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Relationship of Carbon-Arsenic-Nitrogen in Sediment with Respect to Arsenic Release in Groundwater in Bangladesh - A Preliminary Study

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***Abstract*-The relationship of carbon (C)-arsenic (As)-nitrogen (N) was established in sediment to clarify the effect of chemical N fertilizer to release of As in groundwater. The sediment samples were collected from the ground surface to 60 m deep with 0.6-m intervals, and concentration of As, %C and %N were measured in a farming village of southwestern Bangladesh. The $\delta^{15}N$ and $\delta^{13}C$ analysis were performed to identify the sources of N and C in peat and peaty clay sediment, respectively. The groundwater samples were collected from shallow tube wells to measure the concentrations of As, ammonium-N and $\delta^{15}N$ value. It is found from the chemical analysis data that the %C was positively correlated with both concentration of As and %N in sediment. Again, the amount of As is positively correlated with concentration of ammonium-N in groundwater. The source of N in sediment and groundwater were shown to be from chemical N fertilizers, whereas the source of C in sediment shown to be from C_3 or aquatic plants based on $\delta^{15}N$ and $\delta^{13}C$ values, respectively. The above results confirmed that the chemical N fertilizer when percolates to downward, it acts on the peat sediment and accelerate to release As from sediment to groundwater.**

INTRODUCTION

As poisoning in groundwater is one of the most severe environmental problems and the resulting its poisoning has caused in health and social disasters in Bangladesh. The present study was conducted at Samta village, Jessore district, which is located in south western Bangladesh, is one of the most severely contaminated districts in the country. National Hydro-chemical Survey of Bangladesh, found that 35 million people were drinking groundwater containing As above 50 µg/L (Bangladeshi standard), and 57 million people consume water that exceeds 10 µg/L As (WHO standard). The main source of As in Bengal basin is thought to be geological origin, mostly identified within a shallow depth of Holocene aquifers of deltaic region of the Ganges, Brahmaputra and Meghna Rivers [1]. As is a carcinogenic element, which inhibited enzymes containing (SH) group of the body and causes cancer of skin, liver, lung, bladder and kidney [2]. Researches have been conducted on As issues, but the outcome of those researches is not satisfactory to mitigate the problems. The mechanism of As release from sediment to groundwater is still unclear. There are some hypotheses on As release in groundwater, such as 1) reduction of iron hydroxides and release of sorbed As from the sediments, 2) release of As following the oxidation of As-rich pyrite in the sediments, and 3) anion exchange of sorbed As with phosphate from fertilizers [3]. In Bangladesh, however, the mechanism of As release is considered to be the microbial reduction of iron oxyhydroxides (first point, above) [4]. It was observed positive relationship between the As and ammonium-N concentrations in groundwater, where the source of high ammonium-N concentration is due to application chemical N fertilizer [5]. In the present study, the following topics are examined to know the significant of chemical N fertilizer on release of As in groundwater: (1) established C-As-N correlation in sediment, (2) how the concentration of ammonium-N is correlated with As concentration in groundwater, (3) the source of C in sediment is identified from C_3 or aquatic plant, and (4) whether the source of N in sediment and groundwater is chemical N fertilizer. The groundwater and sediment samples were collected from Samta village, Jessore district, Bangladesh.

MATERIALS AND METHODS

Study Area

The study was carried out at Samta village, Sharsha Upazila, Jessore district, is located in southwestern Bangladesh. It is belongs to the high Ganges river floodplain in the agro-ecological region of Bangladesh [6]. The soil type in this region is calcareous dark grey/brown floodplain [7]. The groundwater is used for domestic (drinking, cooking and washing) and irrigation purposes, and a great deal of chemical N fertilizer is used for the crops and vegetables cultivation in these areas. The national average rate of chemical N fertilizer application is 72 and 92 kg/ha for rice and jute, respectively.

Sampling of Sediment and Groundwater

The sediment samples were collected from the ground surface to 60 m deep with 0.6-m intervals and classified as peat, peaty clay, silty clay and sand. The sediment samples were collected in airtight poly-ethylene bags and preserved in a refrigerator until analysis. Thirty groundwater samples were collected in high density poly-ethylene bottles and acidified. The groundwater samples were collected, 2 liters each site randomly selected from 4 wells and brought to Japan for $\delta^{15}N$ analysis. The sampling was done in February 2009.

Analysis of the Samples

The sediment samples were freeze-dried for overnight and powdered with a porcelain mortar and pestle, and placed into a tin capsule to measure %C, %N, $\delta^{15}N$ and $\delta^{13}C$ value by Automated N/C Analyzer-mass spectrometry (20-20, UK) at Kyushu University. The sediments were air-dried and digested using HNO_3/H_2O_2 [8]. Then As concentration of the groundwater were measured by ICP-MS. The concentration of ammonium-N was measured at site using KRK Aqua Tester, Japan. The $\delta^{15}N$ value of water was performed to know the source of N at Shoko Co., Ltd., Japan.

