

## Research Article

# Occurrence, Source, and Mobilization of Iron, Manganese, and Arsenic Pollution in Shallow Aquifer

Md. Shajedul Islam and M. G. Mostafa 

*Institute of Environmental Science, University of Rajshahi, Rajshahi 6205, Bangladesh*

Correspondence should be addressed to M. G. Mostafa; [mmostafa@ru.ac.bd](mailto:mmostafa@ru.ac.bd)

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This investigation is aimed at exploring the mobilization and sources of Fe, Mn, and As in aquifers through geochemical characterization, spatial analysis, and statistical approaches. Analytical results showed that the average concentrations of Fe (10.17 mg/L) and Mn (3.6 mg/L) crossed the guideline value but not As in all samples. In this study, the elevated levels of dissolved organic carbon, salinity, total hardness, and  $\log_{10} p\text{CO}_2$ ; the lower value of dissolved oxygen; and some oxidizing groups of samples influence the reductive environment of Fe and Mn weathering. The investigation illustrated that the source of both metals on the upper platform is consolidated silty soil and aquifer sediment, whereas, in the river valley, they originate from Fe and Mn-bearing nodules in the rich clay layer and tidal mud. In addition, the frequent fluctuation of river flow, excess water mining, and an unbalanced recharge/discharge ratio are the causes of higher concentrations of both metals. The correlation matrix and factor analysis of Fe and Mn with redox-sensitive water variables confirmed that a favourable reductive situation fosters the Fe and Mn release process. Studies have also shown the significant possibility of forming a metal complex with organic and inorganic substances. The higher dissolution of Fe and Mn minerals causes As to be adsorbed on them, leading to less release of As into groundwater. This investigation provides a basis for the rational use of groundwater and the defense of public health in zones with high Fe and Mn contents.

## 1. Introduction

Groundwater is a chief source of drinking and irrigation in the upper Gangetic floodplain area of Bangladesh [1, 2]. Heavy metal contamination in groundwater has become a serious threat to environmental systems and human health [3, 4]. Among the water pollutants, iron (Fe), manganese (Mn), and arsenic (As) are existing in chemicals resulting from both geogenic and anthropogenic sources, including soil, rocks, industrial wastes, and the overmining of groundwater [5, 6, 7, 8]. This type of contamination is very common, and the world is facing the greatest challenge in sustainable water management, including Bangladesh [9] and other countries, such as Italy [10], Argentina [11], China [12, 13], and Indonesia [8]. In such cases, an investigation is required to find the source of the contaminants. Several studies [12–19] have investigated the presence and sources of Fe and Mn in groundwater systems within diverse

topographical and geological settings. The authors have observed that the groundwaters of the north and northwest parts of Bangladesh are highly rich in Ca-hardness, iron, and manganese [1]. Especially in the present study area, the physical appearance and testing of that water can prove this characteristic. However, no investigation has been carried out on the actual dissolution mechanisms of both metals in the present calcite aquifer. So, it is crucial to identify the real origin of these two metals in the local aquifer and how the processes will be taken to control them in groundwater.

Fe and Mn are two of the utmost common metals in the environment, and therefore, humans are exposed to substantial amounts of these elements that are present in natural water and foodstuffs. Fe can stain washing garments and plumbing fittings and lead to deposits of residue in water circulation systems [20]. It is considered undesirable in water when the levels exceed the standard value (0.3 mg/L), causing changes in water turbidity, color, and taste [11, 21].

Adverse health effects of Fe and Mn are known to comprise chronic poisoning, pulmonary embolism, nerve damage, bronchitis, impotence, and parkinsonism [22, 23]. Excess As can have serious health belongings on inhabitants, together with skin cancer and other cardiovascular, haematological, neural, kidney, and respiratory troubles [24]. The countrywide guidelines for drinking water quality of Fe, Mn, and As in Bangladesh are 0.3 to 1.0, 0.1, and 0.05 mg/L, respectively [25]. Irrigation water laden with an elevated level of Fe and Mn causes visual problems for crops/plants, and the soil environment and its accumulation on irrigation equipment can lead to clogged emitters [26, 27].

Fe and Mn extensively originated in soils, sediments, and aquifers, and they have analogous geochemical characteristics in the aquatic environment and occur in dissolved, colloidal, ionic, and organic- and inorganic-metal complexes in water [12, 17, 28]. Their oxidized forms ( $\text{Fe}^{3+}$  and  $\text{Mn}^{4+}$ ) are insoluble, though the reduced forms ( $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$ ) have a larger solubility [21]. Water containing Fe and Mn is colorless, but once in contact with air, it turns cloudy and turbid owing to the colloid formation through the oxidation of  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  [29]. This process occurred through homogeneous oxidation by dissolved oxygen (DO), heterogeneous oxidation by adsorption on the surface, and biological oxidation by iron-oxidizing bacteria [13].

In the saturated zone, weathering of Fe-, Mn-, and As-loaded rocks through the dissolution process of primary minerals is controlled by numerous environmental aspects and redox conditions [11, 12]. Reductive conditions are very common in groundwater systems, and this situation has favoured the dissolution of these metals [30–32]. The potential of this reducing environment generally depends on the concentration of some natural water components, such as dissolved organic carbon (DOC), DO, dissolved  $\text{CO}_2$ ,  $\text{NO}_3^-$ ,  $\text{PO}_4^{3-}$ ,  $\text{SO}_4^{2-}$ , and  $\text{HCO}_3^-$ , and some other parameters like pH, EC, and Eh [12, 17, 30]. Through the participation of these factors, both  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  in rock form may be released into the water phase with the same oxidation number over the various chemical reactions. In this situation, except for DOC, all the above components accept electrons and create a good reductive environment [33–35]. Including this reductive condition, the residence time of groundwater, well depth, and total salinity are the significant factors leading to the dissolution and movement of Fe and Mn to groundwater systems [10, 36]. On the other hand, As dissolution and mobilization mostly depend on Fe and Mn weathering processes. Arsenic is unconfined from the reductive weathering of arsenic-laden  $\text{FeO}(\text{OH})$ . It is a redox course that happens after bacteriological oxidation of organic substance consumes DO,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  (as the nutrients of microorganisms) [37–41]. Among the geogenic processes, anthropogenic actions can also contribute to the release of Fe, Mn, and As into groundwater, as these elements are frequently utilized in agriculture, industry, or landfilling actions [42].

Not only chemical influences but also some natural and external factors are involved in the dissolution rate of Fe, Mn, and As. Numerous studies [e.g., 5, 12, 17] have explored the complex influences of local soil properties, lithological

settings, weather conditions, and geomorphology of the study area on the mobilization rate of Fe, Mn, and As in groundwater. These three metals are dispersed regularly in the following environments: a blanket of clay deposits in the sublayer, unbalanced recharge and discharge ratios of water, variations in river flow, river basin, and deltaic floodplain areas, geological formations rich in organic matter, fine particle sediments/soil, etc. [13, 43].

The Upper Gangetic Plain, a significant part of grain growing in Bangladesh, is a distinctive area with high Fe and Mn levels in groundwater. Several studies of groundwater in the Ganges floodplain area have focused on provincial and local hydrogeochemical features and the origin of groundwater containing raised As, Fe, and Mn levels in specific zones. Though, limited investigations of the several factors and mechanisms involved in the causation of high concentrations of these metals in shallow groundwater at the local scale have been accomplished. The source and mechanism connected to the mobilization of Fe and Mn in the aquifer are not realized completely, and the influences of human actions on Fe and Mn loadings in groundwater at the zonal scale need to be explored. This investigation is aimed at (a) evaluating the dispersal of As, Fe, and Mn in shallow groundwater in the study zone; (b) finding the factors (geogenic and anthropogenic) affecting Fe and Mn levels utilizing statistical techniques; and (c) recognizing the dissolution mechanisms involved in causing groundwater in the study area to hold high Fe and Mn levels and low As levels. The geochemical dataset of the groundwater samples was used for various statistical analyses, multivariate tests, computer programs, and chemistry involving these processes. The outcome of this study is expected to provide data for well-remediation approaches along with protecting the health of the inhabitants.

## 2. Materials and Methods

*2.1. Study Area and Hydrogeological Formation.* The study area was Kushtia District, the middle-west zone of Bangladesh, placed at  $23^{\circ}41'$  and  $24^{\circ}11'$  north latitudes and  $89^{\circ}22'$  east longitudes. The total area of this zone is about 1600 km<sup>2</sup> with a population of 2.367 million [44]. This study area is surrounded by the Padma River (Ganges River), and the other three-branched rivers make a big deltaic alluvium sedimentary plain (Figure 1). The soil layer is categorised by unvarying terrain, which appears to level a landscape with a raise of about 30 ft above the sea surface, but in some parts, it comprises shallow depression and slightly higher edges [45]. The sampling stations with S1 to S10 are situated in a relatively lowland area with a low topographic gradient. A previous study [41] confirmed that the groundwater of the shallow aquifer in the study area is mainly calcite type ( $\text{Ca-HCO}_3$  facies). The primary aquifer of the investigated area covers unconsolidated alluvial sediment that is spread over the surface by permeable sand, silt, and clayey soil. The geology of the study zone comprises young alluvial deposits, deltaic silt deposits, stream and floodplain deposits, calcareous sandstone, and conglomerates [44, 46]. Along with subsurface hydrogeological information, it seems that good

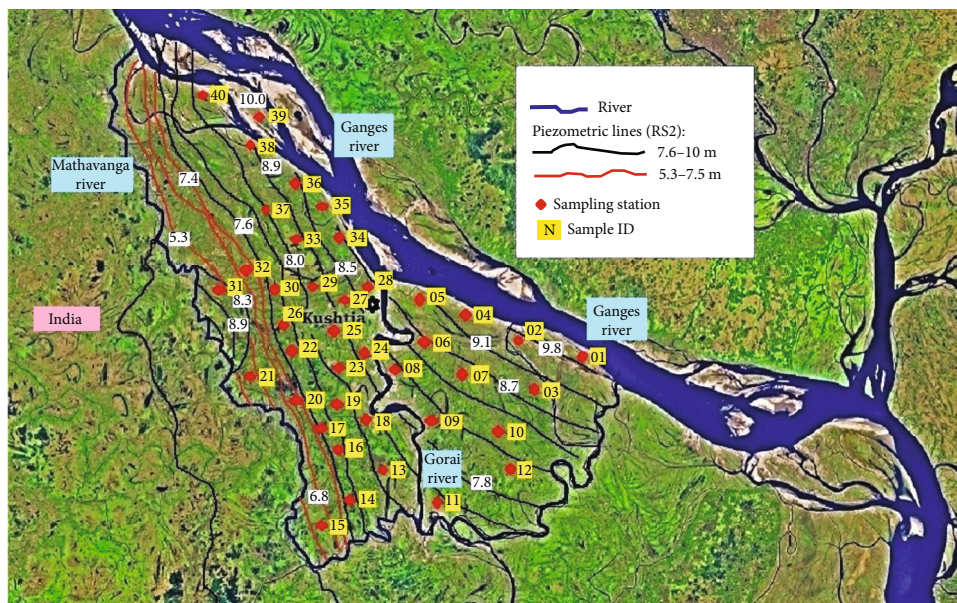


FIGURE 1: Sampling location and piezometric lines (RS2).

aquifers in this part remain between 25 and 140 m in depth [47]. The thickness of the local aquifer varies because of the belongings of basement rock depth and the transverse level of the aquifer. The groundwater movement route in the study area is characteristically from the northeast to the southwest. The study area is enclosed by subtropical humid weather with a warm and rainy monsoon and a pronounced dry time of year in the winter period. A total of 1167 mm/y of rainfall is received in the area [44]. About 95% of groundwater is utilized for agricultural actions, and the enduring portion is used for domestic purposes [48]. So, groundwater quality should be evaluated for irrigation and domestic purposes in this area.

