensively

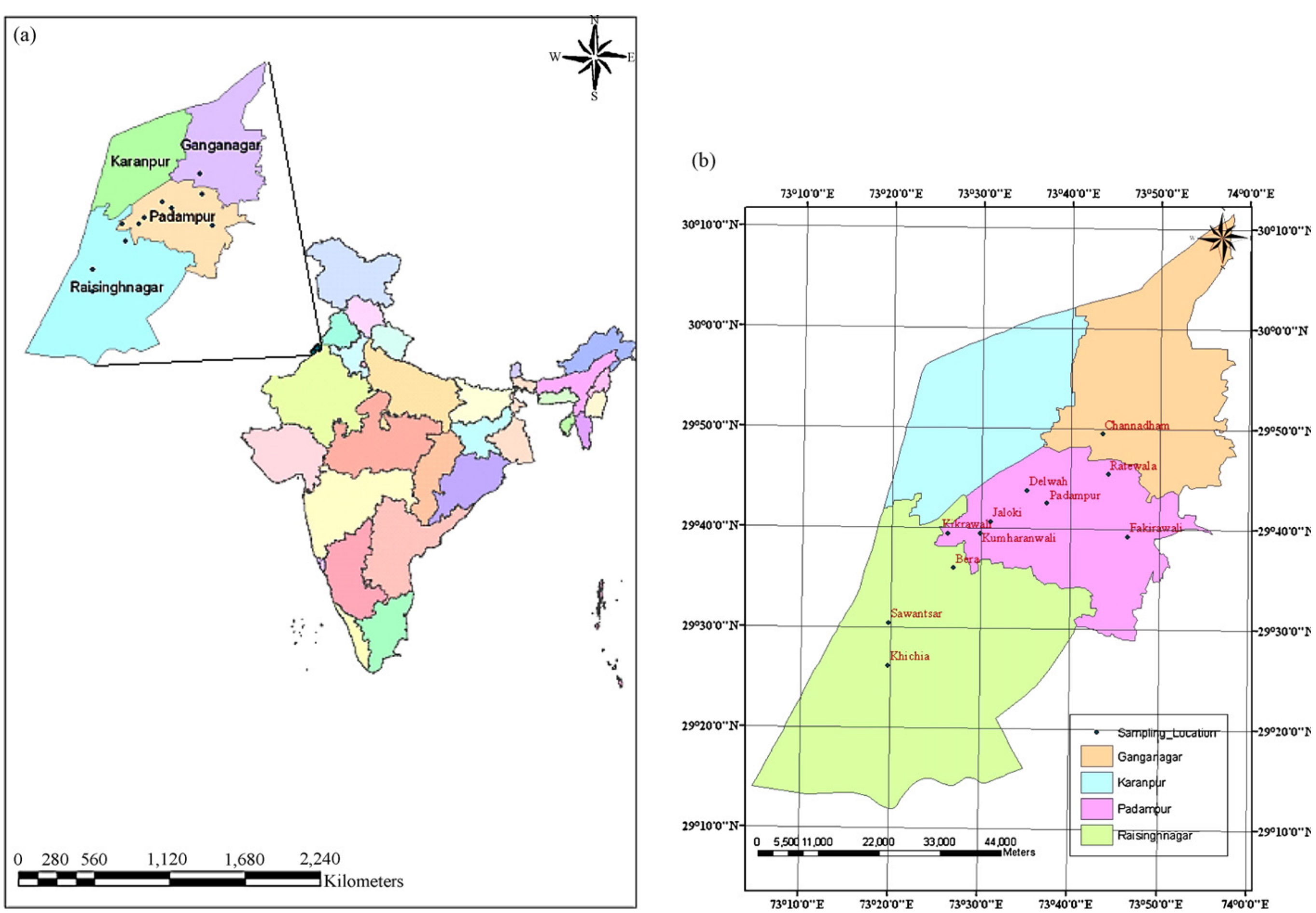

Fig. 1. (a) Location map of study area in India and (b) sampling location in the study area.

Table 4
Sampling sites description.

Village	Sampling in sub-division (if any)	No. of samples collected
Fakirawali	*2 RB, 3 RB*	5
Jaloki	*5 RB, 6 RB, 40 RB, 41 RB*	13
Delwah	*2 DD, 3 DD*	5
Channa dham	*14 BB, 15 BB, 16 BB*	10
Ratewala	*CC Head*	2
Sawantsar	*4 EEA*	2
Padampur	*Padampur main, 17 BB, 19 BB, 20 BB, 21 BB*	15
Bera	*23 BB*	2
Kikrawali	–	4
Kumharanwali	–	3
Khichia	*1 JJ*	3

cultivable Sates: Punjab [8,9], Haryana [10], Maharastra [11], Andhra Pradesh [12], Utter Pradesh [2], West Bengal [4], Rajasthan [13] and Delhi [14] (Table 1). The high NO_3^- in groundwater in these areas was due to heavy use of nitrogenous fertilizer in crop lands. After reviewing current literature, we realized that a little part of the country is explored in terms of groundwater NO_3^- contamination. Since, agriculture is the major land use system in the country and a comprehensive study to evaluate the NO_3^- contamination in groundwater of some other part of the country is still needed. The northern part of Rajasthan is considered as highly cultivated area in the State. It is the part of north-western arid land of Indian. But introduction of canal irrigation system had changed the whole picture of this part during last 50 years of the Green revolution. The fertilizer consumption in this area has been increased day by day (Table 2). Total N-based fertilizer consumption was approximately 1500 tonnes in 1999–2000 followed by a rapid increase in subsequent years. However, there appears to be no report on the groundwater quality with respect to NO_3^- in this part of the country. However, livestock rearing and animal farming is also an important occupation of farmers and local residents. The total livestock population is comparatively higher in this part (Table 3) possibly due to excellent agriculture outputs. Such huge livestock populations produce a considerable amount of N which may raise several environmental problems in this area.

In this study efforts were made to evaluate the level of NO_3^- in groundwater of some selected rural habitations of Sri Ganganagar district of Rajasthan, India (Fig. 1a).

2. Materials and methods

2.1. Study area

The study sites are located in the extreme north of Rajasthan (Fig. 1b). Geographically it is located between the latitude 28°4′–30°6′N and the longitude 72°3′–75°3′E. According to the agro-ecological-zone classification [15], it is an Aeolian plain of northern canal irrigated sub-region. Geologically the area is covered by wind blown isolated sand and alluvium except for a few patches of recent calcareous and sandy sediments associated with gypsite. The oldest rocks of the area belong to Aravalli Super Groups which includes phyllite, shale and quartz vein. The soil is mainly developed from the alluvium of variable texture and at the alluvium is buried under the wind worked sand. These alluvial soils are moderately coarse textured, deep to very deep, underlained by weak concretionary zone and have been classified as Torrifluvents. The climate of this region is semi-arid with extreme temperature conditions in summer (up to 47.3 °C) and winter (up to 1.0 °C). The site is influenced by the Indian southern-west or summer monsoon (June–September) and during winter (December–February) by Siberian anticyclones. The main rainy season is during July and August which receive approximately 80% of the annual rainfall. The annual mean rainfall was 258.1 mm during study year. The total human population of district is about 1.79 million (2001 census) and out of this nearly 74.7% live in rural areas. According to census [16] tehsil-wise urban and human population ratio was 1:0.9 in *Sri Ganganagar* tehsil, 1:7.15 in *Padampur* tehsil and 1:5.7 in *Raisinghnagar* tehsil. The rural human population is dominant in this part of the State and approximately 47.5%, 87.7% and 85% of total human population in *Sri Ganganagar*, *Padampur* and *Raisinghnagar* tehsil, respectively inhabits in rural areas.

