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Assessment of the status of groundwater arsenic at Singair upazilla, Manikganaj Bangladesh; exploring the correlation with other metals and ions

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Abstract

Comparative study was conducted to correlate Arsenic (As), Iron (Fe), Copper (Cu), Manganese (Mn), Calcium (Ca^{2+}), Magnesium (Mg^{2+}), Potassium (K^{+}), Nitrate (NO_3^{-}), Phosphate (PO_4^{3-}), and Ammonia (NH_3) by determining their concentration at different depth of the tube-wells in the selected study area at Singair, Manikganj Bangladesh. Total 99 tube-well water samples were collected from the study area. In most of the sampling points the present concentrations of **As** were less than the previous concentrations. The correlation between As and Fe was positively significant. It can be suggested possible adsorption/coprecipication of As with Fe in shallow aquifer. However, the relationship between As and Mn was not significantly observed. On the other hand, relationship between Cu and As showed a positive significant correlation. The correlation between As and PO_4^{3-} was also significant, although the correlation between As and NO_3^{-} was not significant. PO_4^{3-} may be comes from phosphate fertilizers application and can be a contributer of As in the shallow aquifer. The PCA biplot also indicated the significant relationship between As, Cu, Fe and PO_4^{3-}. Excessive withdrawal of tubewell water along with aquifer dynamics along with ionic interference might be responsible for the mobilization of As in the study area.

Keywords: Groundwater; Chromatography; Contamination; Arsenic geochemistry; Shallow tubewell; Arsenic mobilization.

Introduction

Geochemical cycling of arsenic (As) in the water ecosystems is increasing gradually, which provokes anxiety because of its severe toxicity effects for the human. Ingestion of inorganic As can cause both non-cancer health effects and external and internal cancers in the human body (WHO, 1981; Smith et al., 1992; Tondel et al., 1999). Since the 1970s, the World Health Organization (WHO) has drilled more than two million tube-wells in Bangladesh for the population to have a safe and bacteria-free drinking water supply. Unfortunately, under certain conditions, the groundwater frequently contains elevated levels of natural arsenic, greater than the WHO's maximum contaminant level (MCL) of 0.01 ppm (equivalent to 0.13 mM), and often greater than the Bangladesh's MCL of 0.05 ppm (0.66mM). As, a notorious bio-accumulating poison, has been adversely affecting the health of millions of people in the Bengal Basin (Bangladesh and the West Bengal State, India) for the last twenty years (Das et al., 1995, 1996; Mandal et al., 1996; Dhar et al., 1997; Karim, 2000; Smith et al., 2000**)**. The Bengal Basin lies within in the floodplain of the Ganges and Brahmaputra Rivers. As one of largest river in the world, the GBR is the single largest sediment flux and the fourth highest water discharge to the oceans **(**Milliman and Meade, 1983**)**. The groundwater As occurrences (higher than permissible limit 0.05 mg/L) in 52 out of 64 districts (total area of 118,012 km^2) of Bangladesh were considered as the biggest As calamity in the world **(Karim, 2000).** Arsenic is extensively cycled at the earth surface and has a complex dynamic biogeochemistry. It has multiple oxidation states and it is present in aquatic environment as both in organic and inorganic compounds, which may be interconverted through chemical and biological activity in the natural environment (Ferguson and Gavis, 1972). One of the major sinks of As in aquatic environment is the iron rich sediments (Peterson and Carpenter, 1986) but absorption of arsenic in Mn-oxides has also been reported (Takamatsu *et al*., 1985). Once it is in the sediments, as diagensis can be largely controlled by Fe, Mn and organic matter redox reactions (Branonon and Patrick, 1987). Moreover, if As aquifer has a porosity of 25% and the sediments contains 1 mg/kg of labile As, the complete dissolution of that As would lead to a groundwater As concentration of 7.95 mg /L far in excess of any drinking water standard (BGS, 2001 and DPHE, 2000). Accumulation of As in food crops poses potential health threat to humans. A number of studies have already been reported on As concentrations of soil in Bangladesh. Ahsan et al., **(**2009) reported that the range of As in floodplain soils from Faridpur district of Bangladesh was 18-65 mg/kg whereas the range of As in Dhamrai and Manikganj soils was 3.1-8.9 mg/kg. Contamination of naturally occurring As has recently become an alarming environmental problem in deltaic plain of Ganges-Meghna-Brahmaputra River in Bangladesh and West Bengal (Nickson *et al*., 1988). It is now well documented that more than 35 million peoples in Bangladesh are under detrimental health threat of As poisoning (DH**C** report, 1997). In addition, there have some studies reported that agricultural fertilizers and organic wastes may promote As mobilization by ion-exchange with phosphorus derived from fertilizers.(Acharya et al., 2000; Chowdhury et al.,1999; Anawar et al., 2006 and Brömssen et al., 2008)

Although serious **As** contamination of groundwater in Bangladesh has been frequently reported, there are few articles that discussed the relationship between As, and other common metals and nutrients in the groundwater. In the present study, concentration of As along with other commonly occurring heavy, alkaline earth metals and nutrient ions such as, total $Fe^{2+ \text{ and } 3+}$, Mn^{2+}, Cu^{2+}, Na^{+}, K^{+}, Ca^{2+}, Mg^{+2}, NO_3^{-}, PO_4^{3-}, and NH_4^{-} were determined in the groundwater of the contaminated site. The co-occurrences of these metals and nutrient ions with As were discussed in terms of correlation matrices and step wise linear regression models to focus their competitive effects on the release of As in the groundwater of Manikganj district, Bangladesh.

