Journal of Hazardous Materials 159 (2008) 536–543

Contents lists available at ScienceDirect

Journal of Hazardous Materials

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

An exploration of nitrate concentrations in groundwater aquifers of central-west region of Bangladesh

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ARTICLE INFO

Article history:
Received 22 November 2007
Received in revised form 19 February 2008
Accepted 19 February 2008
Available online 6 March 2008

Keywords:
Nitrate
Denitrification
Groundwater
Aquifer
Contamination

ABSTRACT

Groundwater and river water samples were collected from the study area to investigate the spatial distribution of nitrate (NO_3^-) in the central-west region of Bangladesh. The shallow and deep groundwater nitrate concentrations ranged from <0.10 to 75.12 and <0.10 to 40.78 mg/L, respectively. Major river water NO_3^- concentrations were ranged from 0.98 to 2.32 mg/L with an average of 1.8 mg/L. The average Cl^-/NO_3^- ratio (4.9) of major river water has been considered as reference point to delineate denitrification processes. The alluvial fan, alluvial, deltaic and coastal deposits shallow groundwater having $C1^-/NO_3^-$ values less than that of the average river water value (4.9), suggested denitrification processes within the aquifers. On the other hand, denitrification processes are insignificant in the Pleistocene terraces area aquifers related to relatively higher concentrations of nitrate. Iron pyrite has been found as insignificant effect on denitrification.

1. Introduction

Bangladesh is the most densely populated country in Southeast Asia. Most of the population of Bangladesh lives on agriculture. Since agriculture is widely practiced in Bangladesh, potential nitrate pollution may appear in this country in near future. Because of its large population, there has been a strongly increasing trend towards the growth of more crops to meet the demand. Nitrate (NO_3^-) is the main form of N in groundwater which is usually seen as an agricultural pollutant that often arises from the use of large amounts of nitrogenous fertilizers. Non-agricultural sources of nitrogen, such as septic systems and leaking municipal sewers, are generally less significant regionally but may affect groundwater locally [1].

Mineral fertilizers were introduced into Bangladesh during the early 1950s as a supplemental source of plant nutrients. The use of fertilizers increased rapidly from mid 1960s with the introduction of modern varieties and development of irrigation facilities. The annual urea consumption in Bangladesh is about two million tons per year was in 1980s. From 1989/1990 to 1996/1997, urea consumption grew rapidly registering an average growth rate of 7% per year [2]. Excessive N-fertilizer application is therefore very common, especially in intensive rice, wheat, beans and vegetable producing areas so the occurrence of nitrate pollution might be expected to groundwater.

Topsoil disturbance by cultivation increases the soil aeration and mixes with the available carbon and nitrogen sources in presence of soil micro-organisms. This process leads to nitrate accumulation in the soil due to mineralization and nitrification processes. With sufficient surface water infiltration, soluble nitrate can leach below the root zone to underlying groundwater. Because of its anionic form, nitrate is very mobile in groundwater and it moves in groundwater with no transformation and little or no retardation [3].

Food that is rich in nitrogen compounds can reduce the oxygen transport to blood which may cause serious consequences both for human and cattles. The uptake of high concentrations of nitrogen can cause problems in the thyroid gland and it can lead to shortages of vitamin A. In the stomachs and intestines of animals, nitrates can be converted into nitrosamines, a dangerously carcinogenic kind of substance [4]. According to the World Health Organization (WHO) and the European Community (EC), the maximum contaminant level (MCL) of nitrate is given to be 50 mg/L whereas European Community (EC) describe the guide level (GL) of nitrate as 25 mg/L [5,6].

Unfortunately, no significant research works have yet to be done to get a proximate view of nitrate pollution in Bangladesh. The purpose of this study was (1) to determine the spatial distribution of

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doi:10.1016/j.jhazmat.2008.02.110

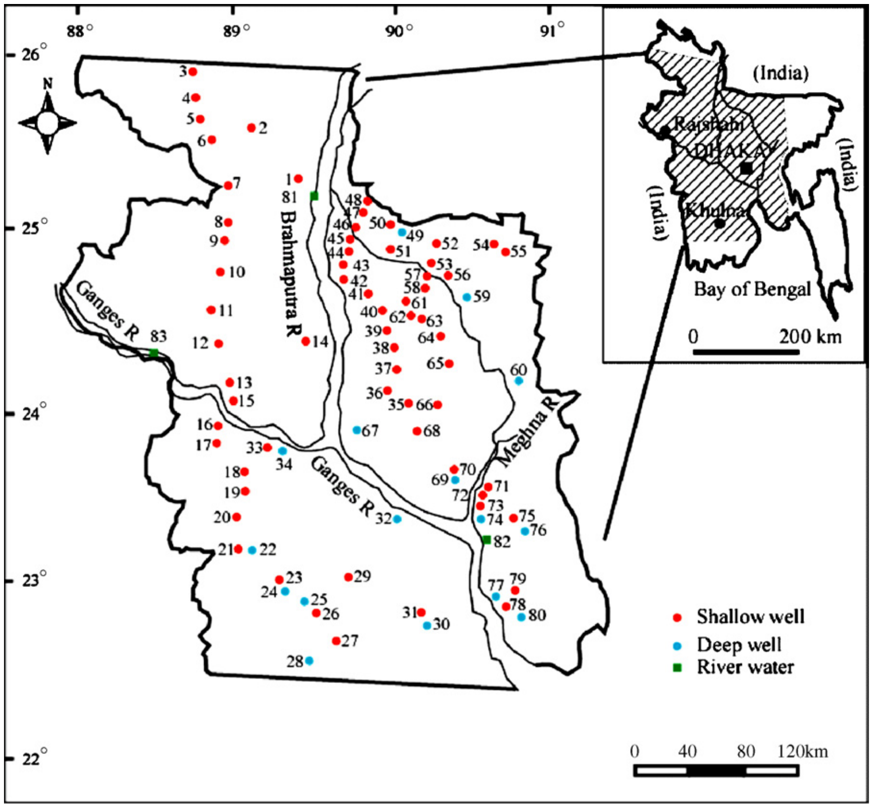

Fig. 1. Geographical map of the study area showing groundwater and river water sampling sites.