About 87.0% of the total land of the district is gross cropped area and 11.9% land is under forest cover. Permanent pasture and other grazing land is approbatory 0.002%, while 3.33% of total land is under fallow land category. The cultivable land is rich in

Table 5
Characteristics of groundwater in different villages.

Village/sub-division	No. of samples	pH			EC (mS)			Turbidity (NTU)		
		Range	Mean	SD	Range	Mean	SD	Range	Mean	SD
2 RB	3	7.29–7.36	7.32	0.04	2.05–2.26	2.13	0.12	0.11–0.12	0.11	0.01
3 RB	2	7.55–7.62	7.59	0.05	1.32–1.39	1.36	0.05	0.006–0.007	0.007	0.0007
5 RB	3	7.47–7.50	7.47	0.02	0.62–0.63	0.63	0.006	0.09–0.10	0.09	0.006
6 RB	3	7.65–7.78	7.72	0.07	9.56–9.70	9.62	0.07	0.006–0.007	0.007	0.0006
40 RB	3	7.29–7.32	7.30	0.02	4.47–4.62	4.56	0.08	0.005–0.006	0.006	0.0006
41 RB	4	5.96–6.06	6.02	0.04	0.001–0.002	0.002	0.0006	0.080–0.090	0.085	0.006
2 DD	3	7.08–7.10	7.09	0.01	0.696–0.687	0.691	0.006	0.090–0.100	0.095	0.007
3 DD	3	7.10–7.20	7.15	0.05	5.60–5.70	5.66	0.055	0.011–0.120	0.047	0.062
14 BB	4	7.10–7.15	7.13	0.02	3.99–4.12	4.05	0.064	0.10–0.11	0.102	0.005
15 BB	3	7.36–7.43	7.41	0.04	2.98–3.18	3.06	0.105	0.14–0.15	0.143	0.006
16 BB	3	7.09–7.13	7.11	0.02	0.00–0.001	0.0006	0.0006	BDL	–	–
CC Head	2	7.29–7.35	7.32	0.04	1.30–1.36	1.33	0.042	0.130	0.130	–
4 EEA	2	7.01–7.05	7.03	0.03	4.18–4.20	4.20	0.021	0.270–0.280	0.275	0.007
17 BB	2	8.29–8.41	8.35	0.08	6.18–6.34	6.26	0.113	0.090	0.090	–
19 BB	3	6.39–6.54	6.45	0.08	1.32–1.49	1.40	0.085	0.090–0.100	0.097	0.006
20 BB	3	7.29–7.30	7.30	0.01	1.23–1.26	1.24	0.015	0.100–0.110	0.106	0.005
21 BB	3	8.01–8.14	8.09	0.07	1.68–1.70	1.69	0.100	0.58–0.59	0.586	0.006
23 BB	3	8.41–8.45	8.43	0.02	1.05–1.12	1.07	0.038	0.089–0.090	0.089	0.0006
Kikrawali	4	7.95–8.01	7.98	0.03	1.69–1.72	1.71	0.015	0.100–0.110	0.105	0.006
Kumharanwali	3	6.82–6.83	6.82	0.01	6.28–6.29	6.283	0.006	0.120–0.130	0.123	0.006
1 JJ	3	7.26–7.31	7.29	0.03	3.48–3.50	3.49	0.010	BDL	–	–
Padampur (urban)	4	7.34–7.37	7.35	0.01	3.50–3.52	3.51	0.01	0.05–0.07	0.06	0.01
Total	65	5.96–8.45	7.32	0.57	BDL–9.70	2.88	2.39	BDL–0.590	0.105	0.0120

Table 6
Characteristics of groundwater in different villages.

Village/sub-division	No. of samples	Chloride ($mg\,l^{-1}$)			Total hardness ($mg\,l^{-1}$)			Fluoride ($mg\,l^{-1}$)		
		Range	Mean	SD	Range	Mean	SD	Range	Mean	SD
2 RB	3	107.0–108.7	107.9	0.85	8.70–9.09	8.90	0.20	0.072–0.081	0.077	0.005
3 RB	2	45.0–45.4	45.2	0.34	7.60–7.69	7.64	0.06	0.125–0.127	0.126	0.001
5 RB	3	2340.1–2401.7	2360.8	35.5	5.90–5.98	5.93	0.042	0.014–0.015	0.014	0.0006
6 RB	3	56.2–96.2	80.6	21.4	4.0–4.01	4.0	0.006	0.023–0.024	0.023	0.0006
40 RB	3	781.0–783.7	782.3	1.34	7.04–7.05	7.04	0.006	0.059–0.061	0.060	0.001
41 RB	4	1579.1–1590.3	1583.9	4.67	6.12–6.20	6.14	0.037	0.034–0.041	0.0385	0.003
2 DD	3	539.6–540.0	539.8	0.29	8.60–8.61	8.61	0.007	0.019–0.020	0.020	0.0007
3 DD	3	635.1–647.2	639.5	6.69	7.44–7.52	7.48	0.040	0.022–0.024	0.023	0.001
14 BB	4	600.1–609.1	604.4	3.71	8.09–8.20	8.16	0.048	0.059–0.065	0.062	0.0025
15 BB	3	340.1–356.1	346.6	8.38	6.81–6.85	6.83	0.020	0.023–0.025	0.0236	0.001
16 BB	3	50.6–52.1	51.2	0.76	7.79–7.84	7.81	0.026	0.043–0.044	0.043	0.0005
CC Head	2	50.8–52.5	51.63	1.22	5.96–6.00	5.98	0.028	0.030–0.039	0.0345	0.007
4 EEA	2	42.3–52.3	47.3	7.1	6.01–6.04	6.02	0.021	0.027–0.029	0.028	0.001
17 BB	2	44.1–53.0	48.6	6.29	9.98–10.02	10.0	0.028	0.096–0.101	0.0985	0.004
19 BB	3	56.7–57.0	56.8	0.149	8.04–8.44	8.28	0.21	0.017–0.100	0.453	0.047
20 BB	3	172.7–179.4	176.6	3.50	9.40–9.47	9.44	0.035	0.070–0.090	0.081	0.010
21 BB	3	169.2–185.6	179.7	9.09	6.60–6.76	6.68	0.080	0.11–0.12	0.113	0.006
23 BB	3	82.6–84.0	83.4	0.76	7.59–7.69	7.64	0.050	0.029–0.036	0.032	0.004
Kikrawali	4	1025.4–1369.5	1217.1	143.2	2.40–4.70	3.52	1.24	0.074–0.081	0.076	0.003
Kumharanwali	3	75.8–89.5	83.1	6.89	5.84–6.08	5.96	0.12	0.021–0.026	0.023	0.002
1 JJ	3	472.7–483.7	479.7	6.11	5.20–6.39	5.86	0.61	0.074–0.080	0.0763	0.003
Padampur (urban)	4	433.1–759.7	524.5	157.1	6.88–10.32	9.09	1.54	0.13–0.13	0.131	0.002
Total	65	42.3–2401.7	639.5	6.69	2.40–10.32	7.48	0.041	0.014-–0.13	0.023	0.001