**2.2. Sampling Campaign and Analysis.** Groundwater samples were collected from two periods, viz., the premonsoon (PRM) and postmonsoon (POM) of 2020. A total of 40 samples were collected from shallow aquifers (depths: 16–54 m), but seven originated from a semideep aquifer (depths: 97–110 m) (Figure 1). The water samples were collected after 15 min of pumping and taken into polyethylene bottles, which were prewashed with a 10% HCl solution and finally rinsed with distilled water. Sampling and all analyses were accomplished following the US-EPA guidelines [49]. All the samples were transported to the laboratory for chemical and spectrophotometric analysis. Groundwater temperature, turbidity, electrical conductivity (EC), pH, and dissolved oxygen (DO) were counted instantly using portable multi-meter instruments after allowing sample contact with the atmosphere. Total hardness (TH) and bicarbonate ( $\text{HCO}_3^-$ ) were measured by titrimetric analysis, and nitrate ( $\text{NO}_3^-$ ) and sulphate ( $\text{SO}_4^{2-}$ ) were estimated using a UV spectrophotometer. The dissolved organic carbon (DOC) was estimated by a total organic carbon (TOC) analyser. Fe and Mn were estimated by atomic absorption spectrophotometry, and As was measured by atomic fluorescence spectroscopy. The

dependability of the sample analysis data was crisscrossed by the comparative error of the cation and anion milliequivalents, and the error of samples was below 5%.

**2.3. Statistical Study.** Correlation and factor analysis were carried out among Fe, Mn, As, and some allied physico-chemical parameters utilizing SPSS 22.0 software program. The influencing factors of metal dissolution in the aquifer were determined using multivariate analyses. The strength of a linear correlation among two variable quantities or the degree of association was judged by Pearson's correlation matrix coefficient,  $r$ . Principal component analysis (PCA) was selected, as it is one of the vital useful tools to analyse the interrelationship among different sets of groundwater [50, 51]. This analysis presented reciprocally interfering factors that are formed by water-rock contacts, water mixing from dissimilar origins, and anthropogenic effects [52]. Once the above two variables were measured simultaneously, multiple linear regression tests were used to evaluate their interrelationship [53]. The partial pressure of  $\text{CO}_2$  in aquifer water,  $\log_{10} p\text{CO}_2$ , was determined by PHREEQC v.3.0 [54]. Lastly,  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  from geogenic origins were appraised using lognormal probability plots.

### 3. Results and Discussion

**3.1. Physiochemical Water Variables.** The basic statistics of selected groundwater parameter values in different seasons of the study area are stated in Table 1. In this study, Fe concentrations were found in water samples ranging between 1.09 and 14.7 mg/L (mean:  $9.63 \pm 3.6$ ) in the premonsoon (PRM) and 1.59 and 17.2 mg/L (mean:  $10.7 \pm 3.56$ ) in the postmonsoon (POM). On the other hand, Mn concentrations of 0.86 to 6.08 mg/L (mean:  $3.44 \pm 1.5$ ) in the PRM and 0.96 to 6.58 mg/L (mean:  $3.76 \pm 1.56$ ) in the POM period were found. The levels of Fe and Mn in all samples

TABLE 1: Statistical analyses of the groundwater data in the premonsoon (PRM) and postmonsoon (POM) seasons, 2020.

|                                  | Turb. | pH   | EC    | TH    | Fe    | Mn    | As    | HCO <sub>3</sub> <sup>-</sup> | SO <sub>4</sub> <sup>2-</sup> | NO <sub>3</sub> <sup>-</sup> | DO   | DOC   | log <sub>10</sub> pCO <sub>2</sub> |
|----------------------------------|-------|------|-------|-------|-------|-------|-------|-------------------------------|-------------------------------|------------------------------|------|-------|------------------------------------|
| Premonsoon (PRM), <i>n</i> = 40  |       |      |       |       |       |       |       |                               |                               |                              |      |       |                                    |
| Min.                             | 1.93  | 6.65 | 366   | 122   | 1.09  | 0.86  | 0     | 271.5                         | 2.91                          | 0.80                         | 0.68 | 0.61  | -2.458                             |
| Max.                             | 19.56 | 7.80 | 1035  | 562   | 14.7  | 6.08  | 0.02  | 703                           | 45.7                          | 14.3                         | 4.35 | 13.60 | -0.884                             |
| Mean                             | 6.26  | 7.02 | 669.6 | 362.2 | 9.63  | 3.44  | 0.008 | 418.6                         | 16.46                         | 4.14                         | 2.13 | 4.46  | -1.952                             |
| SD (±)                           | 4.37  | 0.22 | 172.5 | 93.52 | 3.60  | 1.50  | 0.006 | 110.8                         | 9.29                          | 3.81                         | 1.00 | 3.27  | 0.260                              |
| Skewness                         | 1.57  | 2.10 | 0.36  | -0.07 | -0.55 | 0.05  | 0.11  | 1.36                          | 1.19                          | 1.11                         | 0.49 | 0.39  | -0.933                             |
| Postmonsoon (POM), <i>n</i> = 40 |       |      |       |       |       |       |       |                               |                               |                              |      |       |                                    |
| Min.                             | 2.29  | 7.0  | 662   | 225   | 1.59  | 0.96  | 0     | 248.5                         | 2.95                          | 0.80                         | 0.81 | 0.60  | -3.514                             |
| Max.                             | 26.09 | 8.91 | 1708  | 615   | 17.2  | 6.58  | 0.02  | 817                           | 41.7                          | 18.3                         | 4.02 | 13.71 | -1.260                             |
| Mean                             | 7.85  | 7.83 | 956.8 | 404.7 | 10.7  | 3.76  | 0.008 | 448.8                         | 15.14                         | 3.7                          | 2.07 | 5.71  | -2.272                             |
| SD (±)                           | 5.06  | 0.40 | 206.1 | 94.03 | 3.56  | 1.54  | 0.006 | 124.5                         | 9.25                          | 3.64                         | 0.95 | 3.22  | 0.450                              |
| Skewness                         | 1.66  | 0.02 | 1.31  | 0.59  | -0.46 | -0.07 | 0.11  | 0.61                          | 1.33                          | 2.01                         | 0.31 | 0.38  | -0.009                             |

Note: (a) *n* = number of samples, Turb. = turbidity; EC = electrical conductivity, TH = total hardness; DO = dissolved oxygen; DOC = dissolved organic carbon. (b) The unit of all parameters is in mg/L except pH, turbidity in NTU, and EC in  $\mu\text{S}/\text{cm}$ .

were higher than the native and international guideline values for drinking and agriculture water [21, 55, 56]. Also, these concentrations are higher than the countrywide average values of Fe (3.48 mg/L) and Mn (1.67 mg/L) [1]. The analysis results showed seasonal variations in the concentrations of the two metals. Except for the dissolved oxygen (DO) and log<sub>10</sub>pCO<sub>2</sub>, all measured water parameter values in the POM were higher than those in the PRM period. The pH value of the samples in the PRM season was found to be near neutral ranges (pH: 7.02 ± 0.22), while in the POM, the waters were slightly basic (pH: 7.83 ± 0.4). The electrical conductivity (EC) and total hardness (TH) are both recorded as higher values in the POM than in the PRM season. Overall, the value of these two parameters crossed the national standard guideline [25]. Among the other parameters, such as HCO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and NO<sub>3</sub><sup>-</sup>, the concentration of HCO<sub>3</sub><sup>-</sup> was found in higher ranges in both seasons. In this study, less dissolved oxygen (DO) and higher dissolved organic carbon (DOC) are the significant features of the groundwater. The partial pressure of CO<sub>2</sub> (log<sub>10</sub>pCO<sub>2</sub>) in the aquifer was found to be much higher than that in other dissembler geographic places [8, 57]. The partial pressure of groundwater (log<sub>10</sub>pCO<sub>2</sub> values) in the PRM was lower than that in the POM period due to the heavy percolation of rainwater in the monsoon season (Table 1). Although Bangladesh faces serious toxicity concerns of arsenic (As), the study area is free from arsenic poisoning. The concentration of As in all groundwater samples was found to be less than the national standard (0.05 mg/L) in both sampling periods. Turbidity is an important indicator of the amount of suspended sediment in the water, which makes the water cloudy or opaque. Good water should have less than 5 NTU (nephelometric turbidity unit) of turbidity [13], but we found a higher turbidity value in samples of both seasons. This value in the POM sample (7.85 ± 5.06 NTU) was higher than that in the PRM season (6.26 ± 4.37 NTU). The multivariate statistical analysis utilized typically distributed data. The values of skewness of maximum

parameters varied between 0 and +2, which indicated that they are normally distributed [58].

A study showed that the water facies is Ca-HCO<sub>3</sub> type (major) and that calcite mineral dissolution in the water phase is the main regulating factor for water-rock interaction processes [59]. It was measured by saturation index (SI) with the support of the PHREEQC-3v program. Saturation or undersaturation of minerals in groundwater impacts the water chemistry as well as the weathering processes of metal-laden rocks. The index value showed whether water would tend to precipitate or dissolve a specific mineral. The saturation index (SI) is intended to compare the chemical activity of the dissolved ion of minerals with their solubility product ( $K_{sp}$ ) at a definite temperature. A positive SI value designates supersaturation with minerals and an inclination for the minerals to precipitate from the water solution, and a negative SI value points to the propensity for the minerals to dissolve in the same medium. It was detected from the data that there are approximately 90%, 20%, and 10% of the total samples in both seasons; the index values for calcite, dolomite, and aragonite minerals were observed to be more than 0. These results indicated that groundwater is oversaturated with these minerals, and thus, they are precipitated in groundwater. Therefore, the findings showed that maximum samples were supersaturated regarding calcite mineral (CaCO<sub>3</sub>) and led to higher levels of Ca<sup>2+</sup> and HCO<sub>3</sub><sup>-</sup> in samples in both seasons in the study zone. The higher SI value for calcite focused the opportunity for an extra rise of Ca<sup>2+</sup>, Mg<sup>2+</sup>, and HCO<sub>3</sub><sup>-</sup> concentration in the samples of the study zone owing to the extra dissolution of this kind of mineral. Furthermore, the partial pressure of dissolved carbon dioxide (pCO<sub>2</sub>) has an important role in the mineral dissociation course. The groundwater samples with calcite mineral saturation recommended that these carbonate minerals were the key participants in the host rock. This supersaturation of calcite minerals can mostly impact the water quality and regulate the reactions involved in water at certain pH and temperature levels. The elevated

concentrations of earth metal ions, bicarbonate, and dissolved  $\text{CO}_2$  may influence the Fe and Mn levels in groundwater through a set of chemical reactions.