nitrate concentration, (2) to assess nitrate pollution, and (3) to evaluate the probable cause(s) of groundwater nitrate variability in the central-west region of Bangladesh based on early December 2005 sampling data.

2. Study area

2.1. Geomorphology and climate

The study area, located in the central-west region of Bangladesh between latitude 22°36′–26°00′N and longitude 89°00′–92°41′E, is a very important agricultural region of Bangladesh covering an area of about 90,000 km^2 (Fig. 1). This area forms a part of the sub-areal delta of the Ganges–Brahmaputra–Meghna (GBM) river system of Bangladesh. The study area consists of five major surface geologic units [7] mapped in the Holocene plain lands which includes alluvial fan deposits (AFD), alluvial deposits (AD), deltaic deposits (DD), Pleistocene terraces (PT) and coastal deposits (CD) (Fig. 2). This region is drained by the mighty rivers Ganges, Brahmaputra and the Meghna flowing towards the southeast where it discharges into the Bay of Bengal.

Bangladesh has a tropical monsoon climate with significant variations in rainfall and temperature throughout the country. There are four main seasons: the pre-monsoon (March–May) has the highest temperatures and experiences the maximum intensity of cyclonic storms, especially in May; the monsoon (June–September) when the bulk of rainfall occurs; the post-monsoon (October–November) which, like the pre-monsoon season, is marked by tropical cyclones on the coast; and the cool and sunny dry season (December–February). About 80% of the total rainfall occurs in the monsoon, and the average annual rainfall over the country is about 2320 mm. Precipitation varies from 1110 mm

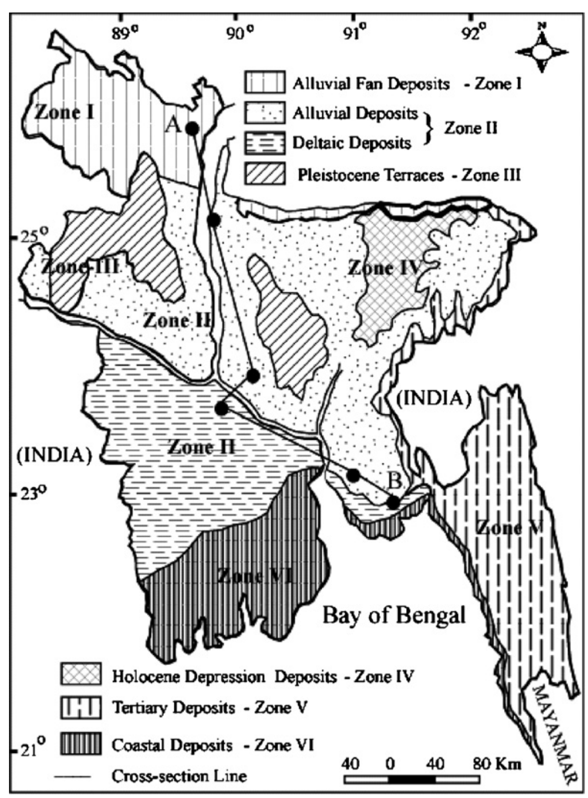

Fig. 2. Geo-hydrological map of Bangladesh.

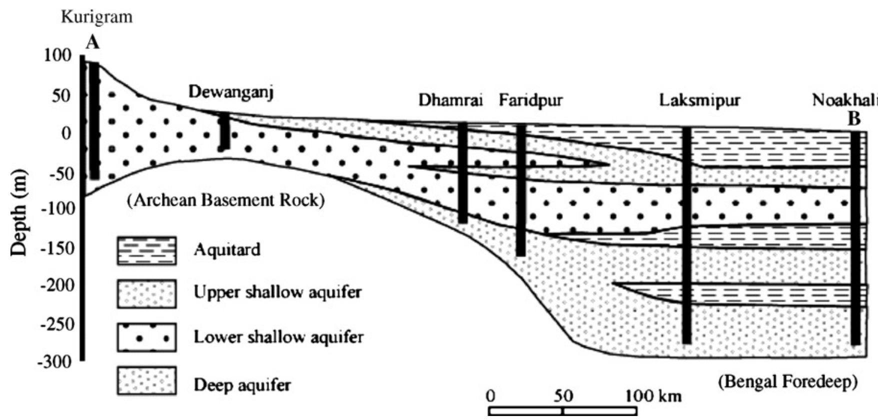

Fig. 3. Simplified hydrostratigraphic cross-section (along line A–B in Fig. 2) of the study area showing shallow and deep aquifers.

in the west to 5690 mm in the northeast. The mean annual temperature is about 25 °C, with extremes of 4 and 43 °C [8].