agricultural products on account of a well-developed system of canal irrigation. A large network of Gang Canal and Bhakhra Canal which along with the Indira Gandhi Canal, had converted the entire area into a green and productive belt of the State. Wheat, cotton, rape and mustered, pulses, gram, sugarcane, etc. are the major crops of this belt. It is considered as the highest wheat production region of the State and popularly known as the 'food basket' of Rajasthan. The total sowed area and production rate for wheat was in the ranges of 173,534–192,668 ha and 481,910–686,230 tones, respectively during the years 2003–2007. The economy of this region is based on agriculture and, therefore livestock population is comparatively higher 1,622,516 (2001 census) in this part of the State.

2.2. Water samples collection

A total of 64 samples were collected from different sites of 11 different villages/towns namely, *Fakirawali*, *Jaloki*, *Delwah*, *Channadham*, *Ratewata*, *Sawantsar*, *Padampur*, *Bera*, *Kikrawali*, *Kumharanwali*, *Khichia* during September–October 2008 (Fig. 1b). The description of sampling sites and sub-sites is given in Table 4. Groundwater samples were collected in pre-cleaned plastic bottles of 1 l capacity after the extraction of water either from privately owned manually operated hand-pumps or from electricity operated bore-wells. The water was left to run from the source for about 4 min to equate the minimum number of well volume and to stabilize the electrical conductivity. Samples were taken by holding the bottle at the bottom to avoid any contamination and were analyzed

Table 7
Characteristics of groundwater in different villages.

Village/sub-division	No. of samples	Nitrate (as NO_3^-) ($mg\,l^{-1}$)			Sulphate (as SO_4^{2-}) ($mg\,l^{-1}$)		
		Range	Mean	SD	Range	Mean	SD
2 RB	3	73.0–76.0	74.3	1.53	220.7–224.8	222.3	2.17
3 RB	2	39.0–42.0	40.5	2.12	118.4–120.6	119.5	1.54
5 RB	3	73.0–78.0	75.0	2.64	28.2–28.9	28.6	0.35
6 RB	3	81.0–82.0	81.3	0.58	652.7–664.8	656.9	6.89
40 RB	3	57.0–59.0	57.7	1.15	430.7–432.2	431.6	0.74
41 RB	4	75.0–77.0	76.0	0.82	640.8–652.2	646.4	5.39
2 DD	3	42.0–43.0	42.5	0.71	32.2–32.7	32.5	0.28
3 DD	3	57.0–58.0	57.3	0.58	635.4–649.6	644.2	7.72
14 BB	4	45.0–48.0	46.2	1.50	562.5–581.1	573.0	8.70
15 BB	3	7.10–7.26	7.18	0.080	227.9–232.0	229.6	2.13
16 BB	3	36.0–37.0	36.3	0.58	632.5–642.5	637.1	5.03
CC Head	2	78.0–79.0	78.5	0.71	62.2–65.1	63.6	2.02
4 EEA	2	65.0–69.0	67.0	2.82	658.6–662.1	660.3	2.52
17 BB	2	63.0–65.0	64.0	1.41	118.4–120.0	119.3	1.15
19 BB	3	20.0–24.0	21.7	2.08	59.9–62.5	61.23	1.31
20 BB	3	68.0–72.0	69.7	2.08	51.1–52.7	52.2	0.94
21 BB	3	18.0–19.0	18.3	0.58	184.2–186.2	185.2	0.99
23 BB	3	58.0–61.0	59.3	1.53	142.1–145.0	143.2	1.59
Kikrawali	4	30.0–32.0	31.0	0.82	190.4–196.1	192.9	2.42
Kumharanwali	3	80.0–82.0	80.66	1.15	638.6–647.0	641.5	4.75
1 JJ	3	59.0–61.0	59.66	1.15	511.5–517.2	513.6	3.11
Padampur (urban)	4	160.0–162.0	161.0	1.15	203.6–204.7	204.2	0.52
Total	65	7.10–162.0	60.57	33.6	28.2–664.8	335.3	242.5

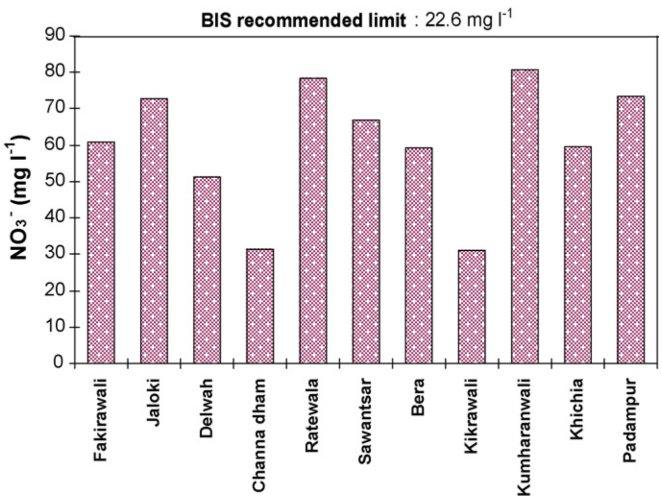

Fig. 2. Concentration of NO_3^- in different villages of northern Rajasthan.

just after the sampling. The collected sample bottles were kept in airtight large plastic ice-cold containers and were transported to laboratory within 6 h of their collection for further processing. In lab samples were stored at low temperatures. Groundwater samples were analyzed for level of pH, turbidity, electrical conductivity (EC), total hardness (TH), fluoride (F^-), sulphate (as SO_4^{2-}), nitrate (as NO_3^-), chloride (Cl^-), etc. by following method as described by APHA [17].

2.3. Analysis

pH and EC of sampled water were analyzed using pH meter (model pH-538) WTW (Germany) and Systronic conductivity meter (model 306), respectively. Turbidity of water samples was measured using ESICO turbidity meter (model 335 E). Titration assembly was used to measure chloride and TH by following method as described by APHA [17]. Nitrate and fluoride content in water samples were measured using ion selective electrodes (Thermo® Electro Co., USA). Sulphate level in groundwater was analyzed by using method described by APHA [17]. ArcGIS 9.2® computer program and IDW technique were used for generation of interpolation grid and isolines for nitrate and sulphate.

2.4. Reagents and standards

Analytical grade (AR) chemicals were used throughout the study without any further purification. To prepare all the reagents and calibration standards, double glass distilled water was used. Deionized water was used throughout the study. The glasswares were washed with dilute nitric acid (1.15) followed by several portions to distilled water [18]. All the experiments were carried out in duplicate. The results were reproducible within ±3% error limit.

