Source of arsenic bearing detrital minerals in shallow aquifer of southeastern Bangladesh

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ABSTRACT: Forty seven (47) sediment samples from two bore holes from different depths and two river bed sediments from Meghna River were collected from arsenic hot spot area of Bangladesh for geochemical analyses. Concentrations of major elements (SiO₂, CaO, MgO, K₂O, Na₂O, Fe₂O₃, Al₂O₃, MnO, TiO₂) and trace elements (V, Cr, Co, Ni, Cu, Zn, Rb, Sr, Y, Zr, Nb, Ba, La, Ce, Nd, Th and Pb) of sediment were analyzed by XRF (X-ray Fluorescence Spectrometer). Total As of bulk sediment was measured by ICP-MS. Total As content in sediment ranges from 1.91 mg kg⁻¹ to 7.97 mg kg⁻¹ (average 3.60 ± 2.04 mg kg⁻¹). Higher content of As (7.97 mg kg⁻¹) is observed in clayey sediment and lower content (1.91 mg kg⁻¹) is found in sandy sediments. The river bed sediment contains arsenic ranges from 1.65 mg kg⁻¹ to 2.27 mg kg⁻¹. The sequential extraction of arsenic along with the strong positive correlation between As, Fe and Mn suggest that As is adsorbed by FeOOH/MnOOH and liberated from sediment into groundwater due to reductive dissolution of FeOOH and MnOOH. The bulk mineralogy of the sediments was determined by X-ray powder diffraction (XRD) method, which shows that quartz, feldspar, mica and chlorite are dominant mineral in sediment of the study area. The positive correlation between As and intensity of mica and chlorite mineral also reveals that the possible source of As in sediment are mica and clay minerals.

1 INTRODUCTION

Arsenic (As) contamination of groundwater of Bangladesh is one of the greatest environmental disasters in the world especially in the southeastern part of Bangladesh (Ahmed *et al.*, 2004; von Brömsen *et al.*, 2007). About 10 million people in Bangladesh are facing various diseases from drinking of arsenic contaminated groundwater. Ganges, Brahmaputra and Meghna carry huge amount of sediment with arsenic rich iron oxides mineral and deposited in the south eastern part of Bangladesh (Seddique *et al.*, 2008). There are several hypotheses regarding the release mechanism of arsenic in groundwater. One of the well established hypotheses is the reductive dissolution of iron oxyhydroxides or manganese oxyhydroxides in presence of organic matter by anaerobic bacteria release arsenic from sediment into groundwater under reducing condition. Few studies have been carried out regarding the source and mobilization of arsenic in the south eastern part of Bangladesh especially in Chandpur district.

2 METHODS/EXPERIMENTAL

2.1 Samples collection in the field

Forty seven (47) sediment samples from two bore holes using hand percussion method at different depths from Hajiganj areas of Chandpur district and two river bed sediments were collected from Meghna river of Chandpur districts in April 2015.

2.2 Samples analyses in the laboratory

The bulk mineralogy of the sediments was determined by X-ray powder diffraction (XRD) using a Rigaku Geigerflex instrument (RAD-IA system) with Ni-filtered Cu K α radiation (30 kV, 10 mA) operating in step scan mode, over an angular range of 2–65° 2 θ with 0.02° 2 θ steps and 2-scount time on 200 mg unoriented side-packed powder mounts. Prior to analysis, 1 g samples, <63 μ m particles passing through 230 mesh, from bulk dried sediments, were ground by hand with a mortar. Approximate relative abundance ratios of major minerals were estimated from the relative intensities of the most intense and specified peak of each mineral. The analytical and sampling-related error in determining mineral abundance was about $\pm 5\%$.

The major element composition of bulk dried and powdered sediments were determined by X-ray fluorescence (VXQ-160S, Shimadzu) using glass bead samples, which were prepared by fusion of sediment samples with lithium borate (1:3 ratio). Calibration lines were obtained using glass beads made by the same procedure with reference sedimentary rock samples (sedimentary rock series, JSd-1, JSd-2, JSd-3, JSI-1 and JIk-1; Geological Survey of Japan). Analytical error for major elements was within $\pm 3\%$, estimated using the duplicate standard samples.