2.2. Hydrogeology and land use

The country can be broadly divided into six major hydrogeological units [9]. However, present study area consists of four major hydrological zones of the country which are: Zone I—alluvial fan deposits, Zone II—alluvial and deltaic deposits, Zone III—Pleistocene terraces and Zone IV—coastal deposits (Fig. 2). The study area can be classified into two major types aquifers: the shallow aquifers (<100 m depth) and deep aquifers (>100 m depth) (Fig. 3). The shallow aquifers mainly consist of Holocene alluvial and Plio-Pleistocene Dupi Tila Formation (Table 1). Except the Pleistocene terraces areas, the alluvial, deltaic and coastal aquifers mainly consist of fine sand with silts and clay lying all over the study area. The

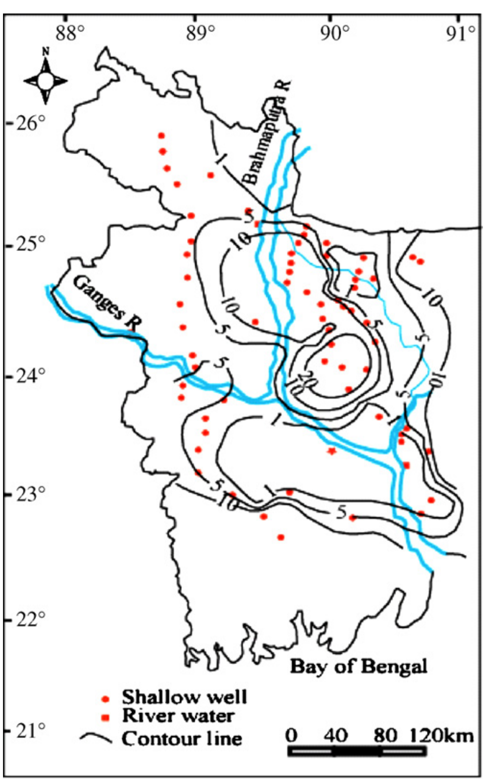

Fig. 4. Spatial distribution of shallow groundwater NO_3^- concentrations in the study area.

Dupi Tila aquifer is characterized by coarse sands with widespread gravels within a depth of 200 m overlain by a thick silty clay layer (Madhupur Clay Formation) of Pleistocene age. The deeper aquifers lying at depths greater than 100 m below the ground surface are separated from one or more overlying clay layers. In shallow aquifers groundwater occur at very shallow depths.

The aquifers are generally multi-layered varying from unconfined to leaky-confined in the shallow alluvial deposits and confined in the Dupi Tila and deeper alluvial deposits [10]. In the study area, groundwater levels lie within a few meters of the ground surface and fluctuate with the annual dry and wet season conditions. The aquifers are recharged during the monsoon season (July–September) when the area receives more than 80% of its annual precipitation (around 2000 mm/year). Huge amounts of annual floodwater standing on a large part of the country also contribute to the recharge process. Annual fluctuations in groundwater levels are controlled by the local hydrogeological conditions and withdrawal of groundwater for irrigation. In general, the seasonal groundwater fluctuations are relatively higher in the northwestern part of the country as compared to the deltaic deposit aquifers.

The low lands (alluvial, deltaic and coastal deposits) in the study area are mainly used for growing cereal crops like rice, jute and sugar cane, and the uplands (alluvial fan deposits and Pleistocene terraces) are cultivated for growing pulses, oilseeds, vegetables, etc. Groundwater is in common use for irrigation in the study area during critical crop periods (November–April) except where a surface water supply is available, such as in the southern part of the study area.

3. Methodology

Eighty groundwater samples were collected from existing domestic tube wells in early December 2005 (Fig. 1) covering the central-west region of Bangladesh. Three surface water samples were also collected from the major rivers namely the Brahmaputra, Meghna and the Ganges. The sampling sites were chosen arbitrarily. Each well was purged for few minutes prior to sampling by hand pump. The approximate depth of each well was noted from the

Table 1
Generalized stratigraphy of the study area (modified after Ahmed et al. [10])

Age	Formation	Lithology
Holocene	Alluvial	Grey clay, silt, fine sand with peat and gravels
Pleistocene	Madhupur Clay	Reddish brown fine to medium sand
Plio-Pleistocene	Dupi Tila	Yellowish brown fine to medium sand with gravels

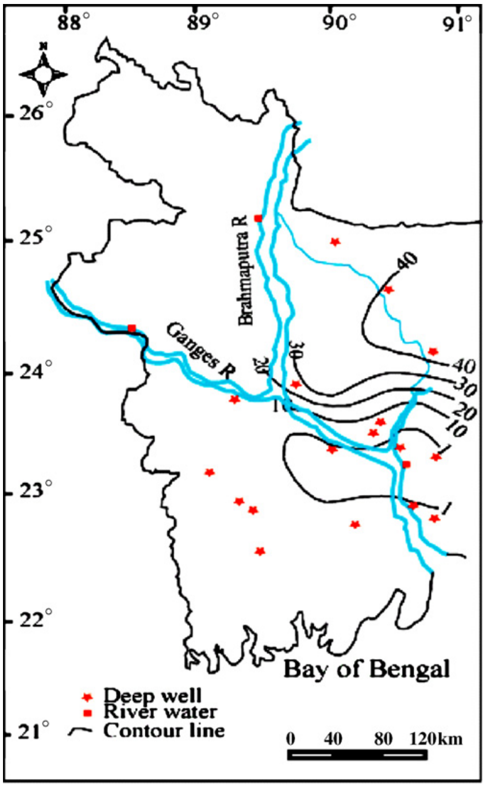

Fig. 5. Spatial distribution of deep groundwater NO_3^- concentrations in the study area.