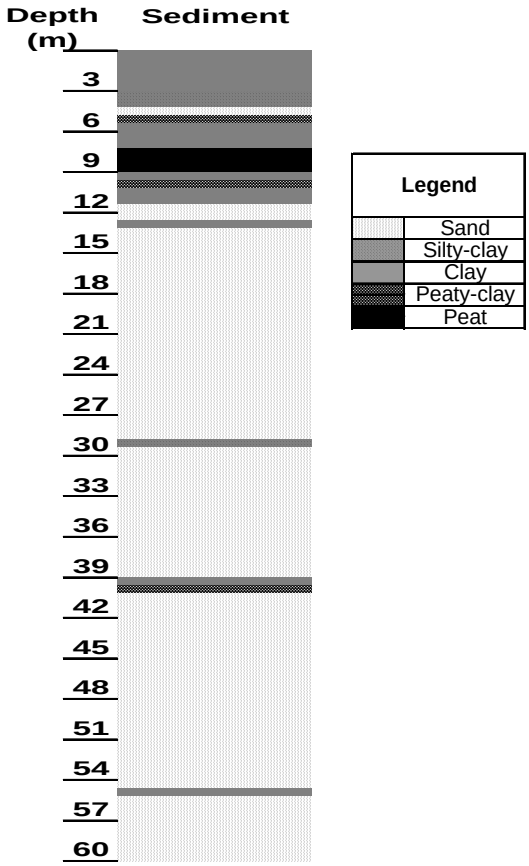

Fig. 1: Geological Profile of the Sediment

RESULTS AND DISCUSSION

Geological profile of the Sediment

The geological profile of the sediment from ground surface to 60 m deep, which consists of peat, peaty clay, clay, silty clay and sand, is shown in Figure-1. Most of the tube wells depth is within 6o meters deep from ground surface in Samta village. The highest concentration of As was found in peat sediment at the level 34.9 to 75.3, whereas concentration ranges in other sediment are 16.5 to 19.8, 5.9 to 15.6, 1.7 to 3.2, 0.7 to 2.8 mg/kg in peaty clay, clay, silty clay and sand, respectively. The average As concentration of the sediment shows in figure-2.

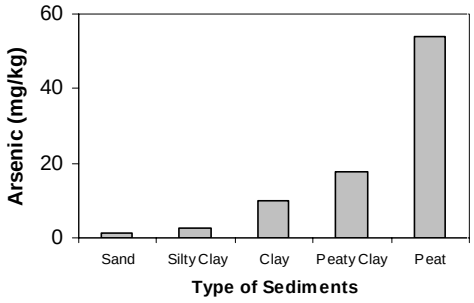

Fig. 2(a): As Concentration of the sediment

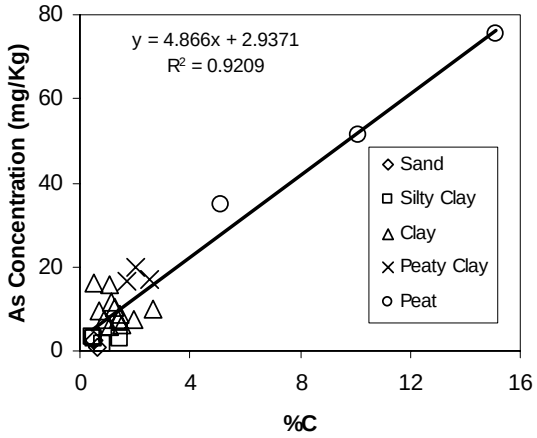

Fig. 2(b): C ~ As relationship in sediment

C-As Relationship in Sediment

The amount of As increases almost proportionally as the %C increases (R^2=0.92) in sediment samples, which is shown in figure-2. The $\delta^{13}C$ value of peat and peaty clay sediment confirmed that the source of C was C_3 plant or aquatic weeds or mixing of these materials, which later absorbed As and deposited to the Holocene aquifers of deltaic region of the Ganges river. During the sampling period, it was observed some decomposed wooden materials in the peat sediment which confirmed the C source is decomposed plants. In sediment, the C-As interaction is to be considered week or inorganic bond, which later release As in groundwater when chemical N fertilizer touches the peat sediment.

C-N Relationship in Sediment

Figure-3 shows, the %N clearly showed an increasing trend with increased %C in sediment (R^2=0.98). In Bangladesh, it is observed that a great deal of chemical N fertilizer was applying in the agricultural field to grow plenty of crops and vegetable [9]. The chemical formula of wood is cellulose ($C_{12}H_{32}O_{14}$) and charcoal (C_7H_4O), which indicates initially N was absent in sediment, because the N source was identified due to apply excessive chemical N fertilizer in agricultural field based on the $\delta^{15}N$ analysis [10]. The C-N correlation is to be considered when chemical N fertilizer percolates downward; then it combined with C in peat sediment. As a result, increased microbial activity and consumed oxygen in peat sediment, thus, decreased oxidation-reduction potential (ORP). Due to enter chemical N fertilizer in peat sediment, N-C interaction become stronger and simultaneously C-As bond become weaker.

Therefore, accelerates to release As in groundwater. A close and positive relationship was confirmed between %C and %N in sediment in this study. Again, Figure-4 shows that the amount of N is positively correlated with concentration of As in sediment. The C-As, C-N and As-N correlation confirmed that the chemical N fertilizer influence significantly to release As in groundwater.

