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An investigation of nitrate and iron concentrations and their relationship in shallow groundwater systems of Kathmandu

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ABSTRACT

Groundwater is vulnerable to contamination by chemicals, including nitrate, which can pass through soil to the shallow groundwater systems. Nitrate-N is commonly used as an environmental indicator to trace the impact of anthropogenic activities on groundwater. An understanding of the fate of nitrate in groundwater is vital for managing risks associated with nitrate pollution to safeguard groundwater supplies. Hence, groundwater samples were collected in ninety wells of shallow groundwater systems of Kathmandu, Nepal including shallow tube wells, dug wells and stone spouts (locally called *Dhunge Dharas*). The samples were analyzed for nitrate-nitrogen (NO_3-N), iron (Fe) along with pH and temperature. The nitrate-nitrogen and iron (Fe) concentrations ranged from 0.0 to 26.4 and 0.0 to 5.24 mg/L, respectively. An understanding of the relationship between nitrate and iron in groundwater is crucial to explore the mechanism of natural denitrification and its ability to reduce nitrate concentrations. In gene ral, elevated nitrate concentrations were not found in sampled groundwater sources in study area where elevated iron concentrations were common. The observed negative correlations between nitrate and iron suggest that the nitrate in shallow groundwater is being consumed. Moreover, this study highlights the current status and trends of the nitrate and iron in shallow groundwater systems and their relationship to water depth and well types.

Keywords: Nitrate-N; Denitrification; Ferrous iron; Shallow groundwater; Nepal

1. Introduction

Groundwater is a major natural resource for drinking purposes in urban area of many developing countries. More than 50% of the water supply is derived from groundwater sources in Kathmandu, the main urban centre of Nepal [1, 2]. Because of the inadequate and intermittent piped water supply, people use a variety of groundwater sources, including dug wells, tube wells and stone spouts (locally called *Dhuge Dharas*) [3]. Because of the high installation cost of deep aquifer tube wells, most of the people use either tube wells or dug wells from shallow aquifer to extract water to fulfill the demand. In addition, stone spouts are one of the alternatives to the municipal piped water supply in the Kathmandu. *Dhunge Dharas* are historic and revered sources that are generally more trusted for drinking water sources for hundreds of years. In urban settings in Kathmandu, Lalitpur and Bhaktapur, they are usually located in low-lying areas and are excavated rectilinear brick-lined pits that tap the shallow groundwater system to a spout or series of spouts. *Dhunge Dharas* are located throughout the Valley, both in dense urban and village settings. Throughout Kathmandu, groundwater

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is vulnerable to contamination by chemicals, including nitrate-N, which can pass through soil to the shallow groundwater systems. Nitrate is commonly used as an environmental indicator to trace the impact of anthropogenic activities on groundwater. Hence, an understanding of the fate of nitrate in groundwater has become a major concern of planners, decision makers and water managers for managing risks associated with nitrate pollution to safeguard groundwater supplies in recent years.

Several researchers have studied the ground water quality of Kathmandu. Khadka et al. [3] examined groundwater quality in stone spouts and springs that revealed widespread sewage contamination based on indicator bacteria, pH, iron and ammonia. Chettri and Smith [4] sampled for few wells in shallow aquifers of urban area of Kathmandu and reported 42% of the shallow tube wells had nitrate-N levels above the old World Health Organization (WHO) limit of 10 mg/L. In 1995, a joint program between the Australian Geological Survey Organization (AGSO) and the Ground Water Resources Development Board (GWRDB) of Nepal conducted the most extensive evaluation to date of the groundwater quality in the Kathmandu Valley [1]. Dug wells and shallow and deep-aquifer tube wells (but no stone spouts) were sampled both before and during the monsoon season and the water was analyzed for 30 chemical and microbial parameters, including heavy metals. Jha et al. [1] reported that the shallow aquifer was extensively polluted by sewage with the highest levels of fecal contamination observed in the populated urban centers of Kathmandu. Similarly, nitrate contamination of groundwater was studied by the Environment and Public Health Organization (ENPHO) in Kathmandu valley who found that 24% of the total stone spouts and 28% of the total dug wells violated the WHO limit [5]. Further, in 2005, a joint program between Japan International Cooperation Organization (JICA) and ENPHO, studied water quality of deep and shallow aquifer wells in Kathmandu and Lalitpur municipality areas [6]. They reported that only 22% of the dug wells and 12.5% of shallow tube wells violated the WHO limit. Recently, Warner et al. [7] reported nitrate contamination most likely from both sewage and agriculture, was common in the shallow aquifer but at concentrations only slightly higher than WHO guidelines in 11% of the water sources sampled. However, 64% of sources were at or above 0.3 mg/L of WHO limit of iron contamination [7]. Possible lower value of nitrate might have been due to the sampled sources include municipal taps and deep aquifer tube wells along with shallow groundwater sources.