**3.2. Explorative Statistical Approach—Multivariate Analyses.** Multivariate statistical techniques exploit normally distributed data [60]. The water quality data for most physical and chemical parameters are positively skewed (Table 1). In the case of statistic and probability theory, skewness is a quantity of the disproportionateness of the statistical probability distribution of the random variable about its mean [61]. If the skewness value is between 0.5 and 1.0, the figure is moderately skewed. If it is less than -1.0 or greater than +1.0, the figure is extremely skewed [62]. It does not imply that data are normally distributed.

Correlation and factor analysis are included in the multivariate statistical technique. The correlation matrix of Fe, Mn, and As with other influencing variables is shown in Table 2. Little temporal variation is observed in the correlation matrix table between the variables of the PRM and POM seasons. First, Fe and Mn are strongly correlated (positive) with each other in both periods ( $r \sim 0.8$ ,  $p < 0.01$ ), which indicates a similar dissolution mechanism of the rock source into groundwater [12]. Fe, Mn, and As have a weak negative correlation with pH (at  $p < 0.05$ ) representing that acidic conditions encourage the dissolution of these metals [63]. Moreover, these three metals were moderate to strongly positively correlated with EC in both sampling periods. A high EC value indicates a high salinity level in the water, and the concentration of these three metals is highly associated with salinity. A strong significant positive correlation of Fe, Mn, and As with TH (at  $p < 0.01$ ) in the POM indicates the influence of inorganic complexes [64]. The significant positive correlation (over  $r = 0.7$  at  $p < 0.01$ ) of turbidity with Fe and Mn suggested the presence of colloidal Fe(III) and Mn(IV) particulates in the water [12, 65, 66].

The oxidizing agents, viz.,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{CO}_2$  are weakly to moderately and positively associated with Fe, Mn, and As. It is less correlated than DO, which may be due to the less favoured electron acceptor after oxygen [35]. However, the positive correlation noted between  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  with those oxidizing agents implies a reduction process that does not have enough influence on Fe and Mn dissolution. Table 2 explores the very high positive and negative correlations ( $r > +0.8$  and  $r < -0.8$  at  $p < 0.01$ ) of Fe and Mn with DOC and DO, respectively, found in both sampling seasons. But As has a moderate positive and negative correlation with these two variables, respectively. If the aquifer contains more organic matter, the level of DO is consequently low. Arsenic was also positively associated with Fe and Mn,  $r = 0.36$  to  $0.38$  and  $r = 0.40$  to  $0.48$  in PRM and POM, respectively. This is because As has a strong geochemical attraction with Fe-Mn-oxhydroxide [41]. The roles of the factors in the dissolution techniques of Fe, Mn, and As are discussed in the subsequent sections.

The factor analysis showed a total variance of 84.14% with an eigenvalue  $>1$  in the PRM season, as determined by three PCs of R-mode (Table 3 and Figure 2). Factor 1 (F1) accounts for 53.41% of the total variation. This suggests

that with the changes in pH and redox state, the inorganic sources of Fe and Mn are dominant and highly dependent on organic matter loading in groundwater, as exposed prior by the correlation matrix. F2 clarifies 16.96% of the total variation for the same season. This factor designates the consequence of total hardness (TH) in samples (Figure 2). It suggests that changes in Ca, Mg, and  $\text{HCO}_3^-$  concentrations (TH) caused by seasonal variation  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  may undergo complexation with  $\text{HCO}_3^-$ . Factor 3 accounts for 13.28% of the variation and is the highly positive loading of pH. It is related to the pH and redox processes of Fe and Mn dissolution. The redox potential of these metals is not significantly associated with the pH of the medium.

During the POM season, the factor analysis showed a total variance of 76.27% determined by only two PCs of R-mode (Table 3 and Figure 2). Factor 1 (F1) accounts for 62.15% of the total variation. This factor suggests that with significant fluctuations in pH and redox state, after the heavy rain of the monsoon season, the inorganic sources of iron and manganese are highly dominant and dependent on organic matter loading in groundwater. Another factor, F2, explains 14.12% of the total variation. This factor designates the relatively low influence of pH on Fe and Mn dissolution. Besides, probably due to temporal variation, this factor showed a negative TH with these metals.

**3.3. Spatial and Seasonal Distributions of Fe and Mn.** Geologically, the study zone is separated into two lithostructural platforms, viz., Gangetic alluvium (*Ga*) and Deltaic alluvium (*Da*) (Figure 3). The geological and soil formations of *Ga* and *Da* are quite different. Both have quaternary formations, but *Ga* consists of floodplain deposits, fine sand silt, and clay with muddy deposits, and *Da* comprises relatively sand, silt, and gravel depositions. The eras of the *Ga* and *Da* are also different, which are the Holocene (0.0117 ma) and early Pleistocene ( $>2.6$  ma), respectively [46]. The results showed that both the Fe and Mn levels of 95% of the water samples from the *Da* platform were lower than those from the *Ga* platform (Figure 3). The maximum piezometric lines (RS2) of 7.6-10.0 m (water tables) crossed *Ga* and 5.3-7.5 m crossed the *Da* platform (Figure 1). Thus, the geomorphology and geology greatly impacted the Fe and Mn levels in the samples of the study area. The study results indicated that the concentrations of these two metals slightly depended on the water depths (Table 2) of the aquifer.

In the premonsoon (PRM) period (March-June), the transboundary river, the Ganges of Bangladesh part, and other branch rivers become nearly dead. During this period, the groundwater recharge from rivers stops, and the water table reaches below the surface. Moreover, in this dry period, huge water mining occurred for vast irrigation in the farmlands of the study area. This situation can impact the changes in water chemistry in aquifers. But during the monsoon and postmonsoon periods, the scenarios are fully reversed where rivers overflow their banks and flood some areas in the Ganges basin. In these periods, huge water recharging occurs with heavy leaching of chemical nutrients from several sources. Thus, the water chemistry varies seasonally in the upper floodplain areas. The results showed

TABLE 2: Matrix table of Pearson's correlation of selected water variable quantity in both the premonsoon (PRM) and postmonsoon (POM) periods.

|                     | Depth          | Postmonsoon (POM) |               |                |                |                |               |                  |                 |                 |                |                |                     |                |
|---------------------|----------------|-------------------|---------------|----------------|----------------|----------------|---------------|------------------|-----------------|-----------------|----------------|----------------|---------------------|----------------|
|                     |                | pH                | EC            | TH             | Fe             | Mn             | As            | HCO <sub>3</sub> | SO <sub>4</sub> | NO <sub>3</sub> | DO             | DOC            | logpCO <sub>2</sub> | Turb.          |
| Depth               |                | 0.02              | -0.30 *       | <b>-0.51**</b> | -0.39 *        | -0.31 *        | -0.19         | 0.23             | -0.09           | -0.11           | 0.21           | -0.29          | -0.09               | <b>-0.41**</b> |
| pH                  | 0.02           |                   | -0.41         | -0.44          | -0.38          | -0.38          | -0.33 *       | -0.35 *          | <b>-0.47**</b>  | -0.04           | 0.34 *         | -0.38 *        | <b>-0.87**</b>      | -0.30          |
| EC                  | -0.40 *        | -0.12             |               | <b>0.59**</b>  | <b>0.68**</b>  | <b>0.70**</b>  | 0.35 *        | <b>0.44**</b>    | <b>0.60**</b>   | <b>0.50**</b>   | <b>-0.56**</b> | <b>0.69**</b>  | <b>0.47**</b>       | <b>0.65**</b>  |
| TH                  | <b>-0.48**</b> | -0.10             | <b>0.58**</b> |                | <b>0.43**</b>  | <b>0.49**</b>  | <b>0.46**</b> | <b>0.77**</b>    | <b>0.53**</b>   | -0.02           | <b>-0.43**</b> | <b>0.49**</b>  | <b>0.58**</b>       | 0.32 *         |
| Fe                  | -0.38 *        | -0.09             | <b>0.49**</b> | 0.20           |                | <b>0.79**</b>  | 0.38 *        | 0.21             | 0.32 *          | 0.22            | <b>-0.84**</b> | <b>0.93**</b>  | 0.38*               | <b>0.80**</b>  |
| Mn                  | -0.34 *        | -0.11             | <b>0.60**</b> | 0.24           | <b>0.81**</b>  |                | <b>0.48**</b> | 0.36 *           | 0.25            | 0.25            | <b>-0.81**</b> | <b>0.85**</b>  | <b>0.42**</b>       | <b>0.75**</b>  |
| As                  | -0.21          | 0.08              | 0.39*         | <b>0.53**</b>  | 0.36 *         | 0.40 *         |               | <b>0.44**</b>    | 0.26            | 0.10            | -0.38 *        | 0.38 *         | <b>0.41**</b>       | <b>0.45**</b>  |
| HCO <sub>3</sub>    | -0.12          | -0.07             | <b>0.72**</b> | <b>0.58**</b>  | 0.20           | 0.34 *         | <b>0.44**</b> |                  | <b>0.49**</b>   | 0.03            | -0.22          | 0.33 *         | <b>0.57**</b>       | 0.21           |
| SO <sub>4</sub>     | -0.09          | -0.27             | <b>0.67**</b> | <b>0.51**</b>  | 0.25           | 0.28           | 0.26          | <b>0.48**</b>    |                 | 0.27            | -0.30          | 0.40 *         | <b>0.54**</b>       | 0.31 *         |
| NO <sub>3</sub>     | -0.10          | -0.12             | 0.24          | 0.15           | 0.18           | 0.30           | 0.10          | 0.03             | 0.27            |                 | -0.21          | 0.30           | 0.04                | <b>0.49**</b>  |
| DO                  | 0.19           | 0.07              | -0.40 *       | -0.10          | <b>-0.87**</b> | <b>-0.83**</b> | -0.26         | -0.06            | -0.17           | -0.18           |                | <b>-0.88**</b> | -0.35 *             | <b>-0.76**</b> |
| DOC                 | -0.26          | -0.09             | <b>0.42**</b> | 0.20           | <b>0.79**</b>  | <b>0.80**</b>  | 0.32 *        | 0.17             | 0.24            | 0.32 *          | <b>-0.89**</b> |                | <b>0.42**</b>       | <b>0.77**</b>  |
| logpCO <sub>2</sub> | -0.11          | <b>-0.82**</b>    | 0.39 *        | 0.32 *         | 0.22           | 0.29           | 0.08          | 0.36 *           | <b>0.45**</b>   | 0.17            | -0.16          | 0.22           |                     | 0.32 *         |
| Turb.               | <b>-0.43**</b> | 0.03              | 0.40 *        | 0.19           | <b>0.77**</b>  | <b>0.71**</b>  | <b>0.42**</b> | 0.18             | 0.22            | <b>0.43**</b>   | <b>-0.76**</b> | <b>0.75**</b>  | 0.09                |                |

Premonsoon (PRM)

\*\*Correlation is substantial at the probability  $p < 0.01$  in 95% confidence interval (2-tailed, bold). \*Correlation is at the probability  $p < 0.05$  in 95% confidence interval (2-tailed, italic).