2.5. Statistical analysis

A regression coefficient was calculated for different chemical characteristics of groundwater to establish the relationship between two parameters. SPSS® statistical package (Window Version 13.0) was used for data analysis. All statements reported in this study are at the $P < 0.05$ levels.

3. Results and discussions

3.1. Groundwater quality: general parameters

Water quality parameters reflect the level of contamination in water resources. In this study the physico-chemical quality of drinking water varied drastically among different sampling sites. To compare the suitability of water for drinking purposes, we referred the standard ranges for different chemicals in drinking water as prescribed by WHO [19] and Bureau of Indian Standards (BIS) [20]. The drinking water samples were free from colour, odour and turbidity. The taste was slightly to moderately saline at some of sampling sites, except in few sites where pH fall in acidic ranges. The pH of all sampling sites ranges from 5.96 to 8.45. The maximum pH range was *23 BB* (8.41–8.45), while *41 RB* (5.96) exhibited the pH in acidic ranges. Similarly, *19 BB* and *Kumharanwali* also showed pH lower than 7 (Table 5). Since, pH has no direct effect on human health, it shows close relations with some other chemical constituents of water and consequently influences chemical kinetics of important constituents of water. EC is an important character of potable water and its value signifies the amount of total dissolved salts, which in turn indicates the inorganic pollution load of water. There were large variations in EC values not only in the samples collected form different villages, but also in the samples collected form the same localities, which suggested the spatial variations in water geochemistry of this region. EC of water ranged BDL–9.70 mS. Some sampling sites, i.e. *41 RB* and *16 BB* showed least values of EC (mS). The maximum range of EC was in samples collected form *6 RB* (9.56–9.70 mS). The higher EC at *Jaloki*, *Padampur* and *Kumharanwali* sub-regions may be the attributed to high salinity and high mineral content in these sites. Also, the greater EC of water is the result of ion exchange and solubilisation in the aquifer. Turbidity in water samples ranged BDL–0.590 NTU in different localities of this region. Sites of *Sawantsar* and *Padampur* villages showed relatively higher turbidity values (Table 5).

Hardness is a very important property of water forms its domestic application point of view. TH of water samples ranged between 2.40 and 10.32 mg l^{-1} for individual sample, while mean of TH for studied locality was between 3.52 (*Kikrawali*) and 10.0 mg l^{-1} (*17 BB*) (Table 6). The acceptable limit of total hardness (as $CaCO_3$) is 200 mg l^{-1}, which can be extended up to 600 mg l^{-1} in case of non-availability of any alternate water source. Chloride is considered as an important inorganic constituent of water, which may deteriorate the quality of drinking water at higher extents. The Cl^- contents ranged between 45.2 (*3 RB*) and 2360.8 mg l^{-1} (*5 RB*) in different localities of northern Rajasthan. The permissible limit of Cl^- in potable water is 200 mg l^{-1}, which may be further relaxed up to 1000 mg l^{-1} for Indian conditions. The greater concentration of Cl^- in groundwater could be associated with chloride rich minerals or likely to be originated from pollution sources, e.g. domestic effluents, fertilizers and septic tanks. The greater level of Cl^- in some rural habitations, e.g. *41 RB*, *Kikrawali* and *5 RB* may be due to leaching processes at animal waste dumping sites. It is also suggested that pit latrines also contribute in chloride enrichment of groundwater of this region, although further detailed study is required to strengthen the hypothesis. In this study fluoride concentration was recorded of least significance. Fluoride ranged between 0.014 and 0.13 mg l^{-1} (Table 6) in this area. Fluoride in groundwater may be due to the presence of fluoride bearing minerals in host rocks and their interaction with groundwater. Also, fluoride accumulation in groundwater in different areas varies according to the geological formation of the area, amount of rainfall and quality of water lost by evaporation. The lower fluoride may be attributed to the absence of fluoride bearing rocks in these areas.

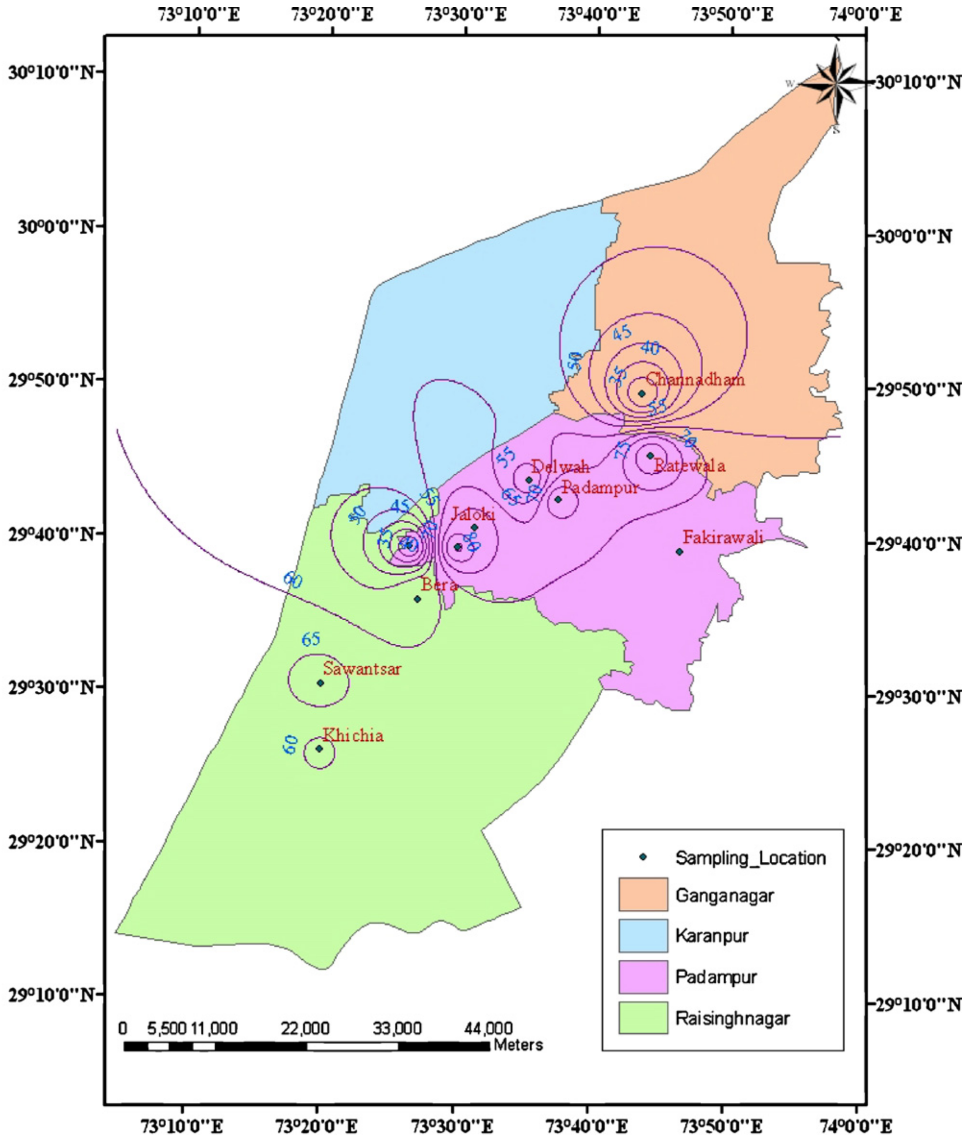

Fig. 3. NO_3^- isolines over the study area.