All the previous studies highlight the severe groundwater quality problems in Kathmandu, which reveals a need for regular monitoring. However the relationship between nitrate-N and ferrous iron to explore the mechanism of natural denitrification and its ability to reduce elevated nitrate concentrations have not been investigated. In this paper, we studied the current status of nitrate and ferrous iron in shallow groundwater aquifer of Kathmandu to explore the possible nitrate attenuation by natural denitrification. Moreover, their relationship to depth to water table was investigated. This study compared nitrate and ferrous iron concentration levels of groundwater from different sources of shallow groundwater systems such as tube wells, dug wells and stone spouts. For this, groundwater samples were collected and analyzed for nitrate and ferrous iron from ninety sources of shallow groundwater systems of Kathmandu, Nepal including shallow tube wells, dug wells and stone spouts.

Generally speaking, the primary pathway for removal of nitrate from groundwater is denitrification, wherein nitrogen serves as the terminal electron acceptor following the general reaction [8]:

$$2NO_3^- + 12H^+ + 10e^- \rightarrow N_2 + 6H_2O \tag{1}$$

Alternatively, other reduced species such as ferrous iron can serve as the electron donor in the autotrophic denitrification process [9]:

$$10Fe^{2+} + 2NO_3 + 14H_2O \rightarrow N_2 + 10FeOOH + 18H^+ \tag{2}$$

It is significant that in all these above reactions, nitrate is converted from a potentially harmful form to a benign form as nitrogen gas [8].

2. Materials and methods

2.1. Study area

Kathmandu Valley is the main urban center of Nepal, which includes five of the fifty eight municipalities of country including three major cities; Kathmandu, Lalitpur and Bhaktapur. The current population of Kathmandu Valley is estimated about 3 millions, which is about 30% of the total urban population. The Kathmandu Valley, shown in Fig. 1, is a roughly circular basin with a diameter of about 25 km that consists of gentle hills and flat lands at elevations of 1300–1400 m with surrounding hills rise to more than 2000 m. Average annual precipitation in the Kathmandu is around 1400 mm, about 80% of which falls in the monsoon period during June and July. Within the Kathmandu, municipal and other water supplies depend on monsoon rains and the stream and groundwater systems fed by this precipitation.

Generally, the groundwater aquifers in Kathmandu Valley can be divided into shallow and deep systems however, this study basically focus to investigate the relationship between nitrate and iron in shallow groundwater systems of depth less than 20 m. Recharge to the shallow groundwater aquifers occurs mostly along the basin margins, directly from precipitation and by supply from many small rivers along with agriculture field. The shallow aquifer of the valley is characterized by a high recharge rate. The lowest recharge was associated with urban land while the dominant part of the study area had more than 254 mm/year [10].

2.2. Water sampling and analysis

Survey was made for groundwater sampling from the shallow aquifer of Kathmandu during September and October 2008, immediately after the monsoon season. Water from a total ninety sources was tested. Sampling locations are shown in Fig. 1, which included twenty three shallow tube wells, nine stone spouts and fifty-eight dug wells. Water depth of all types of sources was measured prior to water sampling. Horizontal and vertical location of sampling points was measured by Global Positioning System (GPS). Total depth and depth to water from ground level in the dug wells was measured directly, whereas depth for all tube wells was based on user provided information. For stone spouts, the total depth was estimated based on the distance of the tap below the ground surface. Based on the median values, stone spouts were the shallowest, followed by dug wells, and shallow tube wells were the deepest. At each sampling site, a single water sample was collected and stored in a 1000 ml low-density polyethylene (LDPE) bottle with zero head space for analysis.