Materials and Methods

Singair Upazilla (Manikganj district) with an area of 217.38 km^2, is bounded by Dhamrai and Manikganj sadar upazilas on the north, Nawabganj (Dhaka) upazila on the south, savar and keraniganj upazilas on the east, Manikganj Sadar upazilla on the west (Fig. 1). Main rivers are the Dhaleshwari, Ghazikhali and Kaliganga.The geographic location of Singair Upazilla is between 23°51´ north and 90°21´ east **(Banglapedia**, **2006).** Ground water samples were collected from the same shallow tube-wells as collected by Uddin et al., 2006 in the study area and the GPS locations were also recorded for GIS mapping (Fig.1). For this study, a total of 99 ground water samples were collected from tube-wells with different depth from the surface of Singair Upazilla from August 23, 2010 to August 26, 2010. The approximate length of the sampling area was 6.5 Km. Samples were collected in 250 ml polypropylene plastic bottles. Acidification was carried out by 2 drops of concentrated HNO_3 to prevent precipitation of dissolved iron as well as adsorption of trace metals on to the bottle surface before filtration. All of the samples were filtered with 0.7 μm glass fiber filters (GF/F Whatman) and diluted before analysis, and sampling bottles were carried in dark containers and stored in freezing condition until analysis.

Analytical Methods

For the analysis of phosphate (PO_4^{3-}) ion, a phosphate analyzer of ASCSII with wavewlength of 880 nm was used. Concentrations of anions and cations were determined by Ion Chromatography method (DX-120, Dionex, USA) using the columns "Ion Pac AS12A" (Dionex, Abbreviation of state, USA) for anions and "Ion Pac CS3" (Dionex, Abbreviation of state, USA), for cations. The inductively coupled plasma (ICP-OES) optical emission spectometer was used (SII Nano Technology inc.) for the determination of **As, Fe**, **Mn,** and **Cu**. The water quality analyses were carried out in the laboratory of ecosystem studies, school of environmental science, The University of Shiga prefecture, Japan. Correlation matrix and Pricipal component analysis (PCA) was done using Origin 9.0 software (OriginLab, USA) and rest of the statistical data analyses was done by MS-excel 2007.

Results and discussion

Concentration of arsenic and other compounds in the study area

The Singair upazilla stands on the active deltaic region on the Ganges basin and the deltaic areas of Bengal basin receives large amounts of land derives and land weathered organic matter rich sediments with high concentrations of trace metals (Tareq et al., 2003). The results of water quality analysis of the collected samples are presented in Fig. 3(a, b). The mean concentration of arsenic was 0.02 mg/L, ranging from less than detectable limit (BDL) to 0.113mg/L, as of the samples collected in August 2010. Among the total sampling point (99), 51 tube-well water contained different levels of As. Where as the previous mean **As** concentration of the study area was 0.08 mg/L, ranging from BDL mg/L to 0.7mg/L in unpublished data **(Fig. 2)** by Uddin et al. (2006), in addition, **As** concentrations of 85 tube-wells out of 102 tubewells exceeded WHO Guideline for Drinking Water Quality (0.01 mg/L) value. Interestingly, in the present study out of 99 tube-wells 50 tubewells were exceeded WHO standard, 47 tubewells showed not detected (BDL) and only 2 samples were below the WHO standard for drinking water. Furthermore, 13 samples exceeds the Bangladesh domestic water quality standard (0.05 mg/L) for As in drinking water. The unpublished data revealed that the highest concentration of **As** was 0.7 mg/L, although the highest concentration of **As** was found 0.113mg/L in the present study. So concentration of As in the present study was lower than that in previous**.** The socio economic study states that there is no mining region, any manufacturing or anthropogenic source of **As** in the study area. The variation of this concentration may be due to the geo-environmental settings in the area. **As** concentration from the Padma–Jamuna confluence along with the possibility of co-precipitation with other metals. Moreover, the mean concentration of Mn, Fe, and Cu were 0.46 mg/L, (0.054-2.72 mg/L), 0.81 mg/L (BDL-19.29 mg/L), 0.002 mg/L (BDL-0.013 mg/L), respectively. The mean concentrations of Mn and Fe were above the WHO Guideline for Drinking water Quality (GDWQ) value for Mn (0.1 mg/L) and Fe (0.8 mg/L). The mean concentrations of NO_3^-, PO_4^{-3}, NH_3, Na^+, K^+, Mg^{2+} and Ca^{2+} were 253.18, 0.10, 10.75, 88.80, 4.14, 49.43 and 195.98 mg/L, respectively. Only 3 samples contained Cd. The concentrations of NO_3 and Na^+ showed higher concentrations mentioned by WHO GDWQ. The high concentration of As, Fe, Mn and PO_4^{-3} may be due to the oxic to sub-oxic conditions of the shallow aquifer of the study area. This result can reflects that the reductive dissolution of iron oxyhydroxide throughout the Holocene alluvial aquifers of Bangladesh and West Bengal, India are driving factor for the mobilization of As (Anawar et al., 2011 and Nickson et al., 2000)