More interestingly Suthar et al. [1] reported relatively higher fluoride in areas of adjoined district, i.e. *Hanumangarh* fluoride. The lower fluoride in these areas indicates the spatial variations in geology of northern part of the Rajasthan. Results of this study suggested that fluoride in groundwater is not a health problem for this region; although fluoride is considered endemic for Rajasthan State.

3.2. Nitrate and sulphate in groundwater

There was great variation in nitrate (as NO_3^-) level in groundwater of this region. NO_3^- concentration ranged between 7.10 and 162.0 mg l$^-$ in different sites for individual samples, while mean NO_3^- content was 7.18 (*15 BB*) to 161.0 (*Padampur urban*) in different habitations (Table 7). The average NO_3^- levels in groundwater was

Table 8
Nitrate contamination in different sampling sites comparison with BIS and WHO standards.

Village	Range (mg l^{-1})	No. of samples showing higher range than standard limit	
		WHO standard (as NO_3-N) 11.0 mg l^{-1}	BIS standard (as NO_3-N) 22.6 mg l^{-1}
Fakirawali (*n* = 5)	39–76	5	5
Jaloki (*n* = 13)	57–82	13	13
Delwah (*n* = 5)	42–59	5	5
Channadham (*n* = 10)	7.10–47	7	7
Ratewala (*n* = 2)	78–79	2	2
Sawantsar (*n* = 2)	65–69	2	2
Padampur (*n* = 15)	18–162	15	10
Bera (*n* = 2)	58–59	2	2
Kikrawali (*n* = 4)	30–32	4	4
Kumharanwali (*n* = 3)	80–82	3	3
Khichia (*n* = 53)	59–60	3	3

60.8 mg l^{-1} at *Fakirawali*, 72.8 mg l^{-1} at *Jaloki*, 51.4 mg l^{-1} at *Delwah*, 31.6 mg l^{-1} at *Channadham*, 78.5 mg l^{-1} at *Ratewala*, 67.0 mg l^{-1} at *Sawantsar*, 59.3 mg l^{-1} at *Beran*, 31.0 mg l^{-1} at *Kikrawali*, 80.7 mg l^{-1} at *Kumharanwali*, 59.7 at *Kichiyan*, 73.4 mg l^{-1} at *Padampur rural* (Fig. 2). The average NO_3^- level for total sampling site was 60.6 ± 33.6 (SD) mg l^{-1}. Results thus indicated that the groundwater of this part of the State is severely polluted due to anthropogenic activities. Considering the health effect of NO_3^-, WHO [19] and Bureau of Indian Standards [20] have set a guideline for the maximum permissible levels, i.e. 10.2 mg l^{-1} (as nitrate-N) and 22.6 mg l^{-1} (as nitrate-N), respectively in drinking water. This study clearly indicated that water from most of the sampling sites was not fit for drinking purposes, if BIS maximum permissible level is taken as reference level. A drastic variation for groundwater NO_3^- within a village, e.g. *Channadham* (7.18–46.2 mg l^{-1}), *Padampur* (18.3–69.7 mg l^{-1}) was also recorded in this area. *Jaloki* village showed a higher range of nitrate (57.7–81.3 mg l^{-1}) which indicates a uniform nitrate distribution in aquifers of this region. Fig. 3 illustrates the patterns of isolines for different NO_3^- level over the study area. It clearly indicates that groundwater of some rural habitations of *Padampur* tehsil (*Padampur main*, *Ratewala*, *Kumharanwali* and *Jaloki*) as well as *Raisinghnagar* tehsil (*Sawantsar*) was severely polluted in terms of NO_3^- level. However, the situation was worst in some villages of *Raisinghnagar* and *Sri Ganganagar* (single village, i.e. *Channadham*) where all the water samples showed higher ranges of NO_3^- in groundwater. About 50% samples from *Padampur* tehsil showed the ranges of 55–77 mg l^{-1} (Fig. 2). A few samples from *Padampur* showed about 3.5–4.5 times higher nitrate level than the maximum permissible limit by BIS (Table 8). As compared to the earlier reports from the other parts of the country (Table 1) the NO_3^- level was relatively higher in this region. Sankararamakrishnan et al. [2] reported only 19% samples exceeding BIS limit at Kanpur district. Reddy et al. [5] reported that about 65% samples of groundwater unfit for drinking purposes from Anantapur district of Andhra Pradesh are due to very high NO_3^- contents.

Sulphate (SO_4^{2-}) is a naturally occurring ion in almost all kinds of water bodies. Moreover, its concentration more than 200 mg l^{-1} is objectionable for domestic purposes. At higher concentration SO_4^{2-} may cause gastro-intestinal irritation particularly when Mg^{2+} and Na^+ are also present in drinking water resources. In this study, sulphate (as SO_4^{2-}) concentration ranged from 28.2 to 664.8 mg l^{-1} for individual sample, while mean level of SO_4^{2-} was between 28.6 (*5 RB*) and 660.3 mg l^{-1} (*4 EEA*) in different villages (Table 7). The average SO_4^{2-} level in groundwater was 181.2 mg l^{-1} at *Fakirawali*, 456.7 mg l^{-1} at *Jaloki*, 399.5 mg l^{-1} at *Delwah*, 489.2 mg l^{-1} at *Channa dham*, 63.6 mg l^{-1} at *Ratewala*, 660.3 mg l^{-1} at *Sawantsar*, 142.2 mg l^{-1} at *Beran*, 192.9 mg l^{-1} at *Kikrawali*, 641.5 mg l^{-1} at *Kumharanwali*, 512.6 at *Kichiyan*, 130.0 mg l^{-1} at *Padampur rural* (Fig. 4). The average SO_4^{2-} level was 335.3 mg l^{-1} for this region that was relatively higher than prescribed limit by BIS (200 mg l^{-1}). The situation was more drastic at *6 RB*, *41 RB*, *3 DD*, *16 BB*, *4 EEA*, *Kikrawali* and *Kumharanwali* villages where mean SO_4^{2-} was 3.28, 3.23, 3.22, 3.19, 3.30, 3.21 and 2.57 times, respectively higher than the maximum permissible limit (Table 8). The isolines over the study area for SO_4^{2-} level (Fig. 5) clearly indicates that groundwater of *Khichia*, *Sawansar*, *Padampur main*, *Delwah*, *Kumharanwali* and *Channadham* is highly polluted in terms of SO_4^{2-} level.