The groundwater samples were also analyzed for temperature and pH, which were examined at the time of water sample collection by using a portable pH meter. Analyses for all samples collected in a day were performed at the end of the day using a DR-890 HACH portable, microprocessor-controlled colorimeter. Nitrate and ferrous iron were examined using the specified colorimetric analyses for each constituent. All analyses were performed using the pre-programmed calibration curves for each contaminant [11]. In addition, 10 water samples were analyzed 2–3 times to estimate reproducibility.

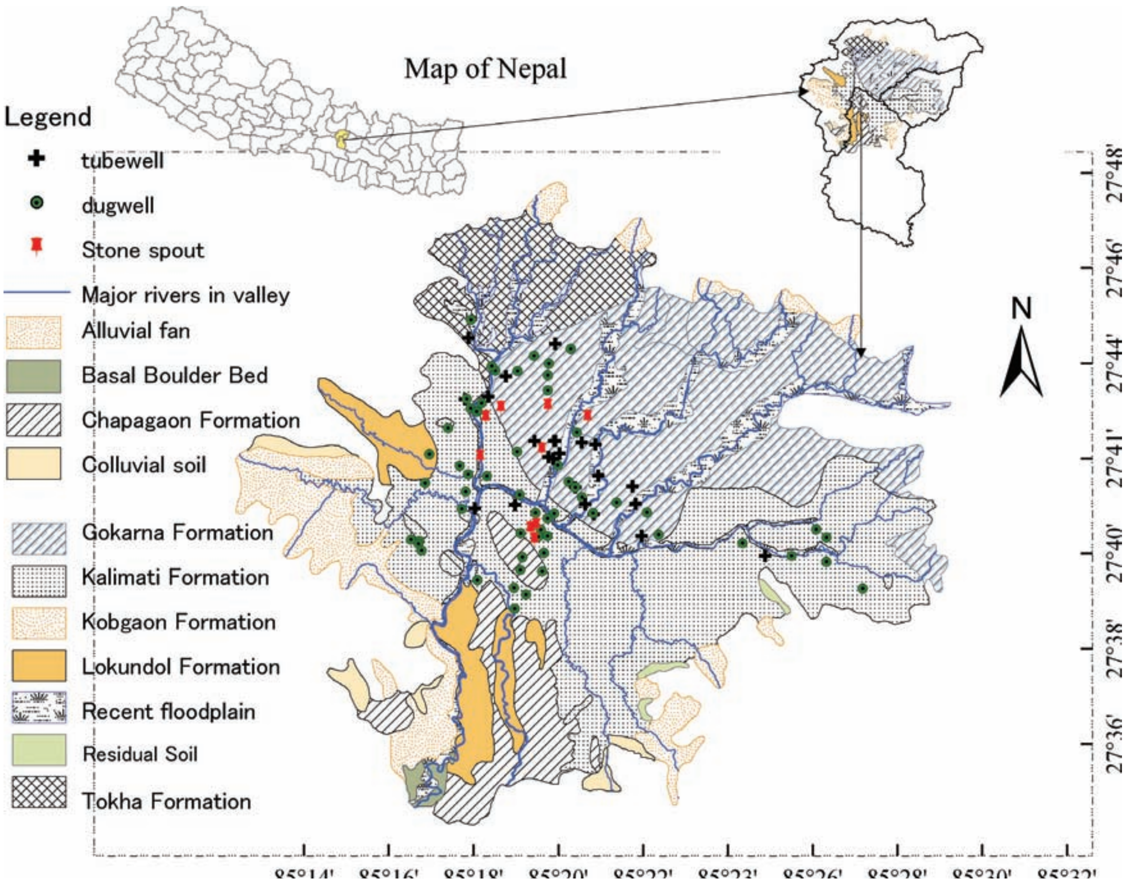

Fig. 1. Sample locations and geological formation within Kathmandu Valley, adapted from engineering and environmental geological map of Kathmandu Valley, Department Mines and Geology, Government of Nepal.

3. Results and discussion

3.1. Distribution of nitrate-N and ferrous iron

The sampled water sources were from shallow groundwater systems of depth ranged from 0.0 to 25 m with average depth 5.1 m however, most of sources were below 10 m. In general, pH of almost all samples (84 of 90) was within the Nepali national water quality standards, which require pH to be between 6.5 and 8.5 [12]. The average temperature of the samples sources was around 25°C.