Correlation of As with elements by PCA

Correlation matrix was calculated in order to search for the relationship between As and other substances including iron (Table 1). The correlation matrix showed the significant positive correlation **(r=0.34)** between arsenic (**As)** and iron (**Fe)** in groundwater in the study area. When the **Fe** concentrations were 0.134, 0.091, 9.262, 1.843 and 19.296 ppm, the **As** concentrations were 0.025, 0.056, 0.041, 0.044 and 0.113 ppm, respectively (fig.3). Many authors found that the As mobilization in groundwater of the Bengal delta and other parts of the world is associated with arsenian pyrite (Peters et al., 1999; Raessler et al., 2000; Schreiber et al., 2000). In this study the positive but weak relationship of Fe with As concentrations confirms the findings of Safiullah (1998) and McArthur et al. (2001). In groundwater of the Bengal delta, the poor correlation between Fe and total As concentration is attributed to the loss of As by precipitation of Fe–As sulfides, a precursor to arsenopyrites, and siderite ($FeCO_3$) (Nickson et al., 2000), and this can be a clue on the possible mechanisms controlling As mobilization. On the other hand, **As** and **Mn** showed significant positive but very weak correlation **(r=0.02).** The reduction of Fe and Mn oxides, a governing process for As mobilization has not been supported by this study is due to weak/very weak relationship among As, Fe and Mn (Anawar et al., 2003 and Nickson et al., 1998) In addition, Cu and As also showed significant positive correlation **(r=0.49).** On the other hand As-PO_4^{3-} showed significant correlation **(r=0.32).** High concentrations of As, especially its reduced species of As(III) along with Fe, DOC (dissolved organic carbon), NH_4^+ and PO_4^{3-} and low concentrations of dissolved oxygen (DO), NO_3^- and SO_4^{2-} in groundwater, indicating reducing condition in the Holocene aquifer, have resulted in suggestions that reductive dissolution of Fe(III)-oxyhydroxides is the primary mechanism for the release of adsorbed and co-precipitated As (Harvey et al., 2002 and Anawar et al., 2003)

Principal component analysis (PCA) is a versatile tool of factor analysis, which can reduce the total number of variables to few factors with most of the variation in the original sets of data and simplify the interpretation (Hair et al., 1998). The number of significant principle component is selected as considering Kaiser Criterion (Kaiser, 1960). PCA on the combined datasets provided two factors with Eigenvalue >1 that can explains approximately 50% of the variability of the data (PC 1 variance of 27.79% and PC 2 variance of 21.58%) (Table 2).

From the biplot analysis of PCA (Fig. 4), when two variables are far from the centre and present closely to each others the closely present variables are said significantly positively correlated (r=1). Fig. 4 depicts the PC where Ca^{+2}, Mg^{+2}, Na^{+}, NH_3 and K^{+} may represented for PC 1, and on the other hand, As, Cu, Fe and PO_4^{-3} belongs to PC 2. From the Fig. 4 As is significantly correlated with Cu (r= 0.49), Fe (r= 0.34) and PO_4^{-3} (r= 0.32); however, there is no significant correlations between As-Mn (r= 0.02), and insignificant negative correlations between As and Na^{+} or K^{+}. There was a good correlation between As-Mn, in addition, correlation between As-Fe was also found in the fine particle core sediment sample by Anawar et al. (2003). However, in this study, a significant positive correlation between As-Fe had existed but another study (Ohno et al., 2005) showed no significant correlation between these two metals. However, NO_3^{-} did not show correlation between two principle components except with Fe (r= 38) and PO_4^{-3} (r= 37).

The mechanism for the accumulation of As in the ground water is adsorption on and co-precipitation with hydrous oxides **(Aggett and O'Brien, 1985).** The As derivatives ($H_2AsO_4^{-}$, $HAsO_4^{2-}$ and H_3AsO_3) in contaminated ground water can easily be adsorbed on to hydrous iron oxides that have a high pH and follow this reaction.