3.3. Source of NO_3^- and SO_4^{2-} contamination in groundwater

The level of NO_3^- and SO_4^{2-} in groundwater of this part of the State clearly reflects the severe contaminations from anthropogenic resources. The following point and non-point sources should have contributed in groundwater contaminations NO_3^- and SO_4^{2-}: (1) fertilizer use: the fertilizer consumption rate is relatively higher in this part due to well-established farming system; (2) poor soil profile: this region constitutes the arid and semi-arid land of Indian Thar desert. The soil of this area is typically sandy or sandy clay (soil type: Site Rozems or Reverina type) with high coarse texture. Such soils have high water filtration rate and possibly contributing in nitrate leaching to underground waters; and (3) irrigation mechanisms: as per the agro-climatic classification this part of the State is categorized under irrigated north western plain. The area is rich in agricultural production on account of a well-developed system of canal irrigation. High irrigation rate may contribute in nitrate leaching in soils of this area. Wheat, cotton, rape and mustered are the major crop of this belt. More interestingly this region is considered as the highest wheat production belt of the State. Due to intensive wheat cropping system, the nitrogenous fertilizer consumption rate is comparatively higher in this region than other agro-zones of the State. The urea is the main nitrogenous fertilizer being used for crop culture in this part. It constitutes about 68.8–70% of the total N-fertilizer consumption in the district followed by di-ammonium phosphate, calcium ammonium nitrate and ammonium sulphate. These nitrogenous fertilizers converted to NO_3^- forms when applied to the soil. These NO_3^- forms are highly soluble and easily leachable on irrigation [21]. It is concluded that heavy fertilizer consumption, highly coarse soils, high irrigation rates, well-developed canal network, etc. are some factors responsible for NO_3^- leaching to groundwater of this region. A trend of spatial variations in groundwater nitrate level was observed among different study sites possibly due to the difference in fertilizer application rate, crop rotation, fertilizer utilization efficiency of sown crops, irrigation arte, soil texture, and local pedo-climatic variability.

This climatic variability may affect the nitrate leaching the soil profiles [8,22]. According to Bijay-Singh et al. [8] soils of several developing countries located in Southeast Asia, Africa and Latin America remain moist part of the year and dry for more than 90 days and these soils possess potential for groundwater contaminations through leaching of nitrates. Here the interesting fact is that wheat is harvested in early summer month in most part of the northern India and next sowing is done in mid summer; after 25–30 days of harvesting of previous crop. Summer is followed by rainy season (monsoon) up to the end of October. Such climatic variability may contributes in NO_3^- leaching, although Bijay-Singh et al. [8] claimed such mechanism responsible for groundwater contaminations in dry areas of Southeast Asia. Similarly Goss et al. [22] reviewed the impact of farming system management on

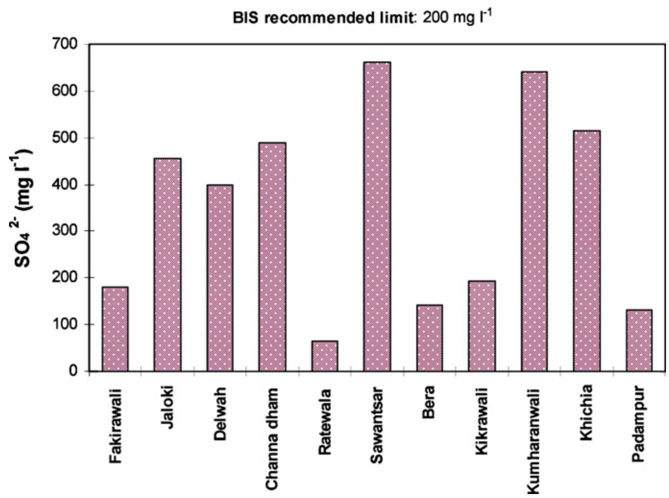

Fig. 4. Concentration of SO_4^{2-} in different villages of northern Rajasthan.

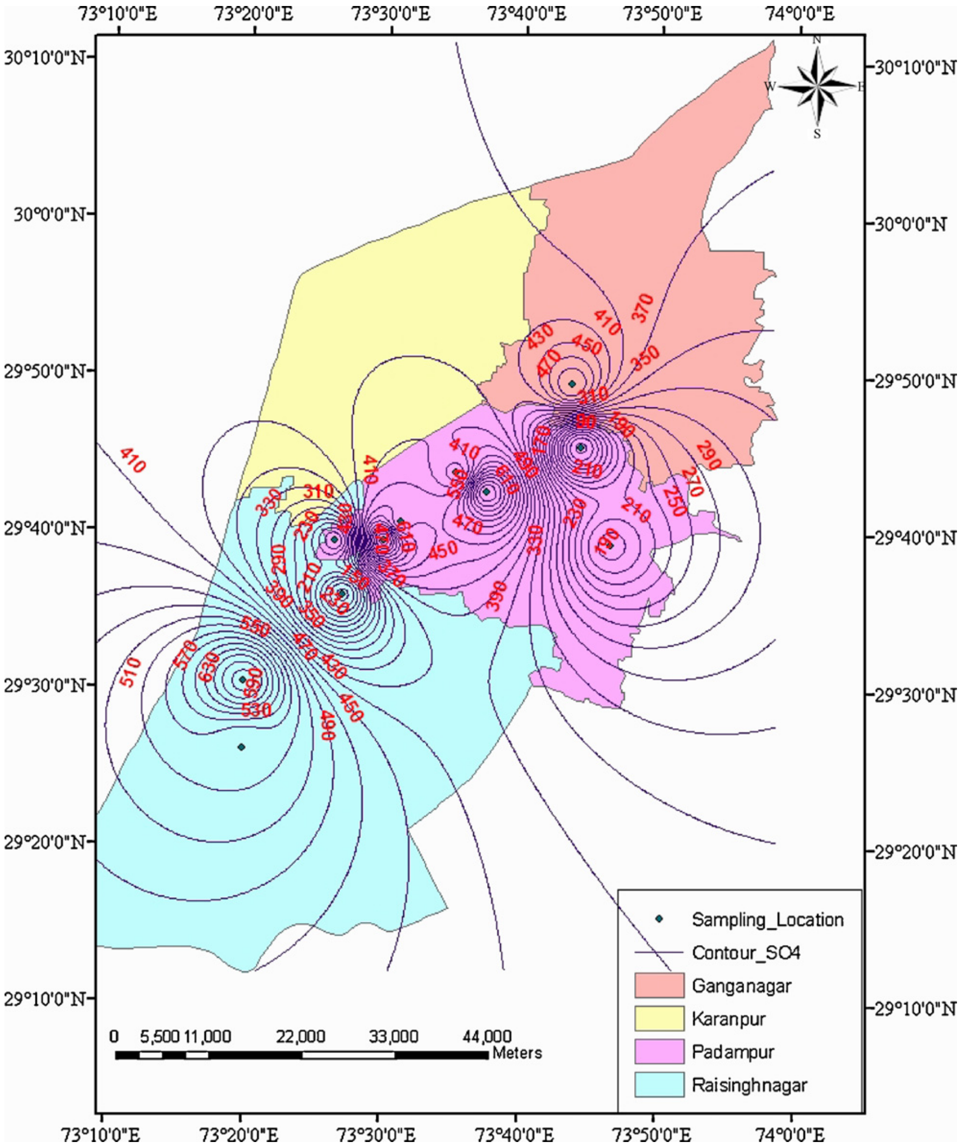

Fig. 5. SO_4^{2-} isolines over the study area.