Nitrate-nitrogen ranged from 0.0 to 26.4 mg/L with average concentration 4.22 mg/L to the shallow groundwater systems of Kathmandu, where 16% of the sampled wells exceeded old WHO guidelines of 10 mg/L as nitrate-nitrogen; however, another 33% wells have impacted levels of nitrate between 2 and 10 mg/L (Fig. 2). Based on the colorimeter analyses, dissolved iron concentration were found ranged from 0.0 to 5.24 mg/L with average concentration 0.79 mg/L. The concentration of iron in most of the observation wells exceeded the maximum allowable limit. 53% of sampled sources were at or above the 0.3 mg/L standard.

Although, nitrate and ferrous iron contamination were widespread in shallow aquifer of Kathmandu Valley, their concentrations were not uniform across the Valley (Figs. 3A, 3B). The spatial distribution map indicates nitrate-nitrogen

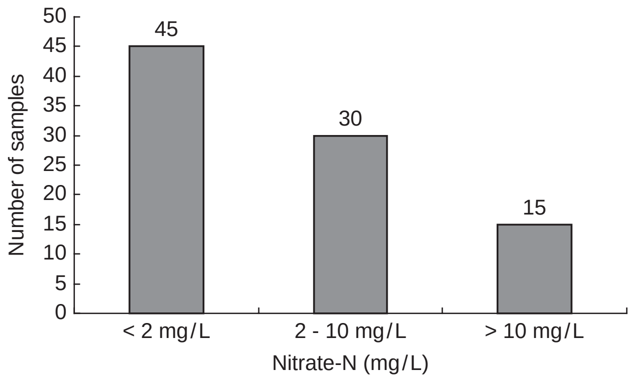

Fig. 2. Nitrate-N concentrations in shallow groundwater systems of Kathmandu Valley.

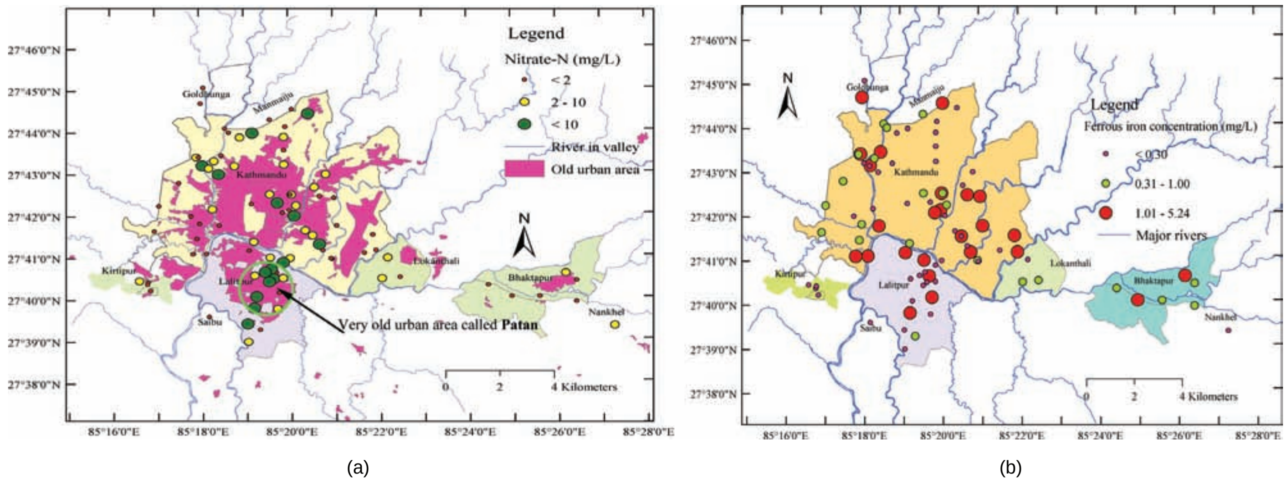

Fig. 3. Spatial distribution of (A) nitrate-N and, (B) ferrous iron in shallow groundwater systems of Kathmandu.

Table 1
Nitrate-N and ferrous iron concentration in different type of groundwater sources within Kathmandu Valley.