$Fe(OH)_3 + H_3AsO_4 = FeAsO_4.2H_2O + H_2O$ ………. (1) **(Kiniburgh et al., 1976)**

The Fig.5 revealed the stepwise linier regreesion between Fe-As, Mn-As, and Cu-As, and revealed there were no significant relationships among them with very small r-square value. Fig. 6 also showed the regression with As and onther ions with very small r-square value except with PO_4^{-3}. Thus the dissolved As can be declined to a very low concentration within a short distance from its source. The BGS and DPHE technical report **(DPHE 2000)** have drawn the As Vs Fe map of ground water for the whole country. They reported that often these 2 elements can be strongly correlated. Indeed, on a well by well basis, the **Fe** concentration in well water generally provides a poor predictor of **As** concentration. It is clear that Fe oxides are closely associated with the development of high **As** ground water in Bangladesh.

The correlation matrix among As and various ions is summerised in Table 2. A significant correlation revealed between As and PO_4^{-3} **(r=0.32).** Like As, PO_4^{-3} is strongly adsorbed by amorphous iron oxide and is released under anaerobic condition **(Jacobs *et al.*, 1970).** Other anions were less effective in displacing As. The order of effectiveness decreasing for As (V) of $H_2PO_4^{-} > H_2AsO_4^{-} > SO_4^{2-} > CO_3^{2-}$ and for As (lII) of $H_2PO_4^{-} > H_3AsO_3 > F^{-} > SO_4^{2-} > CO_3^{2-}$. **Acharyya et al. (2000; 2005)** hypothesized that As anions sorbet to aquifer sediments were displaced to solution by competitive exchange with phosphate from the fertilizers. The correlation of As and NO_3^{-} revealed negative but very weak **(r=-0.02)**, and on the other hand**,**

Na^+ and K^+ showed negative weak correlation (**r=-0.15, r=-0.18)**. As shown in Fig. 6 showed the stepwise linear regression graph between Mg^{+2}-As, NH_3-As, K^+-As, Ca^{2+}-As, NO_3^--As and PO_4^{3-}-As. They posses insignificant relation with **As**.

Conclusion

The study revealed that the mean **As** level in ground water in study area was lower compared to the previous. Excessive withdrawal of ground water might be the reason towards the findings of the study of **As**. However, due to mobilization of **As**, it might be deposited to the lower portion of aquifers. Chemical weathering of biotitic and other basic minerals in the Holocene aquifer could be a primary cause of As mobilization in ground water because of positive correlation of As concentration with some nutrient elements. Iron (Fe) may come from ferro hydroxide minerals, pyritic material and other rock forming primary minerals. For hunting safe and **As** free pure water we need to know the correlation between As and other trace elements. Correlation of **As** with **Fe** and **Mn** may also give an idea about the carrier of these elements. It means that **Fe** and **Mn** are great adsorbent of **As** in solid phase, and ultimately causing the mobilization of As along river flow. However, although in this study the correlation between As and Fe was significant, that between As and Mn showed an insignificant correlation. The concentration may increase or decrease due to some biogeochemical and mechanical factors. It is hard to say about concrete indicator of As in groundwater using metals and other nutrient ions.

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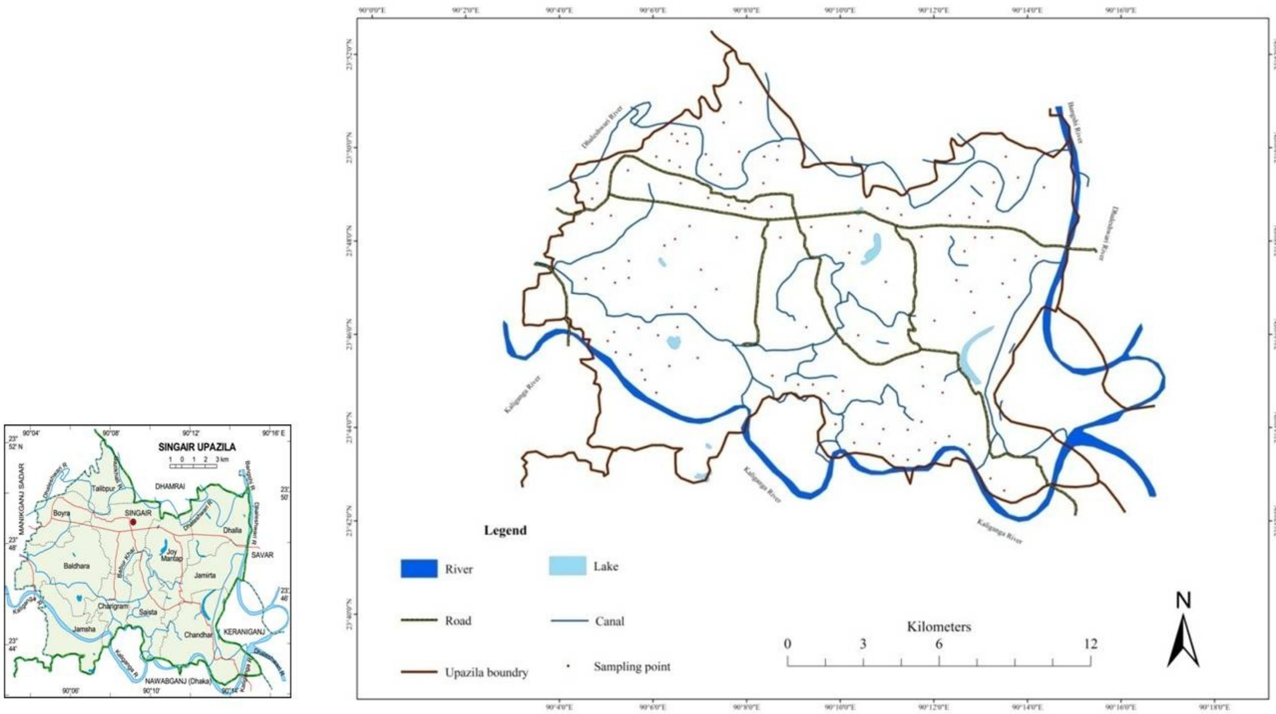