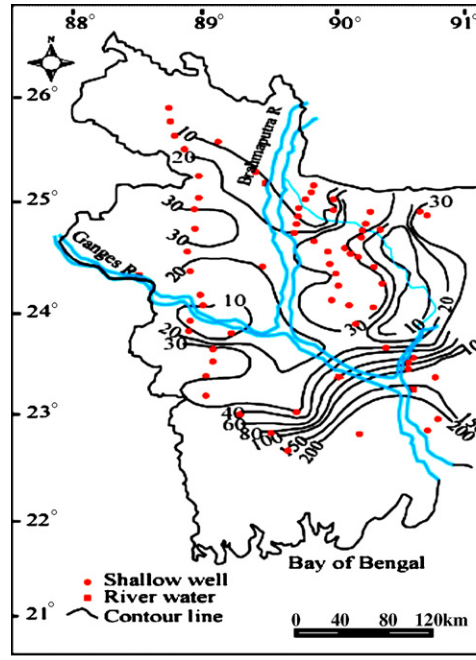

Fig. 6. Spatial distribution of shallow groundwater Cl^- concentrations in the study area.

record preserved by the well owners. The physio-chemical properties such as pH, temperature and electrical conductivity (EC) were measured on site in running water. The field pH of the samples was measured by using a HACH senSION1 portable pH meter and EC by using a HACH senSION5 portable EC meter. Sampling site geo-positions (latitude and longitude) were fixed by using handheld GARMIN 12XL GPS equipment. The samples for major anion analysis were filtered by 0.25 μm polycarbonate filters. In the laboratory Cl^-, SO_4^{2-}, NO_3^- and NH_4^+ were determined by an Ion Chromatograph (ICA-2000) measurement. Bi-carbonate alkalinity was determined by acid based titration.

4. Results and discussion

4.1. Spatial distribution of nitrate

Tables 2 and 3 show the overall picture of nitrate (as NO_3^-) concentrations of the sampling sites. The analysis of groundwater from the study area shows considerable variation in nitrate concentration. The shallow well nitrate concentrations were ranged from <0.12 to 75.12 mg/L. Among the samples reported in Table 2, 69% were found as less than 10 mg/L and only 7.6% exceeded the Bangladesh standard (50 mg/L) indicating that the shallow groundwater had not been severely polluted by nitrate in this agricultural system. NO_3^- concentrations of major river (Ganges, Brahmaputra and Meghna river) water ranged from 0.98 to 2.32 mg/L with an average of 1.8 mg/L. Table 3 shows that NO_3^- concentrations in deep wells were ranged as <0.25–55.23 mg/L with an average value of 4.7 mg/L (Fig. 4).

In the study area, nitrate concentrations show the variations in spatial distribution due to surface geology and agricultural activities. However, the isoline for nitrate concentration map (Fig. 5) for shallow groundwater shows the crowding of contours in the Pleistocene terraces areas. The samples collected from the Pleistocene terraces areas (Fig. 2) have relatively higher (average ~9 mg/L) concentrations of NO_3^- than that of the samples collected from the alluvial fan, alluvial, deltaic and coastal deposits aquifers (average ~7 mg/L). Similarly, few deep groundwater NO_3^- concentrations have also been found as higher concentrations in the Pleistocene terraces and alluvial deposits area (Fig. 6).

Generally, chloride is an inert tracer remaining conservative during passage through the unsaturated zone and recording the rainfall signature modified by evapotranspiration, or the records of human impacts through pollution [11]. The chloride concentrations for shallow groundwater (<100 m) ranged from 0.72 to 606.25 mg/L with an average value of 47 mg/L and deep groundwater chloride concentrations ranged from 0.88 to 491.28 mg/L. However, the patterns of both shallow and deep well chloride concentrations (Figs. 6 and 7) are very similar to that of nitrate. Shallow groundwater samples with a high concentration of $C1^-$ have much lower concentrations of NO_3^- indicating a source of $C1^-$ from non-fertilizer sources (e.g., contamination) and/or from a concentration of mineralization in the zone of capillary fringe caused by evaporation. These results are similar to those obtained by Pawar and Shaikh [12]. A few shallow well samples having elevated NO_3^- concentrations show a higher concentration of $C1^-$ indicating fertilizer as a possible source of nitrate.

The $C1^-/NO_3^-$ ratio for Pleistocene terraces shallow groundwater (Fig. 2) shows the variation from ~0.6 to 9. Considering the average Cl^-/NO_3^- ratio (4.9) of major river water as reference point and comparing it with the values of $C1^-/NO_3^-$ for the aforesaid area shallow wells, it has been estimated that most of the shallow wells exhibit $C1^-/NO_3^-$ values less than 4.9 suggesting a build-up of nitrate in groundwater. Surprisingly, the shallow Pleistocene terraces groundwater samples show low $C1^-/NO_3^-$ ratios confirming the validity of above argument. The relatively higher $C1^-/NO_3^-$ ratios for the shallow well samples collected from the alluvial fan, alluvial, deltaic and coastal region are the indicative of efficient removal of nitrate by denitrification and/or by nitrate reduction in presence of organic matter.