Total As concentration of bulk sediment was determined by ICP-MS (ICP-MS; SPQ 9900) using calibration lines made with commercially distributing standard and spike solutions (WAKO Lmt., Japan). Analytical error is <7% for As elements. BCR method was applied for sequential extraction of As.

Figure 1. As vs Fe₂O₃ in the drilling site 1.

3 RESULTS AND DISCUSSION

3.1 Chemical properties of the sediment samples

Based on XRD analysis, abundant quartz, mica (muscovite and biotite), feldspar (both K-feldspar and plagioclase), and chlorite were observed in both sandy and silty clay sediments. In southeastern Bangladesh, sediments are rich in quartz, mica, feldspar and chlorite (Anawar *et al.*, 2003). The aquifer sediments become gray when the reduction of Fe (oxy) hydroxides is complete and the majority load of sorbed As is released to solution. The depth profile relationships of arsenic with iron, manganese and aluminum oxides and the correlations among these variables suggest that arsenic is strongly associated with iron and manganese oxides and noncrystalline aluminosilicate phases. The correlation analyses (Fig. 1) revealed that the concentrations of As in the sediments are well correlated with those of Fe, Mn and Al contents ($R^2 = 0.666$, $p < 0.05$ for Fe₂O₃, $R^2 = 0.66$, $p < 0.05$ for MnO; $R^2 = 0.666$, $p < 0.05$ for Al₂O₃) in the drilling site 1.

3.2 Mineralogical and geochemical composition of sediments

Positive correlation of As and Fe₂O₃ ($R^2 = 0.666$, $p < 0.05$) observed in the core sediments of Chandpur is consistent with the previous reports by Nickson *et al.* (2000) for the Ganges flood plain sediments, Bangladesh. Ahmed *et al.* (2004) and Hasan *et al.* (2007) also reported that Fe oxyhydroxides are the major hosts of As in the Ganges delta plain sediments.

3.3 Hydrogeochemical factors controlling arsenic mobilization

Sequential extraction method suggests that Fe₂O₃ or MnO phase act as a scavenger of As in sediment (Table 1). A strong positive correlation between As and total Fe (denoted by Fe₂O₃) is observed throughout the sediment column (Fig. 1). Although sediments of the clay unit contain the greatest amounts of Fe₂O₃ (6.96–7.45 wt.%), probably due to enrichment of clay-size chlorite, a correlation with As is observed (Fig. 1). The total As content shows a positive relationship with the XRD intensity of mica and chlorite in both core sediment samples (Seddique *et al.*, 2011).

Table 1. Sequentially extracted As from sediment using BCR method.

Sample	Acid soluble ppm	Reducible ppm	Oxidizable ppm	Insoluble ppm	Residue ppm
D1-10	0.33	0.22	0.62	3.82	0.49
D1-20	0.13	0.44	0.82	2.08	0.00
D1-30	0.16	0.25	0.84	1.77	0.00
D1-40	0.13	0.47	0.76	1.49	0.00
D1-50	0.28	1.30	0.89	5.41	0.59
D1-60	0.14	0.39	0.49	0.90	0.00
D1-70	0.31	1.13	0.93	2.55	0.29
D1-80	0.20	0.68	0.91	1.01	0.20
D1-90	0.20	0.30	0.42	0.52	0.15
D1-100	0.07	0.93	1.37	3.87	0.51
D1-110	0.34	1.08	0.84	4.08	0.51
D1-120	0.28	0.89	0.52	3.19	0.53

4 CONCLUSIONS

Positive correlations among As, MnO, and Fe₂O₃ suggest that reductive dissolution of MnOOH and FeOOH mediated by anaerobic bacteria represents an important mechanism for releasing arsenic into the groundwater. Fe and Mn oxides may be the possible host of As in sediment of Bengal basin. The positive correlation between As and intensity of mica and chlorite mineral also reveals that the possible source of As in sediment are mica and clay minerals.

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