Fig.1 Map of Singair, Manikganj Bangladesh (*Source: Banglapedia, 2006)*
and Location of the sampling points in the GIS map

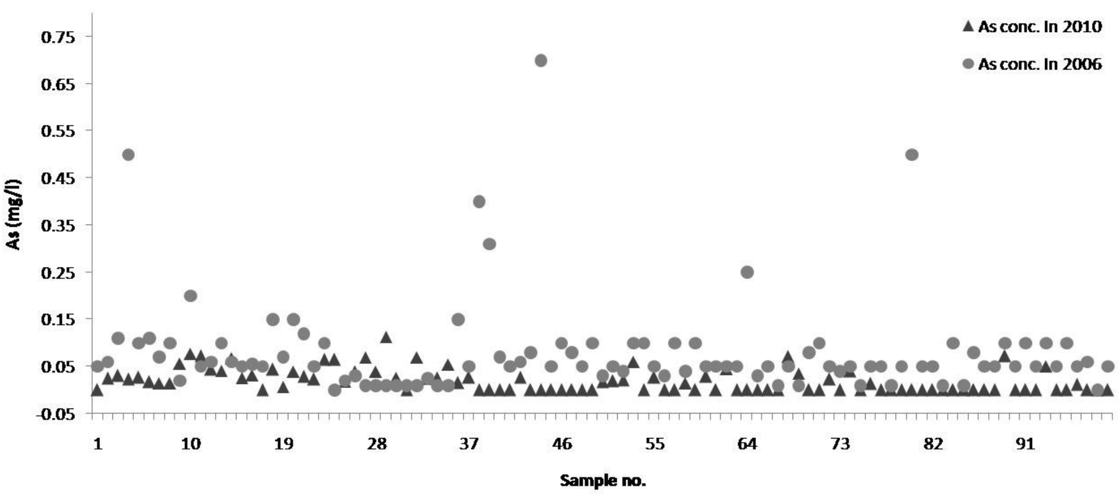
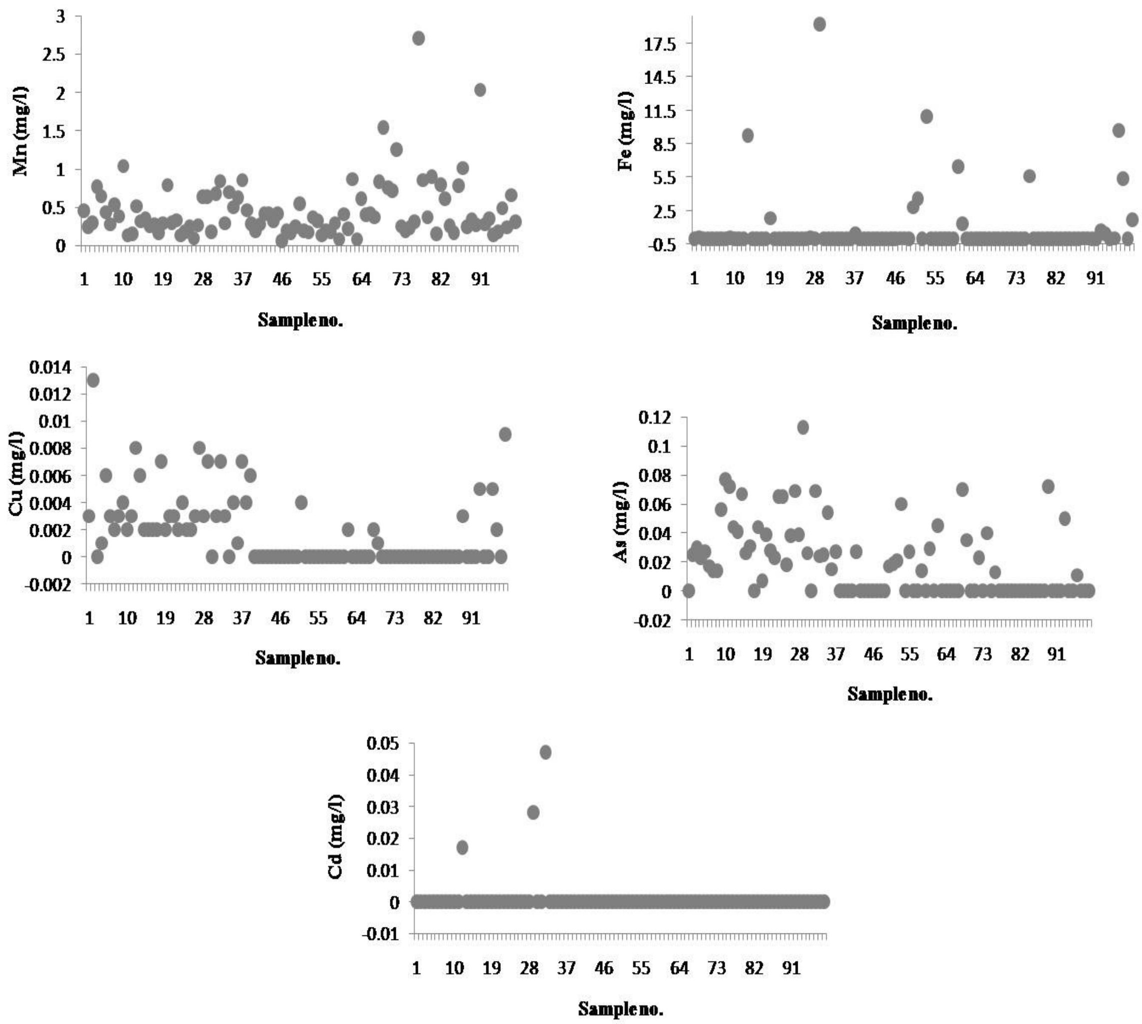