Table 2
Variation of concentrations of major anions and ammonia along with physical parameters for shallow wells

Depth (m)	Geologic unit	pH	EC (μS/cm)	HCO_3^- (mg/L)	NO_3^- (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	NH_4^+ (mg/L)	Cl^-/NO_3^-	SO_4^{2-}/Cl^-
45	FD	6.50	170	85	0.50	1.8	1.60	8.65	3.600	0.89
9	FD	6.35	191	80	2.09	3.43	8.80	4.23	1.641	2.57
12	FD	6.50	1440	65	0.16	6.94	5.59	5.36	42.577	0.81
24	FD	6.64	185	66	4.01	13.32	4.49	4.51	3.323	0.34
13	FD	6.63	210	100	1.52	12.29	1.59	5.42	8.107	0.13
40	FD	7.08	250	160	1.32	0.72	1.73	7.58	0.545	2.41
10	PT	6.05	250	65	4.17	18.12	9.89	1.02	4.345	0.55
20	PT	7.01	375	160	4.50	14.13	7.11	0.45	3.140	0.50
20	PT	7.01	200	102	4.80	22.45	2.57	0.67	4.677	0.11
16	PT	6.55	720	240	6.03	18.73	23.33	2.03	3.104	1.25
40	PT	7.13	336	230	6.94	30.14	0.27	1.12	4.341	0.01
23	PT	6.95	475	205	6.66	28.54	11.16	1.06	4.285	0.39
21	PT	6.85	470	200	5.12	30.61	15.95	1.58	5.979	0.52
23	PT	6.47	550	190	50.86	29.39	19.08	1.22	0.578	0.65
21	PT	6.19	225	70	6.12	28.36	3.38	2.36	4.634	0.12
50	PT	7.16	350	220	6.23	30.45	1.15	1.51	4.888	0.04
54	PT	7.09	354	225	5.01	45.06	1.15	1.86	8.994	0.03
52	PT	7.07	340	220	8.51	31.78	0.54	0.98	3.734	0.02
54	PT	7.00	278	180	11.23	32.48	2.88	0.86	2.892	0.09
36	PT	6.35	150	90	8.35	29.91	6.56	1.57	3.582	0.22
46	PT	6.90	230	135	6.89	33.37	6.94	1.54	4.843	0.21
14	PT	6.8	385	220	6.65	31.24	2.85	1.59	4.698	0.09
24	AD	6.64	273	150	0.97	7.87	0.53	6.38	8.113	0.07
26	AD	6.81	540	260	1.53	19	43.62	6.53	12.418	2.30
22	AD	6.51	810	261	3.31	22.59	19.44	9.63	6.825	0.86
27	AD	6.84	870	560	2.21	4.01	11.05	5.36	1.814	2.76
62	AD	6.90	470	231	0.12	11.4	0.45	10.25	95.000	0.04
26	AD	7.21	785	450	4.88	18.94	4.85	2.87	3.881	0.26
20	AD	6.63	855	180	75.12	109.64	67.32	6.51	1.460	0.61
20	AD	6.52	610	210	0.94	84.01	24.6	16.21	89.372	0.29
26	AD	6.37	380	210	60.12	12.38	2.55	0.41	0.206	0.21
40	AD	6.83	280	175	0.76	2.29	1.24	2.46	3.009	0.54
30	AD	6.74	1290	40	9.87	20.77	7.20	3.46	2.104	0.35
22	AD	7.00	290	150	0.85	12.61	5.68	4.23	14.835	0.45
22	AD	6.88	195	114	0.85	1.34	1.53	5.21	1.576	1.14
15	AD	6.64	370	130	2.50	30.51	11.81	8.54	12.204	0.39
14	AD	6.36	375	70	2.97	41.71	18.38	3.51	14.044	0.44
15	AD	6.77	621	135	2.08	108.64	22.75	7.24	52.231	0.21
45	AD	7.25	336	210	1.16	6.11	1.77	8.45	5.267	0.29
9	AD	6.07	250	110	4.98	14.44	14.11	2.21	2.900	0.98
80	AD	7.24	390	195	15.33	26.06	15.33	1.57	1.700	0.59
50	AD	7.00	380	140	14.20	25.64	13.18	0.81	1.806	0.51
12	AD	6.53	600	145	12.90	107.11	17.81	1.24	8.303	0.17
51	AD	7.01	505	330	2.25	1.52	4.50	5.21	0.676	2.96
58	AD	7.21	342	210	1.99	5.42	4.50	4.59	2.724	0.83
25	AD	6.96	960	230	6.52	0.66	4.72	1.23	0.100	7.21
16	AD	6.93	660	328	4.47	40.43	16.18	2.39	9.045	0.40
90	AD	6.97	725	270	1.13	98.77	0.66	7.45	87.407	0.01
50	AD	6.70	1350	142	1.31	2.70	10.43	5.68	2.061	3.86
25	AD	7.05	1668	612	39.22	235.31	23.32	8.97	6.000	0.10
21	DD	7.00	820	515	4.98	6.72	4.80	2.81	1.349	0.71
38	DD	6.61	850	560	8.92	0.81	4.99	1.81	0.091	6.16
25	DD	6.84	800	518	1.55	4.25	9.27	4.35	2.742	2.18
20	DD	6.68	1070	540	1.85	91.22	38.18	4.96	49.308	0.42
46	DD	6.44	868	475	1.38	44.3	1.75	4.21	32.101	0.04
49	DD	7.21	560	358	2.34	10.03	0.34	5.21	4.286	0.03
60	DD	7.28	1010	615	1.25	47.16	1.60	5.01	37.728	0.03
34	DD	6.90	610	1180	67.02	54.58	32.85	4.25	0.814	0.60
12	DD	7.16	768	400	6.60	36.18	14.08	2.37	5.482	0.39
19	DD	6.80	864	565	0.35	16.4	3.80	1.53	46.857	0.23
9	DD	7.36	415	277	0.45	5.4	0.20	1.01	12.000	0.04
12	DD	7.40	435	322	0.3	6.3	0.30	1.17	21.000	0.05
58	CD	7.20	3010	520	20.53	19.79	47.66	1.12	0.964	2.41
19	CD	8.05	3355	925	12.44	606.25	0.79	8.46	48.734	0.00

The well locations and hydrologic units are shown in Figs. 1 and 2.
Surface geologic units; FD: fan deposits, PT: Pleistocene terraces, AD: alluvial deposits, DD: deltaic deposits, CD: coastal deposits.