TABLE 3: Principal component loading of the analysed parameters in the study area (sorted by size).

| Parameters | Premonsoon (PRM) |             |             | Parameters | Postmonsoon (POM) |       |
|------------|------------------|-------------|-------------|------------|-------------------|-------|
|            | F1               | F2          | F3          |            | F1                | F2    |
| DOC        | <b>0.93</b>      | -0.23       | -0.04       | DOC        | <b>0.93</b>       | 0.25  |
| Mn         | <b>0.92</b>      | -0.26       | 0.02        | Mn         | <b>0.92</b>       | 0.17  |
| Fe         | <b>0.88</b>      | -0.34       | 0.05        | Fe         | <b>0.91</b>       | 0.27  |
| Turbidity  | <b>0.88</b>      | -0.20       | 0.15        | Turbidity  | <b>0.86</b>       | 0.32  |
| EC         | 0.74             | 0.33        | -0.16       | EC         | <b>0.82</b>       | -0.04 |
| As         | 0.59             | 0.53        | 0.26        | TH         | 0.65              | -0.55 |
| TH         | 0.49             | <b>0.80</b> | -0.14       | As         | 0.57              | -0.41 |
| pH         | -0.12            | 0.08        | <b>0.96</b> | pH         | -0.53             | 0.56  |
| Eigenvalue | 4.27             | 1.36        | 1.08        | Eigenvalue | 4.97              | 1.05  |
| % variance | 53.41            | 16.96       | 13.28       | % variance | 62.15             | 14.12 |

Note: bold values denote strong loading with their respective component numbers.

that some water parameter values fluctuated in the sampling periods of the PRM and POM. Iron and manganese in groundwater samples were obtained in higher amounts on the Ga platform than on the Da platform in both sampling periods (Figure 4). On the other hand, the oxidation-reduction environment is the key factor promoting the dissolution process of these two metals. Alternative redox environments are caused by frequent cycles of water logging due to monsoon rainfall followed by the dry summer period. The alternating redox process in the soil is caused by seasonal groundwater level fluctuation, which leads to the formation of Fe-Mn nodules in the soil subsurface layer [67]. Thus, these are another cause of seasonal variation in redox potential as well as Fe and Mn dissolution dissimilarities.

Dissolved oxygen (DO) is the crucial factor in the dissolution of metals at the aquifer level. Although the recharge of atmospheric oxygen in the rainy season occurs relatively higher than in the premonsoon dry season, this oxygenated water will avoid the Fe and Mn from dissolving, and the water pumped from the tube well will have low levels of these metals. In the recharge water, after the oxygen has been consumed, Fe and Mn will again be dissolved, and the water will have dissolved Fe and Mn characteristics [18, 68]. This is a fact, but we get higher levels of Fe and Mn in the postmonsoon season than in the dry premonsoon. So, this is because the sufficiently higher level of DO in the groundwater of the study area is not fully accounted for in the dissolution potentiality of Fe and Mn. Here, instead of

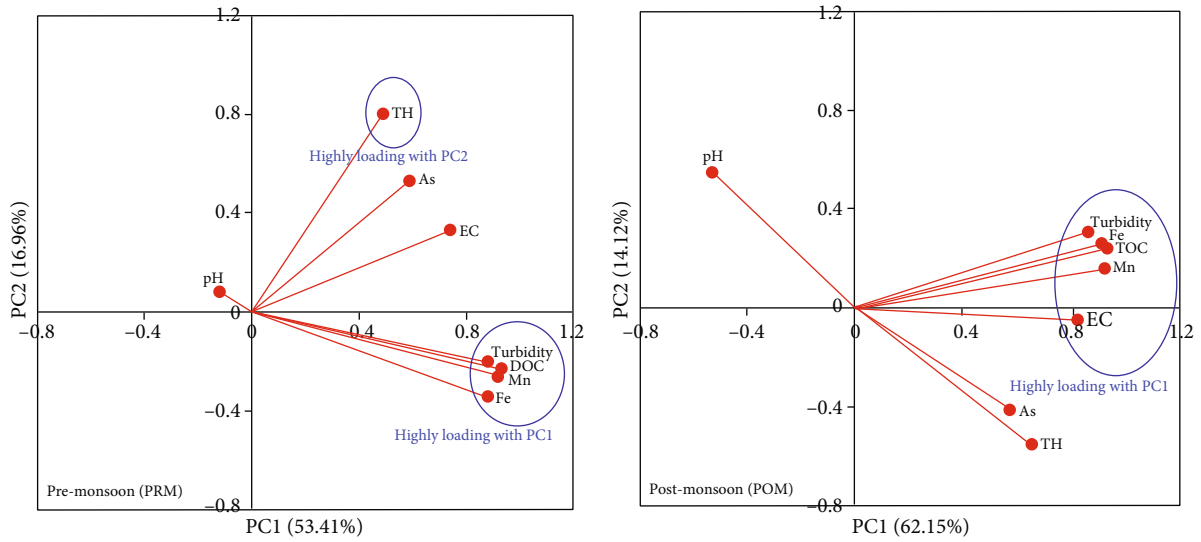


FIGURE 2: Prediction of the factor loading for the different components (PC1 and PC2) for the robust PCA.

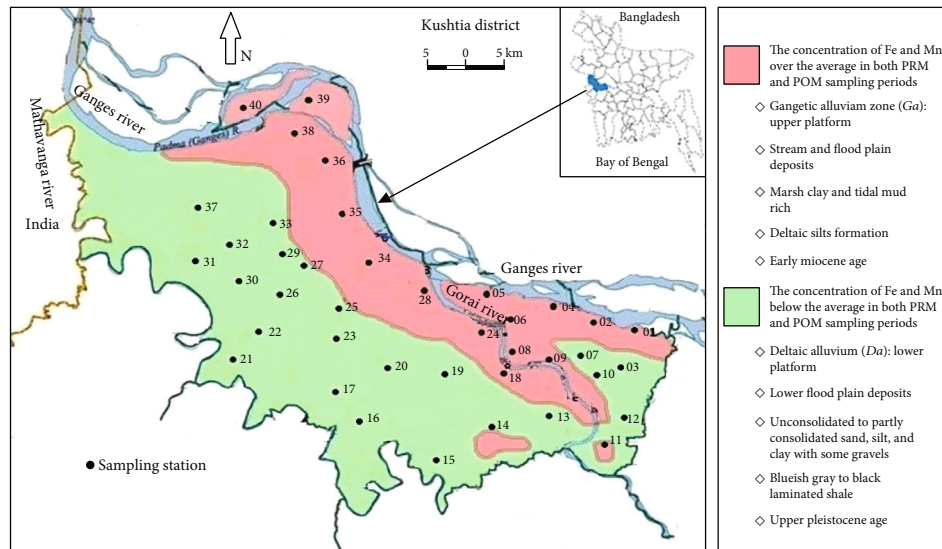


FIGURE 3: Main lithotypes and soil formations of the study area and locations of the sampling stations. The red color is for the Gangetic alluvium (*Ga*), and the green color is for the Deltaic alluvium (*Da*); the result showed that the levels of Fe and Mn are below average in the *Da* aquifer but over average in the *Ga* aquifer.

DO, lithological conditions are the main cause of raised Fe and Mn in the groundwater of the study area.

**3.4. Lithological Impacts on Fe and Mn Dissolution.** The levels of Fe and Mn in groundwater are typically connected to the geology, groundwater flow pattern, well depth, overburden thickness, residence time, and well age [67–69]. It was observed during the field investigation that the extracted colorless groundwater was turned to reddish or yellowish immediately after interaction with air. This designates that the water is in a reduced state. This reductive situation is raised by the amalgamation of organic matter and fine-grained sediments [34]. The geology of the platform in the study area is Quaternary deposits and bears a thick clay layer [46]. Generally, Fe and Mn nodules can be found in the clay

layer [8]. The Gangetic alluvium (*Ga*) formation is composed of loess-like clay (clay with very fine grains) in the river valley, which contains large amounts of Fe and Mn nodules and exhibits Fe and Mn contamination (Figure 3). On the other hand, deltaic alluvium (*Da*) is mostly composed of unconsolidated sand, silty subclay, and fine sand with some gravel (Figure 3). The thickness of the clay layer in the platforms is suggestively different [70]. The clay thickness of this area varies between approximately 20 m and 40 m [46]. This clayey layer overlies the only sandy formation. The elevated level of dissolved organic carbon (DOC) in clays and subclays of aquifer enhances the anoxic conditions that boost Fe and Mn mobilization [71, 72]. A diagram between Fe and Mn in groundwater and the thickness of clay in the sublayers of the sampling well was made to show a

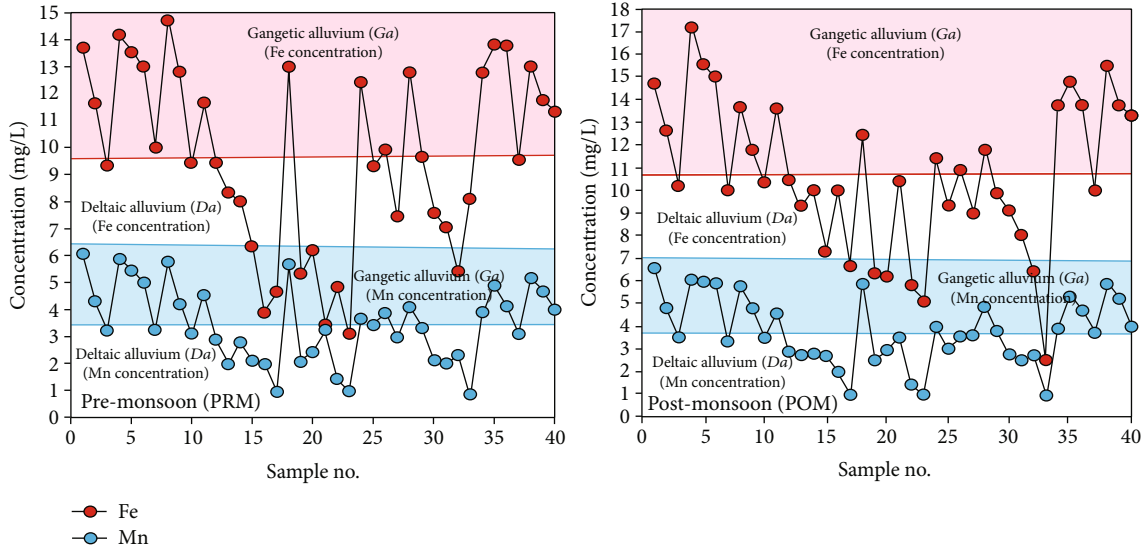


FIGURE 4: Iron and manganese levels in different sampling periods of the Gangetic alluvium (*Ga*) and Deltaic alluvium (*Da*) platforms.