Fig.2 Concentration ranges of As (mg/l) in the study area in 2006 (Uddin et al 2006) and 2010.

a.


Mn (mg/l)
Sampleno.
Fe (mg/l)
Sampleno.
Cu (mg/l)
Sampleno.
As (mg/l)
Sampleno.
Cd (mg/l)
Sampleno.

b.

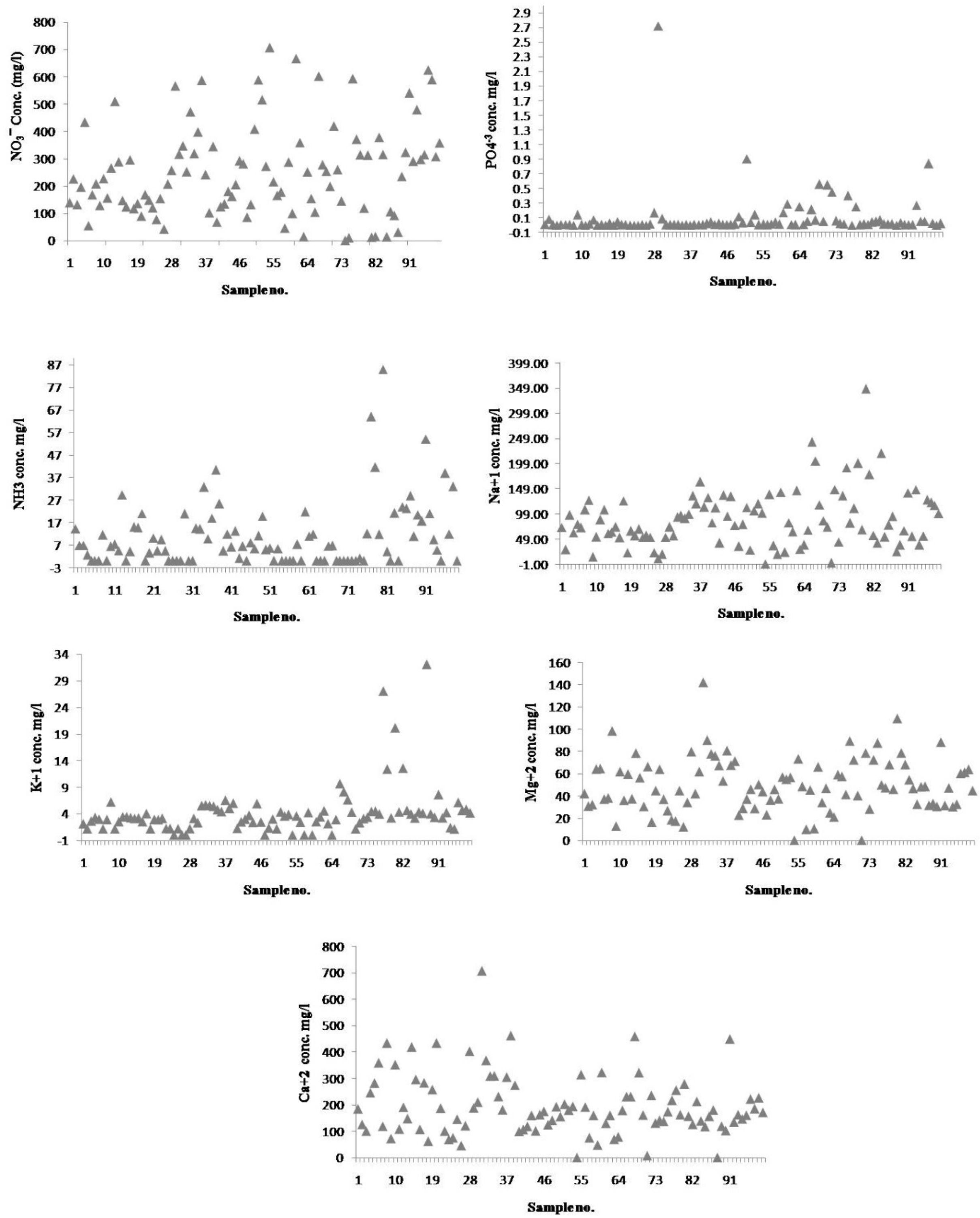


Fig.3 The range of concentration of a. metals (Mn, Cu, Fe, As and Cd) and b. ions (NO_3, PO_4, NH_3, Na^+, K^+, Mg^{2+} and Ca^{2+}) in different tube-well water samples in the study area.