	Number of samples	Min	Max	Mean	Median	Above WHO limit (%)
Nitrate-N						
Dug wells	58	0	20.8	4.12	2	18
Stone spouts	9	3.2	26.4	11.7	10.5	55
Shallow tube wells	23	0	8.8	1.53	0	0
Ferrous iron (Fe)						
Dug wells	58	0.00	3.36	0.50	0.21	46
Stone spouts	9	0.03	0.15	0.08	0.09	0
Shallow tube wells	23	0.06	5.24	1.81	1.48	91

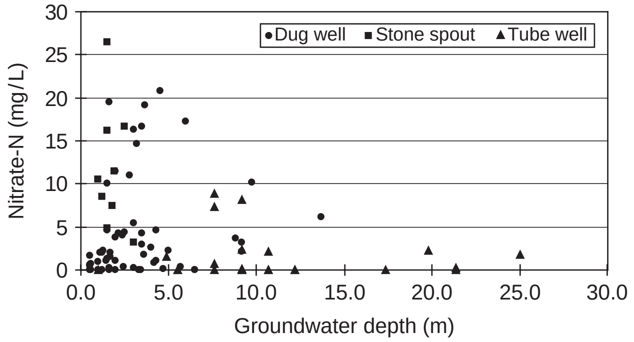

Fig. 4. Relation between nitrate-N and water depth in shallow groundwater systems of Kathmandu Valley.

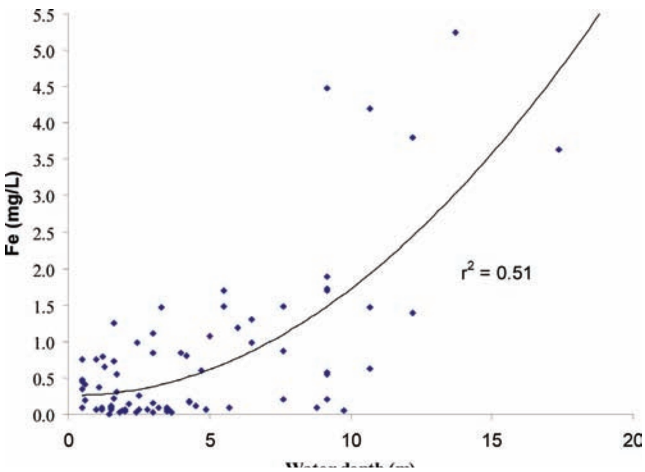

Fig. 5. Relation between ferrous iron and water depth in shallow groundwater systems of Kathmandu Valley.

was found above 10 mg/L (up to 26.4 mg/L) in sampled sources particularly located at old urban centers and northern areas of the Valley that belongs to sandy formation, where many households use septic tanks.

Similar trend had been reported in previous studies [4, 13]. Bittner [13] identified a plume of high nitrate (63.3 mg/L) in the shallow aquifers in Chabahil that is located at northern part of Valley. The summary of descriptive statistic analysis shows the concentrations of nitrate-N and ferrous iron in the three types of sources in shallow groundwater systems (Table 1). Generally, the traditional sources such as stone spouts and dug wells that are considered as 'safe' drinking water sources for hundreds of years were highly contaminated by nitrate nevertheless nitrate rarely detected in shallow tube wells. In contrast, shallow tube wells were most contaminated with iron, but dug wells and stone spouts generally had low levels of iron. Interestingly, about 55% of the sampled sources from stone spouts violated the WHO guidelines of nitrate-N. Only 18% of sampled from dug wells exceeded the WHO limit nevertheless no samples from shallow tube wells were found above the limit value. On the other hand, about 91% and 46% sampled sources from shallow tube wells and dug wells exceeded the WHO iron guideline of 0.3 mg/L, respectively, however no samples from stone spouts were found above the maximum allowable limit.