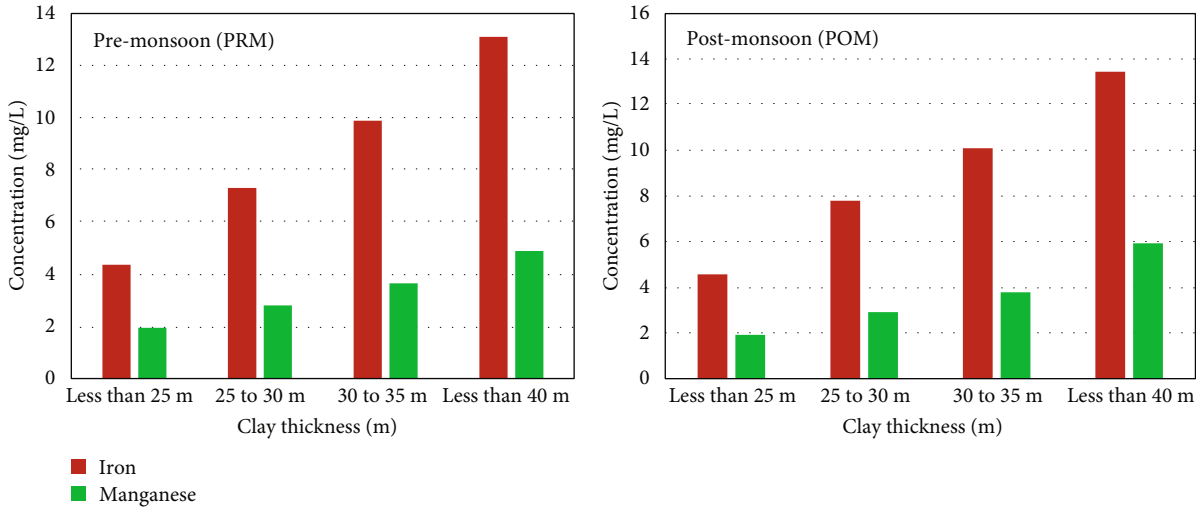


FIGURE 5: Bar diagram of the clay thickness of sample well and average Fe and Mn contents in the PRM and POM seasons.

clear scenario in this study area (Figure 5). The estimated clay thickness data of the borehole sampling well were collected from the local Bangladesh Agriculture Development Corporation (BADC) office. The clay thickness of the *Ga* platform is higher than that of the *Da*. Additionally, Fe and Mn levels were found to be greater in the *Ga* formation than in the *Da* formation. It can be understood from the diagram that the concentration of Fe and Mn in groundwater rises with the rise in clay thickness.

Moreover, the concentrations and correlations of Fe and Mn in the PRM and POM sampling periods are also different. The results showed that Fe and Mn have dissimilar sources in the *Ga* and *Da* platforms. The *Ga* sediments are comparatively young and hold various organic matters. Besides, the lower part of *Ga* is covered by swampland throughout half of the year, which adds extra organic matter to the dead plant in the dry season. Studies [73, 74] have noted that a big portion of organic matter comes from dead

plants and plant debris in the soil. Therefore, the source of Fe and Mn in the samples of the river valley (*Ga*) is not only clay but also whole soils, sediments, and aquifers. The study area has a long history of rice farming. Paddy soils are extensively spread in rice-growing areas. Under the situation of artificial intervallic submergence and drainage, paddy soil was held in reserve in the course of redox fluctuation for a long period and experienced a sequence of exclusive biochemical variations [13]. This is the elementary formation procedure of paddy soil. Enduring waterlogged conditions are advantageous for the deposition of soil organic substance. The use of organic dung manures unceasingly increases the organic matter in the local soil. The Fe- and Mn-oxides in the cultivating layer were abridged to low-valence Fe and Mn compounds under submerging situations and arrived in the aquifer with the water due to gravity action [75]. In another case, the correlation matrix table (Table 2) displays that water depth has a negative



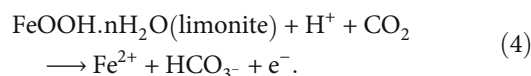
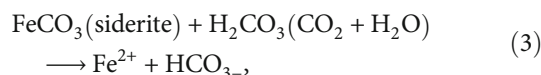
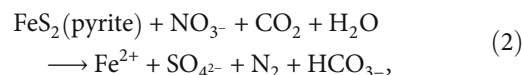
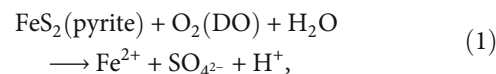
intermediate association with Fe and Mn. A field study showed that the average shallow well depth of the *Ga* area is less than that of the *Da* area. So, the water depth is another cause of the higher concentration of Fe and Mn.

The shallow and intermediate wells with alluvium formation have less As and more Mn and Fe in the study area. It was observed from several studies [e.g., 10-12, 64] that the concentrations, dissolution mechanisms, influencing factors, etc., for Fe and Mn in different types of aquifers or wells are not the same. In the Songnen Plain, Northeast China [76], the maximum wells are gravel-type, and the consequences of these two metals in groundwater are different from those in the present study. Particularly, the Fe and Mn levels in well water are higher in parts covering paddy fields and water shades than in other land-use type parts. The original causes for the high Fe and Mn loads in groundwater in this zone are the Fe and Mn mineral-rich layer and soil with rich organic substance acting as causes of Fe and Mn, and the reductive environment in the lower terrain and areas containing water bodies preferring dissolution of these metals in the groundwater. Besides, the investigation illustrated that contributions of contaminants from farming events have produced the Fe and Mn levels in groundwater to upsurge. On the other hand, the Fe and Mn loads in coastal and arid groundwater are not identical to those in upland groundwater. In coastal alluvial plains, in Indonesia, Rusydi et al. [8] showed that Fe and Mn were natural pollutants in the groundwater of the exploring area. The connections of these metals with redox-sensitive parameters and salinity confirm that saline water has an important effect on the weathering of Fe- and Mn-bearing rocks.

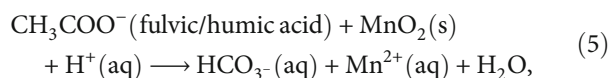
**3.5. Sources and Dissolution of Fe and Mn.** Statistical and chemical analyses showed that the Fe and Mn levels in the aquifer were suggestively associated with climate events (temperature and rainfall), surface topographies (soil type, soil texture, altitude, distance from river, and land use type), and geochemical features (pH, DO, DOC,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and  $\text{PO}_4^{3-}$ ). Multivariate analysis (correlation and factor analysis) indicated that the source and transportation mechanisms of Fe and Mn are almost the same. But, it is not always correct in different geomorphological conditions, such as in central Adriatic Italy [17] and Changchun, north-eastern China [12], in which the mobilization mechanism of these metals is different. Fe and Mn sources are mostly soils, close to the aquifer clay layers, and anthropogenic [33]. Under proper reductive conditions, several existing physiochemical parameters of water participate in the chemical dissolution processes of these metal-laden rocks. Reducing conditions at near-neutral pH ( $\sim 7$ ) is known to initiate the dissolution of iron rocks (mostly iron oxides) and the enhancement of groundwater [10, 77, 78]. These redox conditions are raised by a comparatively high content of organic carbon and fine-grained clayey sediment because organic matter favours the mobilization of Fe and Mn from the soil to the aquifer matrix into groundwater [17, 79–81].

The primary source of Fe in aquifer water can be the dissolution of iron-bearing rocks such as pyrite, siderite, and limonite. Weathering of those minerals in the presence of

organic matter dissolved  $\text{O}_2$  and  $\text{CO}_2$ , and nitrate/sulphate at a particular pH result in an increase in the  $\text{Fe}^{2+}$  concentration in an aqueous medium [82]. The microbial oxidation of released  $\text{Fe}^{2+}$  may be converted to  $\text{Fe}^{3+}$  and reduce the anions by creating an equilibrium state [83]. Various studies [8, 13, 31] have explained the weathering processes through several chemical reactions, such as:



Mn in soil and rocks can be found in diverse phases, such as oxides, silicates, and carbonates. It is also found in adsorbed or exchanging forms on iron oxide surfaces [83]. Similarly, according to some investigations [11, 17], the reductive dissolution of Mn oxides (as  $\text{MnO}_2$ ) with acetate functional groups in acidic conditions occurs according to the reaction (5). For the reverse process, one of the most suitable mechanisms is that the microorganisms first bend and enzymatically oxidize  $\text{Mn}^{2+}$  to  $\text{Mn}^{4+}$  (reaction (6)), which is finally precipitated as  $\text{MnO}_2$  [13].