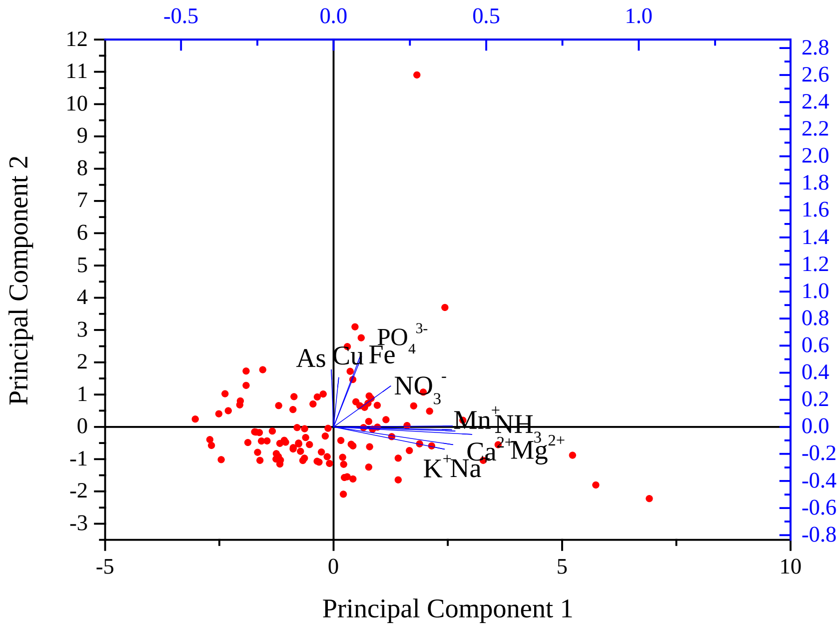


Fig.4 PCA biplot based on correlation matrix

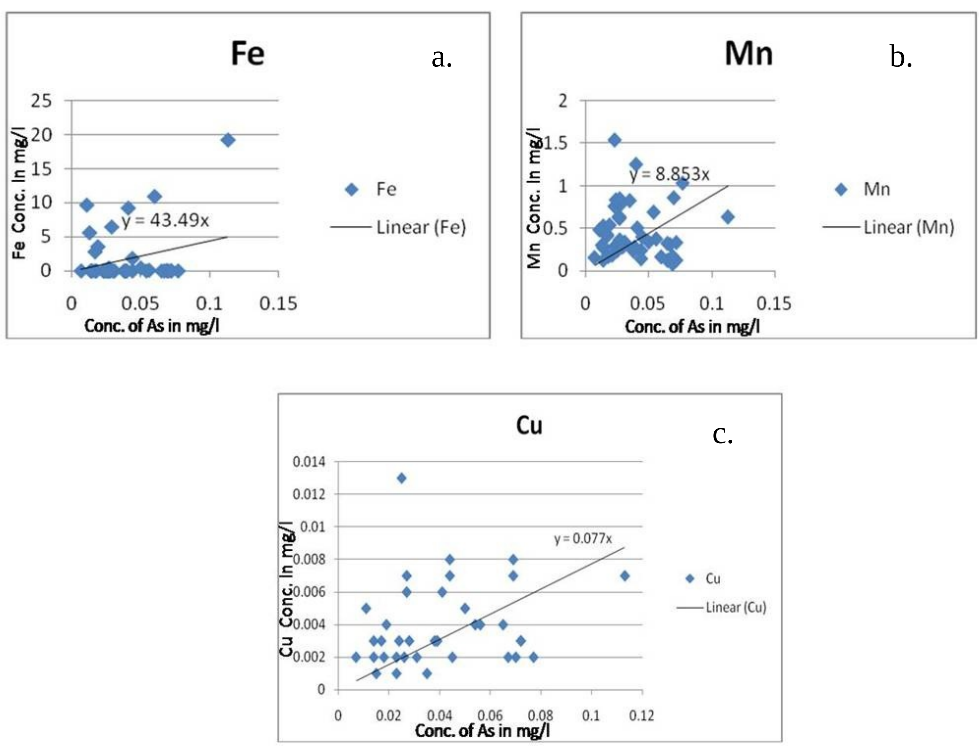


Fig.5 Stepwise linear regression; a) Fe Vs As, b) Mn Vs As and c) Cu Vs As coefficients were insignificant with r^2 value 0.079, 0.001 and 0.040 respectively.