The vertical distribution of nitrate shows nitrate contamination in groundwater of Kathmandu is much more prevalent at shallow depths due to anthropogenic sources. Therefore, in general, domestic wells (mostly tube wells) are installed from deeper layer of shallow aquifer systems in Kathmandu to get water which is supposed to be 'safe' drinking water sources because nitrate, *E. coli* etc. are much

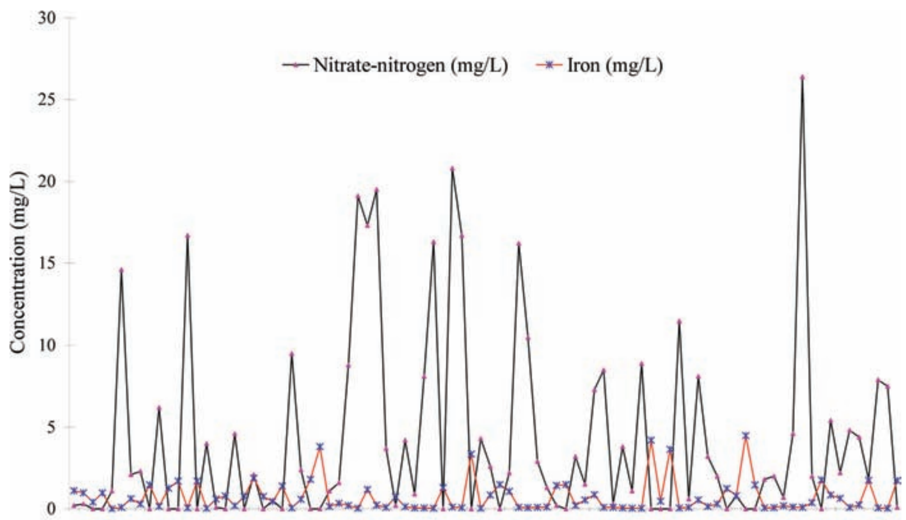

Fig. 6. Nitrate-N and ferrous iron variation in sampled sources in shallow groundwater systems of Kathmandu.

more common at shallow depths. Nevertheless, after few years, the well owners should either abandon those wells or invest huge amount of money to further continue the tapping of water because of the presence of excessive of iron. There is ample observational evidence of the presence of excessive iron in groundwater in deeper depth of the shallow aquifer of Kathmandu, including yellow-red deposits around tube wells and in the water distribution system of private house. Nitrate-N vs. water depth and ferrous iron vs. water depth were plotted to study the relationship of nitrate-N and ferrous iron with water depth in shallow groundwater sources. Fig. 4 clearly shows a gene ral inverse relationship between nitrate-N concentrations and groundwater depth for shallow aquifer systems. Even though nitrate-N concentration decreased on increasing depth of water in well, this relation itself does not explain the mechanisms behind subsurface nitrate transport; this explains only that nitrate is attenuated, diluted, or denitrified with depth below land surface. Ferrous iron concentration, on the other hand, increased with water depth in groundwater sources (Fig. 5). Ferrous iron was well correlated with water depth in groundwater sources with correlation coefficient 0.68. In contrast to the nitrate-N profile, elevated ferrous iron concentrations occurred in shallow groundwater sources with higher depth, which suggest that the aquifer has a suitable redox environment for denitrification, however to obtain a clearer idea of the vertical profiles of nitrate-N and ferrous iron concentrations in the study area, more samples need to be collected and more analysis needs to be performed.

3.2. Possible denitrification

In this study, elevated nitrate concentrations were observed in 16% of sampled groundwater sources up to 26.4 mg/L, where elevated iron concentrations were not found (Fig. 6). A plot of nitrate vs. ferrous iron in the shallow groundwater shows the relationship between these parameters, with the presence of one usually marked by the absence of the other (Fig. 7A). Similar relationships were observed from the previous data set measured in 2005 and 1999 in this area but the magnitude of concentrations was higher (Figs 7B, 7C). Because of the spatially insufficient data, it appears that the ferrous iron available is not enough for denitrification with the current level of data across the entire area based on the stoichiometry of denitrification equation. However, the higher amounts ferrous iron obtained from previous data set measured in 2005 and 1999 from the entire area of shallow aquifer of Kathmandu indicate that this area has good potential for denitrification. A mutually exclusive relationship and the observed negative correlations between nitrate and iron suggest that the nitrate in shallow groundwater is being