**3.6. Effect of Water Variables on Fe and Mn Dissolution.** Both Fe and Mn take part in oxidation ( $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$  and  $\text{Mn}^{2+}$  to  $\text{Mn}^{4+}$ ) and reduction ( $\text{Fe}^{3+}$  to  $\text{Fe}^{2+}$  and  $\text{Mn}^{4+}$  to  $\text{Mn}^{2+}$ ) in groundwater through a particular redox environment [8, 12]. Generally,  $\text{Fe}^{3+}$  and  $\text{Mn}^{4+}$  remain in the solid phase as oxides and salt form in the sediment in aquifers [84]. These metals are weathered from metal-laden minerals into groundwater through various complex redox reactions in which  $\text{Fe}^{3+}$  and  $\text{Mn}^{4+}$  accept electrons from oxidizing agents in the aqueous medium. The sequence of those redox reactions is explained by predicted water parameters such as pH, EC, TH, DO, DOC,  $\text{HCO}_3^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $p\text{CO}_2$ . Except for the DOC, the other chemical parameters act as electron acceptors and physical parameters regulate these reactions in separate ways [12, 17, 32]. The rules of those parameters on Fe and Mn dissolution are discussed below:

**3.6.1. pH and  $\text{HCO}_3^-$ .** Including  $\text{CO}_2$ , numerous causes of acidity are present in groundwater systems and affect the oxidation states of Fe and Mn. The impact of pH on Fe and Mn dissolution in the study area is not strongly significant because of the inconsequential correlation of Fe and

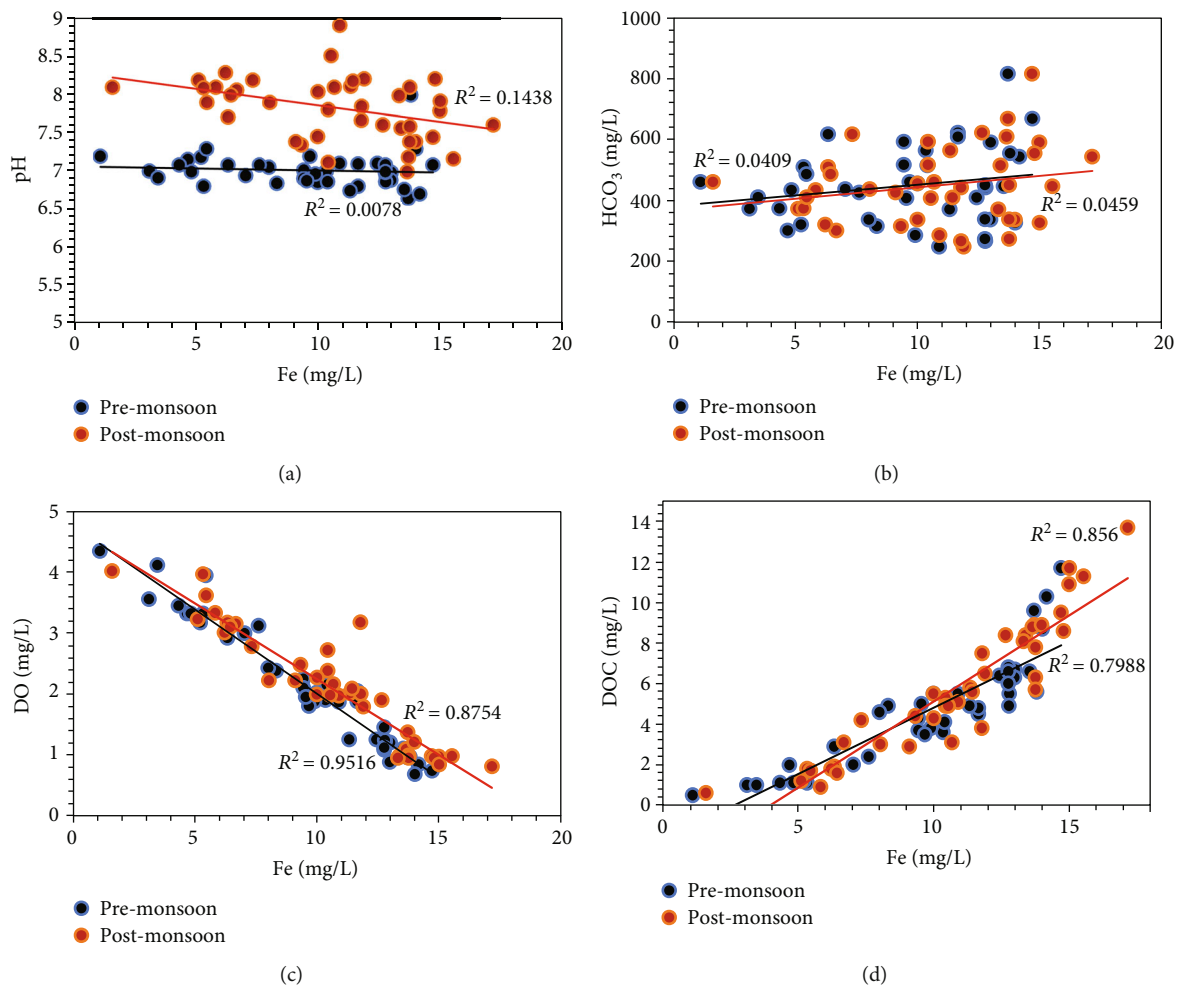


FIGURE 6: Bivariate plots of Fe vs. (a) pH, (b) alkalinity ( $\text{HCO}_3^-$ ), (c) DO, and (d) DOC.

Mn to pH (Table 2 and Figure 6(a)). The overall negative correlation indicates that Fe and Mn dissolution increase with decreasing pH. This is consistent with other investigations that have found that acidic conditions favour the dissolution of both metals [85, 86]. The Fe dissolution rate is lower than that of Mn at a relatively higher pH or in an alkaline medium [73]. Though the average pH of the samples in the postmonsoon season was greater than 7 (alkaline), the concentration of Fe was comparatively higher than that in the premonsoon season, in which the pH values were relatively low (acidic). This is because pH is not the only driving factor that regulates the concentration of Fe in water.

In the Fe dissolution process (reactions (1)–(4)),  $\text{H}^+$  was consumed simultaneously in a neutral to slightly acidic medium. At low pH, this  $\text{Fe}^{2+}$  produces solid phases of  $\text{Fe}^{3+}$ -oxide/hydroxide during the action of reducing bacteria, creating a  $\text{Fe}^{2+}/\text{Fe}^{3+}$  equilibrium [87]. Oxidation of  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$  occurs more slowly at lower pH values, and overall equilibrium is highly impacted by pH [88, 89].

Regarding the Fe and Mn dissolution processes,  $\text{HCO}_3^-$  in groundwater decreases the acidity and increases the probability of metal-complex formation exhibiting dual natural functions [17]. In this study, the significant positive correlation of  $\text{HCO}_3^-$  with Fe and Mn may be owing to the forma-

tion of metal complexes that increase the solubility of the metals [12, 90]. Some soluble  $\text{Fe}^{2+}$  may remain in the water as carbonate complexes [88]. In natural waters containing  $>1$  mEq/L carbonate alkalinity,  $\text{Fe}^{2+}$ -carbonate complexes, such as  $\text{Fe}(\text{CO}_3)_2^{2-}$ ,  $\text{FeCO}_3$ , and  $\text{Fe}(\text{CO}_3)(\text{OH})^-$  are the dominant forms of  $\text{Fe}^{2+}$  [91]. In the studied groundwater samples, the average concentration of  $\text{HCO}_3^-$  is near 7 mEq/L, and a very high loading of alkalinity results in a big chance of forming metal carbonate complexes. The over-excess bicarbonate is not strongly correlated with dissolved iron (Table 2 and Figure 6(b)). Only a negligible amount of total bicarbonate is used for complex formation.

**3.6.2. DO and DOC.** Dissolved oxygen (DO) is one of the most sensitive components for oxidation-reduction assessment. Several reports have shown that oxic environments may happen once DO is  $\geq 0.5$  mg/L [18]. In contrast, a study [92] reported the high opportunity of the reductive environment at DO less than 1 or 2 mg/L. The DO levels of the samples (Table 1) are as sufficient as needed to supply the oxygen for the oxidation of iron and manganese. The average contents of DOC were 4.46 and 5.71 mg/L in separate sampling periods, which were higher than the global average of 3.8 mg/L in groundwater [93]. Dissolved

organic carbon (DOC) levels of less than 1 mg/L are unwanted in groundwater, and then high levels can encourage the mobilization of hazardous heavy elements [94]. Thus, DOC played a significant role in the vast weathering of Fe- and Mn-laden minerals.

Atmospheric oxygen is weakly soluble in water, only  $1.22 \times 10^{-3}$  mol/L at 25°C and 1 atm. Air pressure [18]. This oxygen in groundwater mostly comes from leached rainwater. Due to the coarse sandy soil, the hydraulic conductivity of the study area is very high [95]. Thus, oxygen-rich rainwater enters the aquifers and sufficiently increases the dissolved oxygen (DO) level in the groundwater of the study area. As the favoured electron acceptor, less DO levels suggest that O<sub>2</sub> has been utilized in organic substance decomposition procedures to endorse a reductive environment [32, 96]. An investigation [34] stated that the reduced environment controls the weathering of Fe and Mn-bearing rocks. Figure 6(c) shows a strong negative correlation between Fe and DO in the water samples. Huge oxygen was consumed by both redox reactions,  $\text{Fe}^{2+} \rightleftharpoons \text{Fe}^{3+}$  and  $\text{Mn}^{2+} \rightleftharpoons \text{Mn}^{4+}$ . Thus, lower DO means higher Fe and Mn in the water medium.

Figure 6(d) represents a very strong correlation ( $R^2 = 0.7988$  and  $0.8558$  in PRM and POM, respectively) between DOC and iron concentration in groundwater. The elevated levels of Fe and Mn in groundwater are not caused by dissolved free  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$ . Several forms of dissolved Fe and Mn may be present in water. In the presence of dissolved organic matter, it can strongly maintain  $\text{Fe}^{3+}$  nanoparticles in water and create a suspension. This unstable solution is generated by the combination of the electrostatic (attraction) and steric (repulsion) effects of their negatively charged nanoparticles [31, 97]. In this process, the total iron may bind with the fulvic and humic macromolecules and form soluble complexes [18]. The equilibrium constant for the formation of the  $\text{Fe}^{3+}$  complex with fulvic and humic acids is much higher than the corresponding constants for  $\text{Fe}^{2+}$ . Thus, once these complexes are formed, either by direct contact between  $\text{Fe}^{3+}$  and organic matter (OM) or by oxidation of a preexisting  $\text{Fe}^{2+}$ -OM complex,  $\text{Fe}^{3+}$  can resist precipitation and subsidize the raised level of Fe [98–100]. Like Fe, Mn also forms complexes using the oxidation number of +4 [101, 102]. So, the higher levels of DOC in the samples are used for direct reactions with Fe and Mn minerals and then form a colloidal complex. The turbidity values indicated the formation of a colloidal macromolecule complex. The average turbidity value of the samples was greater than the normal freshwater standard (<5 NTU) (Table 1). The correlation coefficient between turbidity and Fe and Mn was very high ( $r = 0.7$  to  $0.8$  at  $p < 0.01$ ) in both sampling periods (Table 2). Sample ID S38 in PRM and S04 in POM showed the highest values of turbidity with 19.56 and 26.10 NTU; accordingly, these two samples carry the highest amount of iron at 14.01 and 17.17 mg/L, respectively. This result indicated that some Fe remains as a  $\text{Fe}^{3+}$ -colloidal macromolecule complex.