4.2. *Relationship between NO_3^-, HCO_3^-, SO_4^{2-} and NH_4^+*

Fig. 8 shows the relation between nitrate concentration and well depth in the study region. The sites mentioned here is the highest cultivated area of Bangladesh. The farmers usually use excessive amount of N-enriched fertilizers and organic manure for increased food production. Logically, year round cultivation may cause high-level nitrate concentration in shallow groundwater around sites mentioned here. However, surprisingly it was found that almost all shallow wells (<100 m depths) of alluvial fan, alluvial, deltaic and

Table 3
Variation of concentrations of major anions and ammonia along with physical parameters for deep wells and river water

Depth (m)	Geologic unit	pH	EC (μS/cm)	HCO_3^- (mg/L)	NO_3^- (mg/L)	Cl^- (mg/L)	SO_4^{2-} (mg/L)	NH_4^+ (mg/L)	Cl^-/NO_3^-	SO_4^{2-}/Cl^-
132	DD	7.28	620	390	0.85	12.76	0.85	0.31	15.08	0.066
280	DD	7.65	1640	405	4.94	371.00	11.17	0.53	75.10	0.030
270	DD	8.84	845	445	3.18	54.77	3.40	0.48	17.22	0.062
265	CD	8.20	2670	440	1.55	491.28	0.25	3.26	316.95	0.001
301	CD	7.22	1350	340	5.28	118.56	0.41	2.46	22.45	0.003
230	DD	6.75	1310	325	0.21	261.5	21.30	1.25	1245.24	0.081
200	DD	6.90	995	654	0.30	194	0.20	1.36	646.67	0.001
170	AD	7.04	260	150	1.19	0.88	1.26	4.65	0.74	1.426
205	AD	7.03	340	215	55.23	3.81	0.36	15.32	0.07	0.094
280	AD	7.01	332	150	52.36	5.32	1.65	16.81	0.10	0.310
210	AD	6.96	810	380	40.78	115.32	0.91	14.26	2.83	0.008
220	AD	7.21	970	233	1.37	213.00	4.63	2.58	155.47	0.022
210	AD	6.50	1940	70	2.70	597.00	26.6	1.28	221.11	0.045
210	AD	6.22	2985	615	0.80	233	23.58	2.39	291.25	0.101
374	DD	6.99	391	253	0.4	3.5	0.50	1.42	8.75	0.143
240	DD	6.74	610	215	0.25	125	2.70	1.02	500.00	0.022
0	River water	6.95	200	122	2.1	2.03	12.21	ND	0.967	6.01
0	River water	8.3	230	120	0.98	10.52	14.98	ND	10.735	1.42
0	River water	7.72	420	250	2.32	7.02	12.74	ND	3.026	1.81

Well locations and hydrologic units are shown in Figs. 1 and 2.
Surface geologic units; AD: alluvial deposits, DD: deltaic deposits, CD: coastal deposits.

coastal deposits areas (Fig. 2) had shown low nitrate concentrations (<20 mg/L) and only four samples exceeded 50 mg/L (Fig. 8). Low nitrate concentrations in the shallow groundwater aquifers of alluvial fan, alluvial, deltaic and coastal deposits aquifers (Fig. 2) may be due to denitrification process resulting from chemically or biologically mediated reduction of nitrate to N_2 in presence of organic carbon and denitrifying bacteria. The Pleistocene terraces area shallow groundwaters showed relatively higher nitrate concentrations than that of shallow groundwaters from the fan, alluvial, deltaic and coastal groundwaters.

Starr and Gillham [13] reported that the presence of nitrate, labile organic carbon, denitrifying bacteria and a reducing environment are suitable for denitrification. Denitrification is expected to occur only in the zones that are devoid of dissolved oxygen and rich in denitrifying bacteria [14]. In this study, most of the shallow samples collected from the alluvial fan, alluvial, deltaic and coastal deposits are characterized by high HCO_3^- concentration. The shallow groundwaters are also characterized by low dissolved O_2 [15]. Such a bicarbonate and oxygen scarcity environment should generate a strongly reducing environment within the shallow aquifers. Above all, Bhattacharya et al. [16] noticed that the Holocene alluvial sediments of Bengal delta are rich in organic matter and the aquifers are mostly reducing in nature [17], which can create ambient conditions for the growth of aerobic bacteria [18].

Nitrate reduction by organic carbon leading to high concentration of HCO_3^- can be expressed as

$$4NO_3^- + 5C + 2H_2O \rightarrow 2N_2 + 4HCO_3^- + CO_2 \quad (1)$$

In this study, shallow groundwater nitrate concentrations of fan, alluvial, deltaic and deltaic deposits areas decrease with an increase of HCO_3^- concentrations (Fig. 9) that is a clear evidence of denitrification within the above-mentioned aquifers. Deep groundwaters also show similar decreasing trend of NO_3^- that is also an evidence of denitrification. However, the Pleistocene terraces shallow groundwaters NO_3^- concentrations remain more or less stable (Fig. 9) with respect to HCO_3^- concentrations. It is a clear indication of low or absence of denitrification within the Pleistocene terraces aquifers.