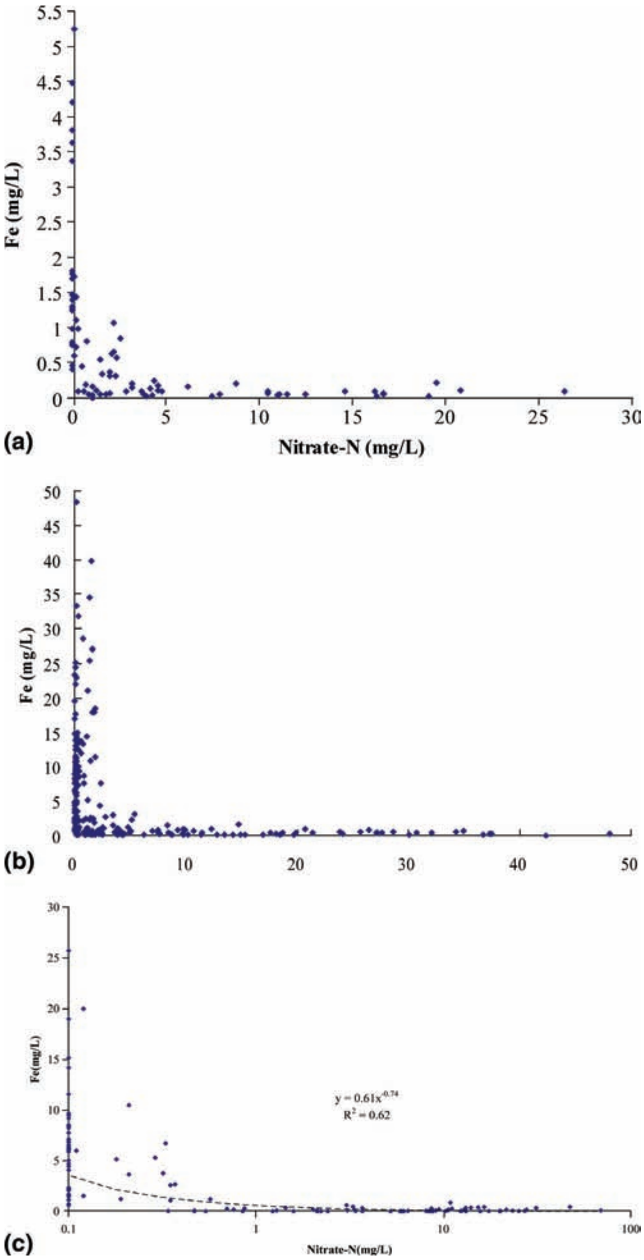

Fig. 7. (A) Relationship between nitrate-N and ferrous iron (Fe) in groundwater sources from (A) present dataset (2008), (B) 2005 data set and, (C) 1999 data set (X-axis in log-scale).

consumed that can provide clues to potential for the natural denitrification. It is suggested that ferrous iron is not only responsible for nitrate reduction, however that iron is serving as indicator of denitrification process. Given the spatial distribution in nitrate concentrations and the fact of temporal variation of nitrate input, it is difficult to determine the exact extent to which nitrate reduction has taken place within the shallow groundwater systems with the current level of data. It is first attempt to investigate the relationship between nitrate-N and ferrous iron to explore the mechanism of natural denitrification and its ability to reduce elevated nitrate concentrations in shallow aquifer of Kathmandu. Hence, it is recommended more geochemical studies of the groundwater in this area to evaluate the potential for biogeochemical processes within the aquifer to understand and control the fate of nitrate.

4. Conclusion

The spatial distribution map reveals high nitrate-nitrogen is abundantly found in the shallow groundwater, especially in dug wells and stone spouts (up to 26.4 mg/L), which suggests it is still major threat to groundwater quality in shallow aquifer of Kathmandu. At the same time, elevated nitrate concentrations were not found in sampled groundwater sources in study area where elevated iron concentrations were common. Generally, it is difficult to determine the extent of nitrate reduction which has taken place within shallow groundwater aquifer due to the fact of the spatial and temporal variation of nitrate input. However, the mutually exclusive relationship between iron and nitrate-N in shallow groundwater systems of Kathmandu suggests nitrate reduction in some extent by natural denitrification process. Similar relationships were obtained from the previous data set measured in 1999 and 2005 from shallow groundwater sources of Kathmandu. In general, the negative correlation between iron and nitrate-N indicates that there has been at least some denitrification. Nevertheless, it is suggested that iron is not only responsible for nitrate reduction, however that iron is serving as indicator of denitrification process. More studies are required in order to quantify the amount of nitrate loss from the shallow groundwater aquifer of Kathmandu due to denitrification.

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