**3.6.3.  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{CO}_2$ .**  $\text{NO}_3^-$  is the more powerful oxidizing agent (just behind oxygen) than  $\text{Fe}^{3+}$  and  $\text{Mn}^{4+}$ , but  $\text{SO}_4^{2-}$  and  $\text{CO}_2$  are weaker oxidizing agents than others,

and the most favourable are reduced first based on this order [8, 17]. This course continues until the electron acceptor and donor have been completely used [103].  $\text{NO}_3^-$  is very stable under oxic conditions [104], and therefore, groundwater may be undergoing  $\text{NO}_3^-$  reduction processes. The concentration of  $\text{NO}_3^-$  was very low in the samples (Table 1). Nitrate and sulphate have weak positive correlations with Fe and Mn (Table 2) and have no chance of oxidation of  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  through reduction itself [11, 105]. Fe and Mn could be bound with  $\text{SO}_4^{2-}$  as Fe- and Mn-sulphide and precipitated together with sulphate reduction [106]. Instead, the elevated levels of  $\text{SO}_4^{2-}$  (Table 1) led to higher salinity that potentially supported the leakage processes of Fe and Mn from soil to groundwater over ion-exchange courses. The higher salinity supports the leakage processes of Fe and Mn from rocks to groundwater, perhaps through the same procedures [10, 107].

The sources of  $\text{CO}_2$  in groundwater samples are percolated atmospheric  $\text{CO}_2$ , dissolution of carbonate minerals, oxidation of organic carbon (OC) through microbial actions in oxic environments, and plant root zone respiration [108]. Carbon dioxide has a very poor oxidizing efficiency and does not participate in the redox reaction of the metal dissolution process [109]. Generally,  $\text{CO}_2$  controls the equilibrium state of Fe and Mn in an aqueous medium through its acid balance capacity [110]. The percolating water plays an active role in rock weathering, especially when it is saturated with  $\text{CO}_2$  [8]. In the study area, the  $\log_{10}(p\text{CO}_2)$  values in groundwater samples vary from -2.46 to -0.88 with an average of  $-1.95(\pm 0.261)$  in the premonsoon (PRM) and -3.51 to -1.26 with an average of  $-2.27(\pm 0.446)$  in the postmonsoon (POM) period (Table 1), which is advanced than the partial pressure of  $\text{CO}_2$  in the atmosphere ( $p\text{CO}_2 = 10^{-3.414}$  or  $\log_{10}p\text{CO}_2 = -3.41$ , or 385 ppm) under equilibrium conditions [57]. Thus, the  $\log_{10}p\text{CO}_2$  value for groundwater is sufficiently high. This saturated  $\text{CO}_2$  reacts with water and produces  $\text{H}^+$  through an equilibrium reaction that is used in Fe and Mn dissolution reactions.

**3.6.4. Salinity and Total Hardness (TH).** Both correlation and factor analysis indicate a strong positive correlation and loading (at  $p < 0.01$ ) of EC with Fe and Mn (Tables 2 and 3). The EC value represents the total salinity of water [111], and higher EC values of the samples were found in the study area (Table 1). Perhaps through ion-exchange processes, high salinity possibly supports the leaching of Fe and Mn from soil minerals to sublayers [13, 85]. Under the stimulus of the salt effect, the rise in EC leads to an upsurge in ionic strength and a diminution in activity coefficient, such that the weathering of Fe and Mn in the inorganic complex may happen effortlessly [8, 18]. A higher EC in the POM indicated a higher concentration of Fe and Mn in the same period (Table 1). The studies suspected that Fe and Mn could form inorganic complexes with existing anions such as  $\text{SO}_4^{2-}$  and  $\text{HCO}_3^-$  in water, thereby increasing their concentration [64].

Especially in the POM season, total hardness (TH) has a strong positive correlation with Fe and Mn in this study (Table 2). It is measured mainly by the concentrations of

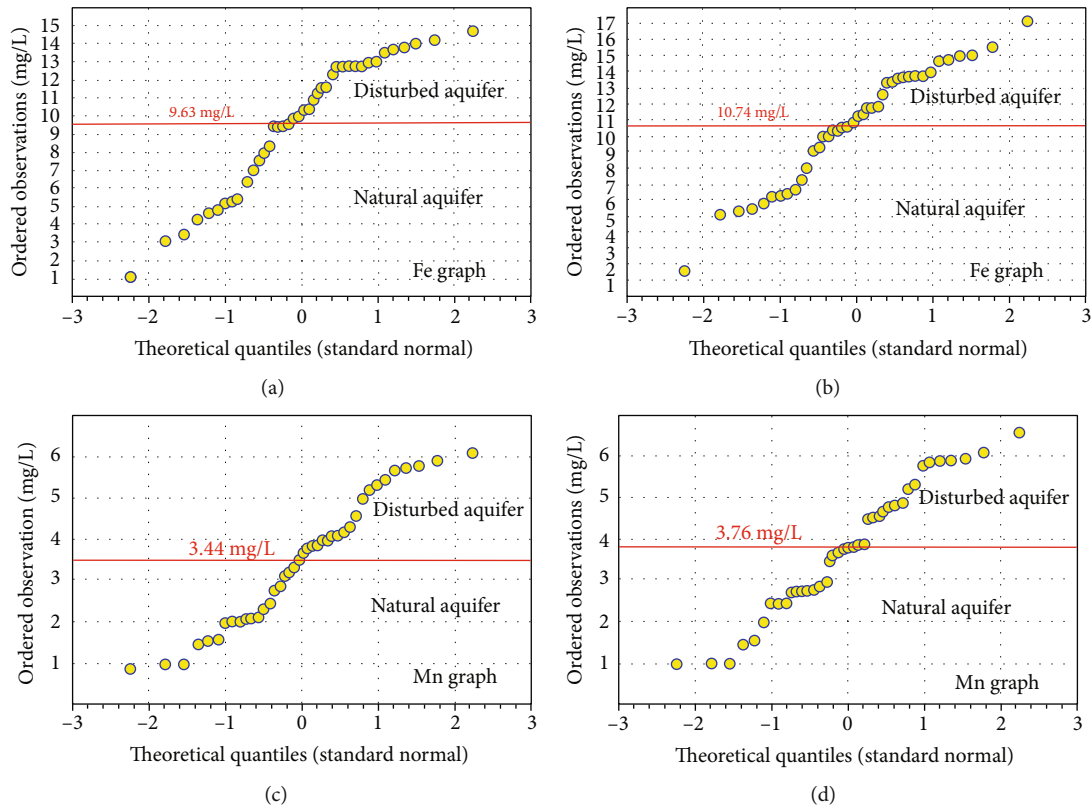


FIGURE 7: Lognormal probability plots of iron (a, b) and manganese (c, d). Proof of identity of the slope changes consistently to the natural background level, NBL (red line).

$\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{HCO}_3^-$ , and  $\text{SO}_4^{2-}$  [112]. The hydrochemical classification of this area shows that samples from the alluvium aquifers have  $\text{Ca-HCO}_3$  facies with frequent enrichments of  $\text{Mg}^{2+}$  and  $\text{SO}_4^{2-}$  [113]. Those facts propose the effect of inorganic complex formations related to  $\text{HCO}_3^-$  because the water was mainly determined by  $\text{Ca-HCO}_3$  type hardness.

**3.7. Natural and Anthropogenic Impacts on Iron and Manganese Concentrations.** The lognormal diagram in Figure 7 helps assess the higher amounts of Fe and Mn delivery connected to the aquifer basement. The transition of the slope of the curve in this figure discriminates whether the metals were present in the groundwater under geogenic conditions or were influenced by peripheral disturbers [114, 115]. In the graph of the lognormal distribution of Fe, samples show high possibilities of naturally taking place in disturbed groundwater at levels of  $<9.63$  and  $<10.74$  mg/L in the PRM and POM periods, respectively. Besides, for Mn, these values are  $<3.44$  and  $<3.76$  mg/L (Figure 7). Statistically, nearly half of the samples (50%) are cited in the externally influenced aquifer area of the graph (upper part of the line). The undisturbed confined groundwater is characterized by relatively lower salinity (EC and TH), lower clay thickness, lower DOC, and higher DO and is located in the southern sector. The disturbance criteria may be either anthropogenic or natural or both. The field study showed that no human activities such as vast industrial activities, no use of iron-manganese-bearing chemical fertilizers in agriculture, no waste

disposal inland, and no landfill were observed, but heavy water mining for irrigation has occurred. So, over groundwater exploitation, substantial fluctuations in river water flow, unbalanced recharge/discharge ratios, and some soil erosion are the main causes of disturbance in aquifer systems. Therefore, along with the physicochemical impact on higher Fe and Mn levels in groundwater, these geogenic and human activities are additional causes for this factor.

The study observed that higher loadings of Fe and Mn may reduce the rate of As dissolution into groundwater, except for any special causes. Very recently, Driscoll et al. [116] investigated the geogenic release of As, Mn, and Fe in the aquifer water of Southeast Asia. They found that drier climatic conditions are related to a tendency toward elevated Mn, while high Fe tends to be found in a more humid climate with high levels of soil organic carbon. Besides, the results confirmed that high Fe levels are often associated with high As levels, which is not similar to the findings of the present study, but areas of high concentrations of Mn and As are often found adjacent to each other. Likewise, the highest Mn and Fe hazards are found in densely populated and alluvial deposits in northwest Bangladesh. Another study [13] in the Quaternary alluvial sedimentary formation of Northeast China showed that in the groundwater, the source of Fe and Mn is the Fe and Mn nodules in the clay layer, whereas it originates from the soil and the whole aquifer in the river valley plain. Another finding of this study is that the TDS, variation in groundwater depths, and

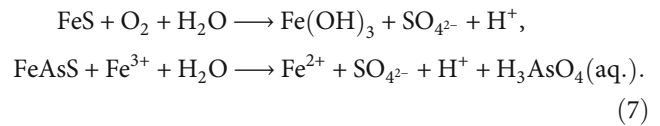
residence time are vital aspects affecting the concentrations of Fe and Mn in water samples. The weathering of Fe and Mn rocks causes As adsorbed on their surface to be unconfined into groundwater.