NO_3^- leaching processes. They claimed that dry and moist soil duration may directly influence the nitrate leaching in arable lands. On the other hand the efficiency of surface crops in NO_3^- intake is also important for leaching of agrochemicals to soils. The literature suggested that wheat utilizes as much as half of the applied N [8,23]. Arora et al. [23] concluded that maize, which was grown on coarse-textured alluvial soil, utilized only 11–22% of the total N application in the experimental field. Thus unutilized fertilizer may contribute in NO_3^- enrichment of groundwater, although further detailed studies are still needed to establish the hypothesis.

Except to non-point sources some other point sources also contribute to groundwater NO_3^- pollution. Wakida and Lerner [24] reviewed the non-agriculture sources of groundwater nitrate and suggested that waste disposal network, animal wastes including livestock and human excreta, industry, river–aquifer interactions, house building, etc. are some important factors indirectly enriches nitrate in groundwater. The animal excreta could be an important source of groundwater NO_3^- in this area. Since cattle farming has been adopted as an important occupation by rural communities of this region. Therefore total livestock population in the district is comparatively higher, i.e. 1.62 million [16]. We calculated that such huge livestock population may produce a considerable amount of N (i.e. 42,055.07 tones year^{-1}). It has been observed that open dumping (in heaps) is the common practice to dispose the livestock excreta in this region. Due to weathering and open dumping, a considerable amount of soluble forms of nitrogen is leached into deep soil layers especially during wet months of the year. Other sources such as human excreta may contribute in nitrate leaching in this region. Pit latrines may enhance NO_3^- in groundwater, but further detailed study on groundwater microbiology is needed to support the hypothesis.

Data clearly indicates that SO_4^{2-} level was relatively higher at some sites. About 57.1% sampling sites showed more SO_4^{2-} than permissible limit by BIS (200 mg l^{-1}). Approximately one-third sampling sites showed groundwater SO_4^{2-} level >600 mg l^{-1}. There are a number of potential sources of S that contribute to groundwater sulphate which includes accession of inorganic sulphate via atmospheric deposition of marine sulphate aerosols, dissolution of sulphate minerals such as gypsum ($CaSO_4{\cdot}2H_2O$), oxidation of reduced sulphide minerals such as pyrite (FeS_2), or mineralization of organic S compounds with the soil zone [25]. Some anthropogenic sources such as fertilizers, animal excreta, and urban

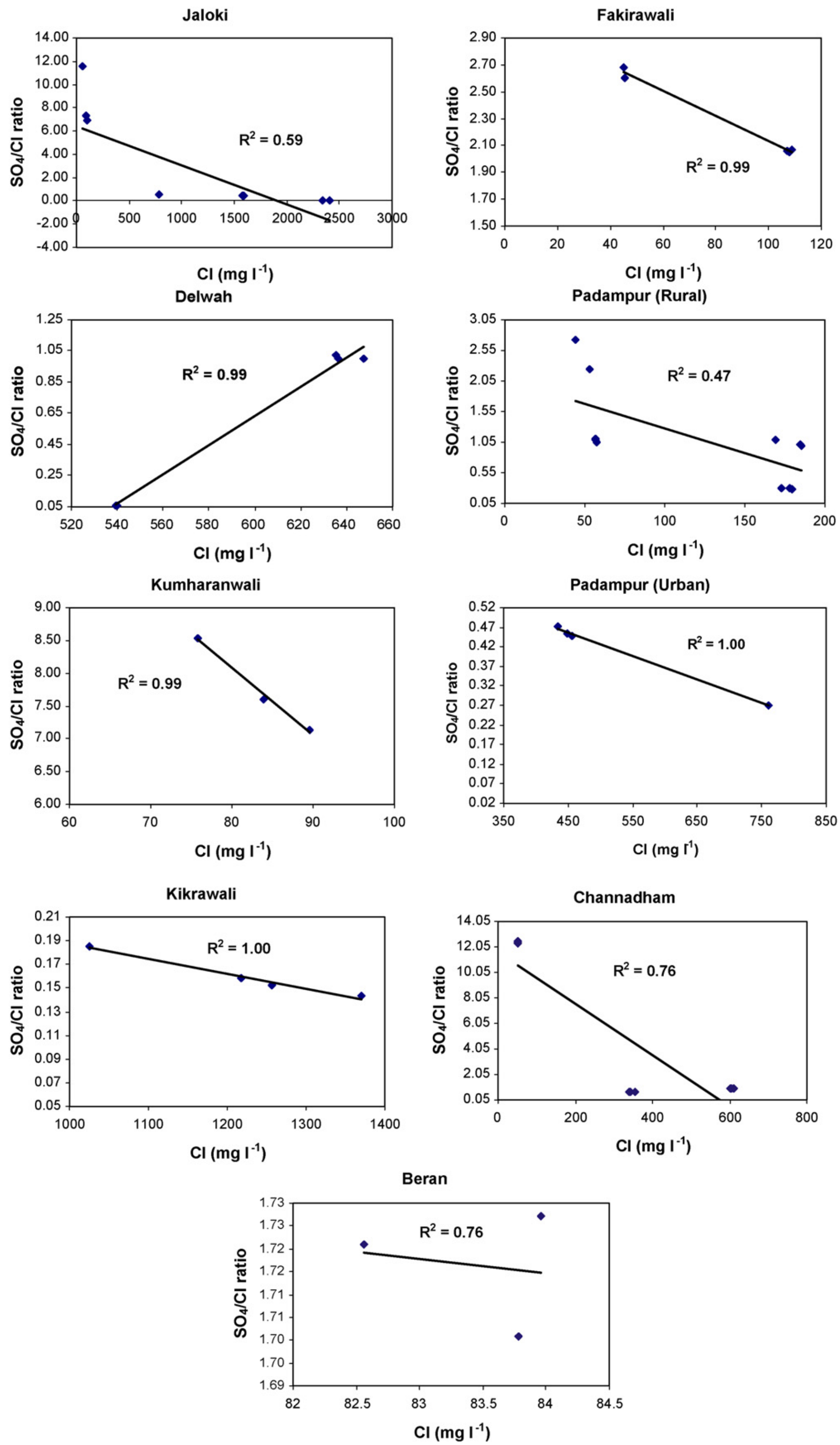

Fig. 6. Relationship between SO_4/Cl ratios and Cl contents for groundwater at different sites.