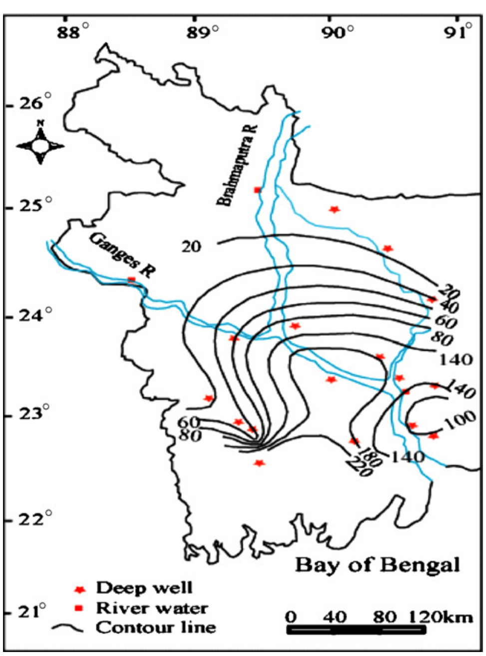

Fig. 7. Spatial distribution of deep groundwater Cl^- concentrations in the study area.

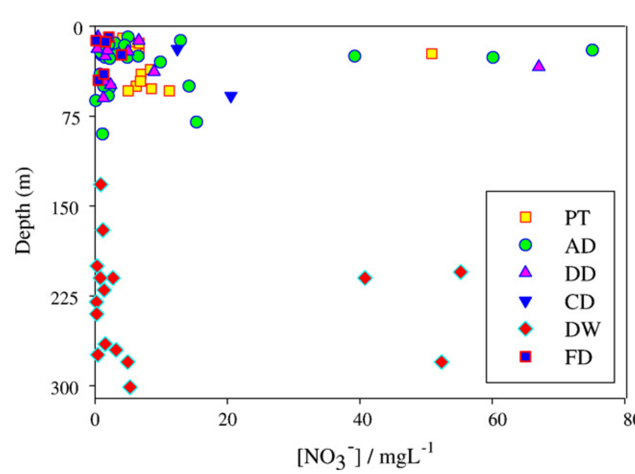

Fig. 8. Dependence of NO_3^- concentrations as a function of depth. PT: Pleistocene terraces, AD: alluvial deposits, DD: deltaic deposits, CD: coastal deposits, FD: fan deposits, DW: deep well.

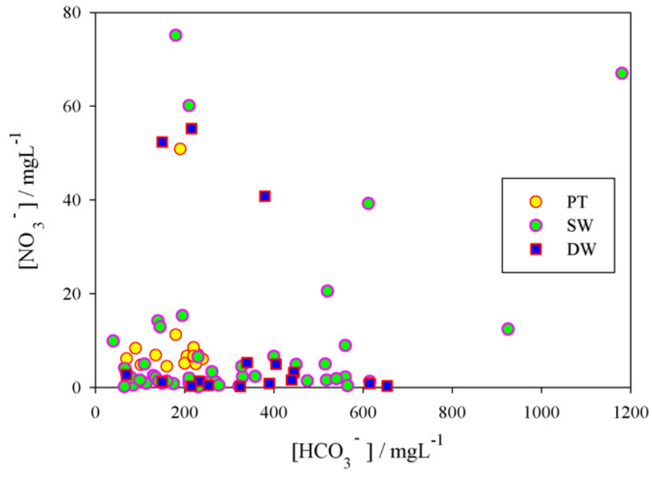

Fig. 9. Relationship between HCO_3^- and NO_3^- concentrations. PT: Pleistocene terraces, SW: shallow well, DW: deep well.

Denitrification is expected to occur in presence of denitrifying bacteria and can be summarized as [19];

$$4NO_3^- + 5CH_2O \rightarrow 2N_2\,(gas) + 5HCO_3^- \quad (2)$$

$$NO_3^- + H_2O + CH_2O \rightarrow NH_4^+ + 2HCO_3^- \quad (3)$$

Fig. 10 shows the relation between NO_3^- and NH_4^+ concentrations. The presence of NH_4^+ (ranging from 0.41 to 14.45 mg/L) and high HCO_3^- concentrations in shallow groundwaters (of fan, alluvial, deltaic and coastal deposits aquifers) suggest the possible bacterial denitrification as stated above. Microbial degradation of organic matter in those aquifers caused the reduction of NO_3^-. The increasing concentrations of NH_4^+ (up to 13.2 mg/L) in shallow groundwater were also observed by Bhattacharya [20]. Lower NH_4^+ concentrations (Fig. 10) were observed in groundwaters in Pleistocene terraces areas (Dupi Tila aquifers) compared to shallow aquifers of the study area which may be due to insignificant denitrification (both chemical and bacterial) processes in the Dupi Tila aquifers in the absence of sufficient organic carbon and denitrifying bacteria. According to BGS [21] report, Dupi Tila aquifers (Table 2) were thoroughly oxidized during its geological evolution which resulted a decrease in organic matter concentration in the aquifers. In addition, Ueno [22] found that bacteria may consume organic matters more easily from younger sediments than that