**3.8. Concentration and Mobilization of Arsenic.** Arsenic is a category-1 carcinogenic element [117] and arises naturally in groundwater supplies throughout maximum parts of Southeast Asia. From 59 out of 64 districts in Bangladesh, over 75 million people were brought to be at risk of potable water contaminated by arsenic [41]. The key source of As in the groundwater was natural since it was confined in the deposits of the shallow Holocene aquifer of the Ganges Delta plain [5]. In the present study, maximum water samples contain less arsenic ( $8 \mu\text{g/L}$ ) compared to the middle part of the country. In this region, over 60% of water samples from 10 districts cross the level of the national guideline of arsenic for drinking water of  $50 \mu\text{g/L}$  or 0.05 ppm [5, 118, 119]. Numerous investigations established that the shallow aquifers in this area enhanced Fe-, Mn-, Al-oxides,  $\text{PO}_4^{3-}$ ,  $\text{NH}_4^+$ , and  $\text{NO}_3^-$  with organic material containing moderately high levels of arsenic [120–122]. In this study, a weak correlation of arsenic with influencing components in water (e.g., Fe, Mn,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , DO, and DOC) was observed (below 0.4 at  $p < 0.05$ ) (Table 2). Iron and manganese directly take part in the dissolution and distribution of arsenic in the aquifer system. At an elevated level of Fe and Mn in aquifer water, lower levels of arsenic are expected [38]. Oxidizing agents convert  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$ , which forms a solid phase of  $\text{FeO}(\text{OH})$ . Arsenic (+3 and +5) could be adsorbed on the solid surface of  $\text{FeO}(\text{OH})$ , and then both are precipitated [119]. A previous study by the authors confirmed that the abovementioned factors affect arsenic mobilization, but this influencing empowerment mostly depends on the local lithological conditions [41]. If the lithology is heavily clayey, peaty, and silt-clay in nature and has a rich bacteriological community, then the factors such as Fe, Mn,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , DO, and DOC may show the action properly, otherwise not [123]. The present study area consists of sandy, silt, and silt-loamy land, which disfavours the action of those influencing factors.

The approach of occurrence and mobility of As in sedimentary aquifer may be measured by complex interactions of microbially facilitated actions and hydrogeochemical processes sensitive to site-specific lithology and sediment category, as well as manmade actions, for instance, the use of agrochemicals. Numerous investigations have been conducted in the last few decades but have been unsuccessful in exploring the mobilization mechanisms of As in the Ganges basin areas. Over time, four mechanisms have been suggested to explain these processes of contamination of the Bengal delta plain by arsenic:

- (1) *Oxidation of pyrite.* Near the year 2000, several researchers, e.g., Chowdhury et al. [124], Bhattacharya et al. [125], and Nickson et al. [126] assumed that arsenopyrite and iron-pyrite were oxidized by the entry of atmospheric oxygen into aquifer sediments and then As was released as arsenate into

the groundwater of the Ganges basin



The reasonableness of this hypothesis is not correct since the groundwaters in this area are (a) anoxic type, with insignificant DO; (b) at near-neutral pH or reasonable alkalinity; (c) characterized by low  $\text{SO}_4^{2-}$  levels with no relationship to As; and (d) categorised by the very low level of  $\text{NO}_3^-$ . This process was suspended at that time, and the following methods were established later.

- (2) *FeOOH reduction.* Microbial metabolism of DOM in the aquifer leads to anoxic environments, therefore leading to the reduction of lepidocrocite [ $\text{FeO}(\text{OH})$ ] rock and then resultant in the release of confined As into the groundwater [38, 117, 127]
- (3) *Redox process.* As is unconfined by the reductive weathering of arsenic-rich  $\text{FeO}(\text{OH})$  rock, and it is a redox procedure that happens after bacterial oxidation of organic material has used up DO,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  [38, 39, 118]
- (4) *Ion exchange.* The arsenate ion ( $\text{AsO}_4^{3-}$ ) adsorbed to aquifer rocks is evacuated into the water medium by a modest exchange of  $\text{PO}_4^{3-}$  after As contamination in groundwater [39, 119–121]

The abovementioned processes of 2 to 4 were recognized after the 2000s. Along with lately published articles on adequate microbial communities, the accessibility of DOM, an abundance of highly positive charge-baring metal rocks, a higher  $\text{PO}_4^{3-}$  concentration, and a low value of redox potential (Eh) with moderately high pH in aquifers are the main criteria to increase the As concentration in shallow aquifer systems. The study area is situated in the northwest position of the country and holds very fertile cropland relative to another region of Bangladesh. Approximately 72% of total lands are arable, of which 30% are triple-cropped areas [44]. The largest part of cultivable farmland is loamy, sandy, or sandy-loam; however, clayey or silt-clay formations were not found in this area. A previous study [128] confirmed that the soils of these areas comprise less organic matter and that the Eh values of the soil are comparatively high. But this area is free from any As toxicity, though the chemical manure consumption rate and water mining are very high. It is assumed that in the study area, other influences, for example, soil DOC, pH, and redox potential (Eh), which are the crucial operators of As dissolution, favoured less As contamination. Some investigators [129, 130] assumed that water depths are drawn down by vast irrigation and allow atmospheric  $\text{O}_2$  to penetrate the aquifer, which may quicken the release of As by oxic oxidation of pyrite rocks. This statement is not true for the study area because irrigation activities are very common in these zones, and in hot summers,

the groundwater level suggestively deteriorates owing to the overmining of groundwater. Nath et al. [131] and Reza et al. [132] designated the comparative role of mobilizing agents in the utmost and least As-affected parts of Bangladesh. This study supported their work and reported that in the least affected areas, the lower level of As is not subjected to  $\text{PO}_4^{3-}$ , dissolved organic matters, and soil alkalinity and As released from the soil with low level owing to less desorption of As or settled out of  $\text{FeO}(\text{OH})$ . The low concentration of detection suggests that sorption of As on iron-oxide precipitates, or other solubility controls, may limit its dissolution in groundwater.

**3.9. Impact of High Fe and Mn on Irrigation and Aquaculture Water Quality.** Over 70% of cultivable land in the study area is under irrigation with shallow groundwater through an engine pump. The high contents of Fe and Mn in groundwater may adversely impact irrigation water as well as the soil environment, which impacts plant growth and production. Although  $>5$  and  $>2$  mg/L of Fe and Mn are undesirable in irrigation water [133], potential problems due to high Fe in irrigation water mostly center around the staining of concrete surfaces, plugging of irrigation equipment and drainage tiles, and unsightly deposits of rust-colored material on ornamental plants. High Fe concentrations in spray tank water can clog screens or nozzles and negatively affect the performance of some pesticides. If concentrations are very high (over 10 mg/L) and iron-laden water is used frequently and continuously, some nutrient deficiencies may result. The maximum number of water samples in the study area are highly mineralized and contain huge amounts of soluble salts. Increasing the level of dissolved salts in the soil raises the osmotic pressure of the soil solution, and in that situation, plants cannot adapt effortlessly. On the other hand, in general, Fe is not lethal to plants in fizzy soils, but additional soil iron can make it rigid for plants to absorb other essential salts such as P, Mg, and Mo [26]. Plants need P for cell division and photosynthesis. Plants need Mg to yield chlorophyll, which gives them their green color. With elevated concentrations of Fe in soil, plants miss out on these vital minerals and can expect the plants to experience staining, distress-controlling photosynthesis processes, and weak roots. Unbalanced concentrations of numerous other significant elements, like Zn and Cu, are also probable with too much Fe present. Concerning Mn, it is toxic to several crops at 1 or less than 1 mg/L, but typically only in acidic soils [26]. Especially in the premonsoon season, samples from the study area have lower pH values, increasing Fe and Mn toxicity to plants.

In aquaculture waterbodies, if the sediments become anoxic, precipitates of  $\text{Fe}^{3+}$  can be reduced into  $\text{Fe}^{2+}$ , leading to a darker coloration of the layered sediment. Though Fe can also be helpful by mitigating the toxicity of  $\text{H}_2\text{S}$ , formed by anaerobic bacteria, by precipitating it into insoluble  $\text{Fe}_2\text{S}_3$ , if water containing a high level of  $\text{Fe}^{2+}$  is used in hatcheries, the formation of  $\text{Fe}(\text{OH})_3$  can be problematic. This precipitate can settle on eggs and damage them, and it can clog the gills of fish. It has been proven that in many

aquaculture practices, the presence of Fe at levels above 0.1 mg/L will damage the gills of fish [134]. This level is much lower than the concentration usually reported in the literature (see Table 1). The adverse impact of Fe will be subject to the species and size of the fish. The gills of the fish act as a mechanical filter, and small particles of Fe with dimensions of a few microns become stuck in the gill lamella. The presence of these particles irritates the gill tissues, leading to gill injury and secondary bacterial and fungal infections. Fe acts as a catalyst in water and will inhibit the dissociation of  $\text{O}_2$  in water to form a very reactive free radical. However, on the surface of the gills, the free radicals formed by Fe will cause oxidation of the nearby tissue, which will lead to massive demolition of the gill tissue and anemia. Manganese has a low toxicity relative to iron, and the bioaccumulation rate in fish bodies is very low [135].

#### 4. Conclusion

This study identified the environmental issues responsible for elevated levels of Fe and Mn in the calcite-type groundwater of the Ganges River basin. The results revealed that the levels of Fe and Mn ( $\sim 10$  and  $\sim 3.5$  mg/L, respectively) in groundwater crossed the national and international acceptable levels but not As for all the samples. Including the geological and soil formation of the study area, the influencing features on elevated Fe and Mn concentrations in groundwater are water acidity or alkalinity, DO, DOC,  $p\text{CO}_2$ , and some anions. The high salinity (EC) of the samples is supposed to foster the leaching of Fe and Mn from soil and minerals as ionic forms through ion-exchange processes. It demonstrated that the sources of Fe and Mn in the river valley plain were both metal-bearing nodules in the thick clay layer and tidal mud, while on the upper platform, these metals originated from the consolidated silty soil and whole aquifers. According to seasonal variation, changes in Fe and Mn concentrations in samples were observed. The correlations of redox-sensitive variables with these two metals illustrated that heavily mineralized groundwater has a less significant impact than reductive environmental conditions upon the dissolution of Fe and Mn from their minerals. Studies have demonstrated the substantial possibility of forming Fe and Mn complexes with organic and inorganic substances. Moreover, the lognormal curves of Fe and Mn were effectively used to differentiate two groundwater conditions in the study area. Nearly half of the samples (50%) are cited in the externally influenced aquifer areas. Excess groundwater mining, significant fluctuations in river flow, and some soil erosion are identified as the main causes of disturbance in aquifer systems, which are found in the northern river valley part of the study area. The higher weathering rate of Fe and Mn minerals causes arsenic to be adsorbed on them, which leads to less release of As from arsenic-laden rocks into groundwater. This study provides a basis for the rational consumption of groundwater and information concerning higher Fe and Mn contents in groundwater. Deep wells should be drilled in river valleys, avoiding areas with a thick clay layer.

## Data Availability

Data are available on request, and the corresponding author (M.G. Mostafa) will provide if someone is interested.

## Conflicts of Interest

Both authors declare that there is no conflict of interest regarding the publication of this article.

## Authors' Contributions

M. S. Islam was responsible for the conceptualization (lead), investigation (lead), data curation (lead), formal analysis (lead), methodology (equal), software (equal), resources (equal), supervision (supporting), visualization (supporting), validation (supporting), writing—original draft (lead), and writing—review and editing (supporting). M. G. Mostafa contributed to the conceptualization (equal), methodology (equal), investigation (equal), resources (equal), project administration (equal), validation (equal), software (equal), supervision (equal), and writing—review and editing (equal).

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