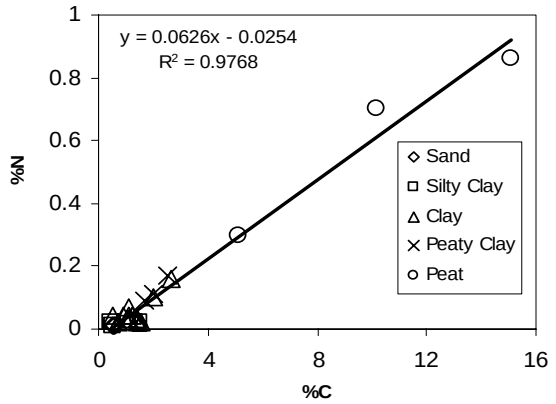

Fig. 3: C ~ N relationship in sediment

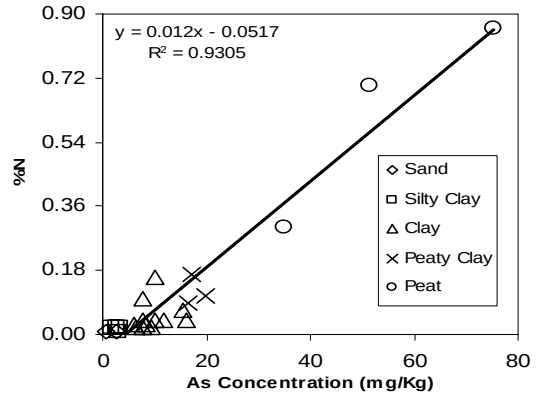

Fig. 4: As ~ %N relationship in sediment

As and ammonium-N Relationship in Groundwater

Figure-5 shows the relationship between the concentrations of As and ammonium-N in groundwater. The concentration of As increases as ammonium-N increases in groundwater samples. In one sample it is found that the concentration of As is as low as 104 µg/L, whereas concentration of ammonium-N was as high as 5 mg/L. The well depth was 14 meters; it was adjacent to an inactive dug-well which was 10 meters deep with concentration of ammonium-N 4.05 mg/L. Therefore it seems that high ammonium-N was percolated from inactive dug-well to that well.

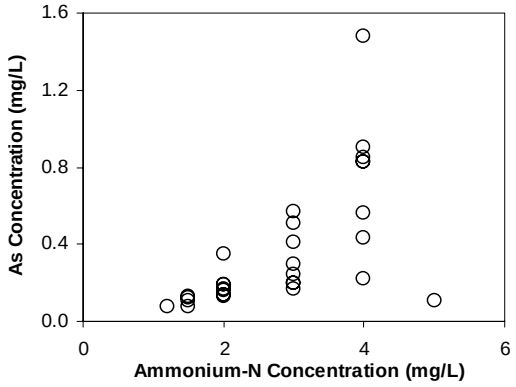

Fig. 5: As ~ ammonium-N relationship in water

Source of N in Water and Sediment, and C in Sediment

The amount of N in peat and peaty clay sediment, and groundwater were found relatively high in Samta village, shown in Table-1. The $\delta^{15}N$ value is to be controlled by the source of nitrogen, and the values range from -7 to 7‰, which indicates that the source of N is chemical fertilizers [10]. The $\delta^{15}N$ values of the groundwater, peat and peaty clay sediment were found within the ranges -7 to 7‰. Therefore, the source of N in groundwater, peat and peaty clay sediment were identified to be from chemical N fertilizer. The $\delta^{13}C$ value is to be controlled by the source of C, and the values range from -23 to -35‰, indicates that the source of C, is C_3 plant or aquatic weeds. The $\delta^{13}C$ values of the peat and peaty clay sediment were found within the ranges -18.18 to -24.67‰, which indicates the source of C in peat and peaty clay sediment were identified to be from C_3 plant or aquatic weeds.

Table-1: Results of $\delta^{15}N$ and $\delta^{13}C$ of the sediment and $\delta^{15}N$ for groundwater

Sediment Samples					Groundwater Samples		
Sl. No	Depth (m)	Sediment Type	$\delta^{15}N$ (‰)	$\delta^{13}C$ (‰)	Well No.	Depth (m)	$\delta^{15}N$ (‰)
1	5.0	Peaty Clay	6.31	-18.81	2	33.5	2.54
2	7.5	Peat	5.52	-19.28	7	16.8	2.39
3	8.5	Peat	2.81	-23.98	16	27.4	1.83
4	40.0	Peaty Clay	-5.07	-24.67	18	15.2	2.28

CONCLUSION

In the present study, it was found that the %C is positively correlated with both concentration of As and %N in sediment. Again, a close and positive relationship between As and ammonium-N concentrations was also confirmed in groundwater. The source of C in peat and peaty clay sediment were identified from C_3 plant or aquatic weeds, whereas the source of N in groundwater and sediment were shown to be from chemical N fertilizer. The above results revealed that the chemical N fertilizer when percolates to

downward, it acts on the peat sediment and accelerate to release As from sediment to groundwater.

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