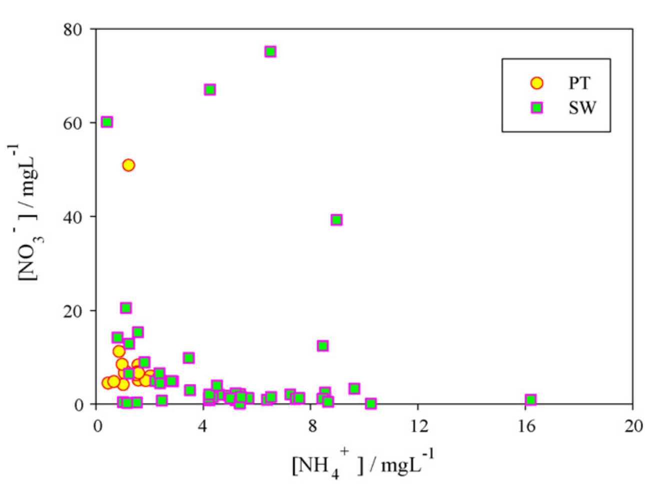

Fig. 10. Dependency of NH_4^+ on NO_3^- concentrations. PT: Pleistocene terraces, SW: shallow well.

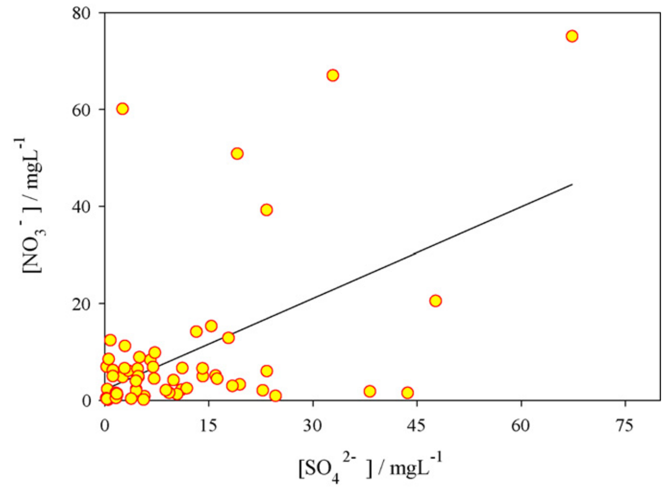

Fig. 11. Relationship between SO_4^{2-} with NO_3^- concentrations for shallow wells. Number of wells: 64, r^2: 0.27.

of older sediments. Above observation also validates that the low and/or absence of denitrification process within the older Dupi Tila aquifers which may allow a rise of relatively higher concentrations of NO_3^- in that aquifers. High nitrate concentration (>40 mg/L) was also observed in alluvial deposits deep wells. The high concentrations of NO_3^- in alluvial deposits deep wells may be due to excessive use of nitrogen fertilizers and animal waste leading to the percolation of NO_3^- into the highly transmissive (2325 $m^2\,d^{-1}$) [21] coarse grained deeper aquifer materials. This NO_3^- accumulation may be a possible evidence of nitrate pollution in deeper alluvial deposits aquifers in near future.

Denitrification in presence of pyrite is also possible if generation of high SO_4^{2-} concentration occurs according to the following scheme

$$14NO_3^- + 5FeS_2 + 4H^+ \rightarrow 7N_2 + 10SO_4^{2-} + 5Fe^{2+} + 2H_2O \quad (4)$$

Most of the shallow groundwater shows low SO_4^{2-} concentrations having no straight relationship ($r^2 = 0.27$) with SO_4^{2-} and NO_3^- (Fig. 11). In an addition, Ahmed et al. [10] found an insignificant amount of pyrite in shallow aquifer sediments of Bengal Delta that might not be responsible for nitrate reduction. Therefore, our observation exhorts that denitrification in presence of pyrite is not dominant within aquifers of the study area. In addition, Galy and Frace-Lanord [23] reported that denitrification in presence pyrite is related to high SO_4^{2-}/Cl^- ratio. In the present study, the groundwater samples show low SO_4^{2-}/Cl^- ratio (Tables 2 and 3) which is not favorable for nitrate reduction in presence of pyrite.

5. Conclusions

The results suggest that the NO_3^- concentrations of Pleistocene terraces groundwater aquifers are relatively higher than the alluvial fan, alluvial, deltaic and coastal deposits aquifer groundwater. Most of the deep groundwaters have shown insignificant concentrations of NO_3^- having a few high concentrations in some pocket areas. The high NO_3^- concentrations in alluvial deposits deep groundwater may be due to infiltration of NO_3^- into the deeper aquifers through the highly transmissive (2325 $m^2\,d^{-1}$) coarse-grained alluvial materials. Thus, the proper installation of deep wells and careful land-use planning may alleviate future contamination of deep groundwater aquifers. The relatively higher $C1^-/NO_3^-$ ratios for the shallow well samples collected from the alluvial fan, alluvial, deltaic and coastal region are the indicative of efficient removal of nitrate by denitrification and/or by nitrate reduction in presence of organic matter. Nitrate reduction in presence of organic carbon and denitrifying bacteria are dominant in the younger alluvial fan, alluvial, deltaic and coastal deposits aquifers. However, denitrification is not prominent in the Pleistocene terraces area aquifers, which may lead to nitrate pollution in Dupi Tila aquifers in near future.

Acknowledgement

The authors ruminate for the earnest co-operation of local peoples during sampling.

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