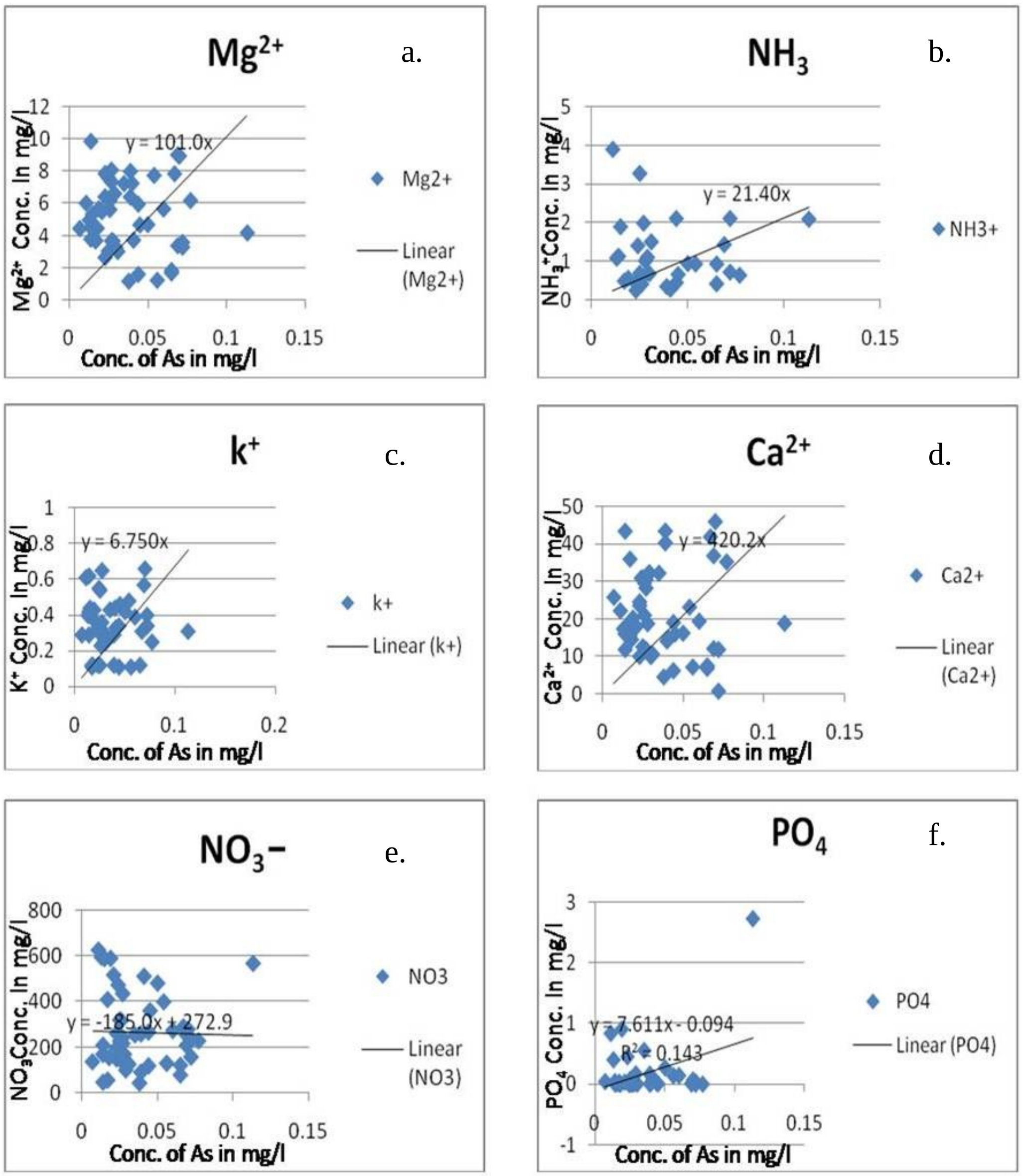


Fig.6 Stepwise linear regression; a) Mg^{2+} Vs As, b) NH_3 Vs As, c) K^+ Vs As d) Ca^{2+} Vs As e) NO_3^- Vs As f) PO_4^{3-} coefficients were insignificant except PO_4^{3-}.