wastewater also contribute to enrichments of groundwater with sulphate. The dissolution of gypsum, fertilizer, animal excreta, and wastewater are probably the most important source in this part of the State. Krouse and Mayer [26] suggested that when sulphate concentrations in groundwater exceed 100 mg l^{-1}, lithogenic sources of sulphate such as evaporitic sulphate minerals or oxidation of pyrite or organic matter are often predominates. However, the variability among sampling sites for SO_4^{2-} level in groundwater was due to spatial distribution of gypsum in sub-soil layers and surface water recharging. Therefore, SO_4^{2-} level in groundwater can be explained partially by gypsum dissolution and partially by surface sources. Gypsum is widely distributed in subsurface layer of this part of the State and in general, its dissolution produces mineral SO_4^{2-} as:

$$CaSO_4{\cdot}2H_2O(S) \rightarrow Ca^{2+} + SO_4^{2-} + 2H_2O$$

Calcium is another important product of this reaction and it directly contributes to hardness (TH) of groundwater. We observed a good correlation between SO_4^{2-} level and TH at *Jaloki*, *Channadham* and *Fakirawali* villages, which indicates the possible source of SO_4^{2-} in these villages. Moreover, SO_4/Cl ratio may be a good indicator of SO_4^{2-} leaching to groundwater from surface sources. The variation in SO_4/Cl ratios indicates net changes to SO_4^{2-} due to biogeochemical processes in the groundwater prior to recharge, or within the aquifers. The surface precipitation, irrigation or evapotranspiration lead to Cl^- enrichments in groundwater, which untimely lowers the SO_4/Cl ratios of groundwater. The spatial variability in SO_4/Cl ratios was recorded (0.01–13.50) at different sampling sites of this area (Table 9). Although, at some sites we observed a good positive correlation between sulphate and chloride concentrations (Table 10), but at some site SO_4/Cl ratios tend to decrease with increasing chloride concentration (Fig. 6). In this context, we concluded that surface input was the major sources of SO_4^{2-} in areas where SO_4/Cl ratios was relatively lower. Cattle dung, fertilizers, wastewater disposals and even human excreta (through pit latrines) should be the main source of groundwater SO_4^{2-} especially in *Jaloki*, *Padampur*, *Kikrawali*, *Kumharanwali*, *Beran*, etc. The significant and negative correlation between SO_4/Cl ratio and Cl^- concentration clearly supports the proposed hypothesis (Fig. 6). The statistical analysis of groundwater clearly indicates the diverse sources of groundwater SO_4^{2-} contamination in different regions rather than a common source. In general, human excreta and sewerage are the major source of sulphate apart from chemical fertilizers. Possibly high SO_4^{2-} level in groundwater should be due to pit latrines at some sites. Dzwairo et al. [27] reported high ranges of SO_4^{2-} and NO_3^- in groundwater due to pit latrine system in rural habitations of Marondera district, in Zimbabwe. The lack of adequate financial resources for construction of safe onsite sanitation devices people still rally on pit latrines in rural areas of northern Rajasthan. The greater level of NO_3^- and SO_4^{2-} in groundwater supports this hypothesis, although further detailed microbial study of groundwater is required to trace the direct source of sulphate in groundwater.

Table 9
SO_4/Cl ratio of groundwater samples in different sampling sites.

Village/sub-division	No. of samples	SO_4/Cl ratio
2 RB	3	2.05–2.07
3 RB	2	2.61–2.68
5 RB	3	0.01
6 RB	3	7.30–11.6
40 RB	3	0.55
41 RB	4	0.41
2 DD	3	0.06–1.02
3 DD	3	1.0–1.02
14 BB	4	0.94–12.5
15 BB	3	0.65–0.67
16 BB	3	12.3–13.5
CC Head	2	1.18–1.26
4 EEA	2	12.9–12.65
17 BB	2	2.23–2.72
19 BB	3	1.08–1.10
20 BB	3	0.28–0.31
21 BB	3	0.99–1.09
23 BB	3	1.70–1.73
Kikrawali	4	0.14–0.19
Kumharanwali	3	7.14–8.54
1 JJ	3	1.06–1.08
Padampur (urban)	4	0.27–0.47
Total	65	

The relationship between major chemical parameters of groundwater of different sampling sites is presented in Table 10. A straight and very strong relationship was observed between NO_3^- and SO_4^{2-} level in groundwater at *Fakirawali* (r^2 = 0.99), *Delwah* (r^2 = 0.99), *Channadham* (r^2 = 0.84), *Beran* (r^2 = 0.88), *Kikrawali* (r^2 = 0.92) and *Kumharanwali* (r^2 = 0.99). Such high correlation indicates the contamination of groundwater with nitrate and sulphate form point sources, i.e. fertilizers, sewage and animal wastes. Majumder et al. [28] reported a non-significant relationship between NO_3^- and SO_4^{2-} while studying nitrate concentrations in groundwater of central-west region of Bangladesh. Similarly, Rao [12] reported a weak relationship between NO_3^- and SO_4^{2-} for groundwater at some sites of Andhra Pradesh. NO_3^- showed close affinities with Cl^- contents at *Fakirawali*, *Delwah*, *Kikrawali* and *Kumharanwali* as the r^2 was significantly higher. Similarly, SO_4^{2-} also exhibited good relationship with Cl^- for groundwater sample from *Fakirawali* (r^2 = 1.00), *Delwah* (r^2 = 0.99), *Kikrawali* (r^2 = 0.79), *Kumharanwali* (r^2 = 0.80) and *Padampur (urban)* (r^2 = 0.78), which suggested the contribution of animal and human excreta deposition on groundwater contamination. Reddy et al. [5] suggested non-point and point sources of nitrate in groundwater using correlation plots between different parameters of water. They summarized that fertilizers form one of the source of nitrate apart from the domestic sewerage and organic manure. The interrelationships between important pollution indicating parameters of water (Table 10) suggested that major sources of NO_3^- and SO_4^{2-} in groundwater of this region were nitrogenous fertilizer, sewerage, animal waste, organic manure, etc.

Table 10
Correlation between different parameters of groundwater of different sampling sites.

Parameter interaction	Regression coefficient (r^2)								
	Fakirawali	*Jaloki*	*Delwah*	*Channadham*	*Padampur (rural)*	*Beran*	*Kikrawali*	*Kumharanwali*	*Padampur (urban)*
$NO_3^- \times SO_4^{2-}$	0.99	0.04	0.99	0.84	−0.22	0.88	0.92	0.99	0.07
$NO_3 \times Cl^-$	0.99	0.01	0.99	0.08	0.005	0.09	0.96	−0.84	0.37
$SO_4^{2-} \times Cl^-$	1.00	0.47	0.99	0.012	0.09	0.48	0.79	−0.80	0.78
$EC \times NO_3$	0.90	0.01	0.99	0.002	0.14	0.80	0.002	1.00	−0.50
$pH \times NO_3$	−0.94	0.01	0.42	−0.88	0.05	0.79	0.02	−0.25	0.47
$SO_4^{2-} \times TH$	0.95	0.10	0.99	0.83	0.45	−0.85	0.01	−0.71	−0.72

4. Conclusions

The average NO_3^- and SO_4^{2-} level for this part of the State was 60.6 ± 33.6 (SD) and 28.6–660.3 mg l^{-1}, respectively. The nitrogenous fertilizers and organic wastes containing high NO_3^- (livestock excreta, sewerage, organic garbage, etc.) could be main sources of NO_3^- and SO_4^{2-} in groundwater in this region. A spatial variation in samples for different level of chemical parameters (NO_3^-, Cl^-, SO_4^{2-}, TH, etc.) was observed which indicates different sources of contaminations. This is the first comprehensive report on NO_3^- contamination in groundwater of this region; further detailed study is still required to trace out the potential contaminations source. Results thus indicate that this part of the State may be a danger zone for NO_3^- toxicity risks in humans.

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