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<https://doi.org/10.5109/1955382>

出版情報：九州大学大学院農学研究院紀要. 63 (2), pp.207-210, 2018-09-01. Faculty of Agriculture, Kyushu University

バージョン：

権利関係：



Arsenic, Toxic Heavy Metals, and C–N–P in Peaty Sediments in Samta Village, Southwestern Bangladesh

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(Received April 12, 2018 and accepted May 8, 2018)

Concentration of Arsenic (As), other toxic heavy metals, and C, N, P of the peaty sediments collected from Samta Village, Bangladesh were analyzed. The sediment layers were located in 6.5–9.0 m in depth from ground surface. According to the results, As, N (total), $\text{NH}_4\text{-N}$ and C (total) concentrations were larger in peat than peaty clay layers. As is, therefore, may be related to N (total), $\text{NH}_4\text{-N}$ and C (total) in concentrations. There were significant amount of ash, which was inorganic residue (minerals), in the sediments. P_2O_5 in the sediments may be sourced from phosphate fertilizers. The concentration of Fe, Al and Mn were high, while the concentrations of Cr, Cu, Zn, Pb were low. Based on the correlation analysis, the higher As may be related to higher C, N, H in concentrations. Thus, organic substances may be related to the higher As concentration. P_2O_5 and $\text{NH}_4\text{-N}$ were not closely related to the As concentrations. Positive correlations of As with Cu, Zn, Cd, and Pb in concentrations were observed, while any correlation of As with Fe and Mn was not observed.

Key words: Arsenic, sediment core, nitrogen, carbon, heavy metals

INTRODUCTION

In Bangladesh and West Bengal, shallow groundwater aquifers (mostly <100 m) provide over 90% of drinking water (McArthur *et al.*, 2001), however, one million water wells in these areas are contaminated by As with concentrations of $>50 \mu\text{g.L}^{-1}$. Previous reports indicated that a high As concentration in groundwater was associated with a high prevalence of arsenicosis (McDonald *et al.*, 2006).

The As concentration varies with the depth of aquifer as well as the hydrological characteristics of the area. The As concentration in the aquifer sediments in Bangladesh was controlled by the dissolution of iron, manganese and aluminium oxy-hydroxides under anoxic conditions (Bhattacharya *et al.*, 2009; Kumar *et al.*, 2016). In anoxic conditions, As was released when the As-rich iron and manganese oxy-hydroxides were reduced in a process that solubilizes iron and manganese and its absorbed load and increases bicarbonate concentration (Das *et al.*, 1996; Nickson *et al.*, 1998). In addition, As concentration in groundwater may be affected by phosphate fertilizers (Campos, 2002; Lin *et al.*, 2016), and chemical nitrogen fertilizer application (Uddin and Kurosawa, 2011). In the recent study, Whaley–Martin (Whaley–Martin *et al.*, 2017) showed that the oxidation of human/livestock waste coupled with reductive dissolution of iron–oxy–hydroxides may enhance the As release to groundwater from sediments.

In many previous studies, As concentration was very high in peat layer than in others in the As contaminated areas in Bangladesh. Therefore, the authors collected peat or peaty sediments in a highly As contaminated area in Bangladesh, and analysed their concentrations of toxic heavy metals, carbon (C), nitrogen (N), phosphorus (P) and some others as well as their As concentrations. Based on these results, the authors clarify the factors relating to high As concentration in the peaty sediments.

MATERIALS AND METHODS

Study area

The study area of Samta Village is situated on the southeastern part of Bangladesh in Jessore District, neighboring to India. Samta Village is a highly As contaminated area (Alam, Snow and Tanaka, 2003). In the area, groundwater is used not only for drinking purposes but also for irrigation purposes in dry season. The driest month in the area is December with 8 mm precipitation. Most precipitation falls in June, with 314 mm in average. In the rainy season, groundwater levels up to 1.5 to 2.0 m compared to the dry season. Hand-dug wells and many tube-wells are shallow (Biswas *et al.*, 1998). The total geographical area of Samta Village is 3.2 km², where the main portion of the area is used for residential and paddy field purposes.

Sampling methods

Two boreholes in order to collect sediments (Core1 and Core2) were drilled from 0 m (ground surface) to 10 m in depth in Samta Village with hammering method. The horizontal distance between C1 and C2 cores was about 10 m. Sediments collected from the cores were classified as peat, peaty clay, clay and silty clay accord-

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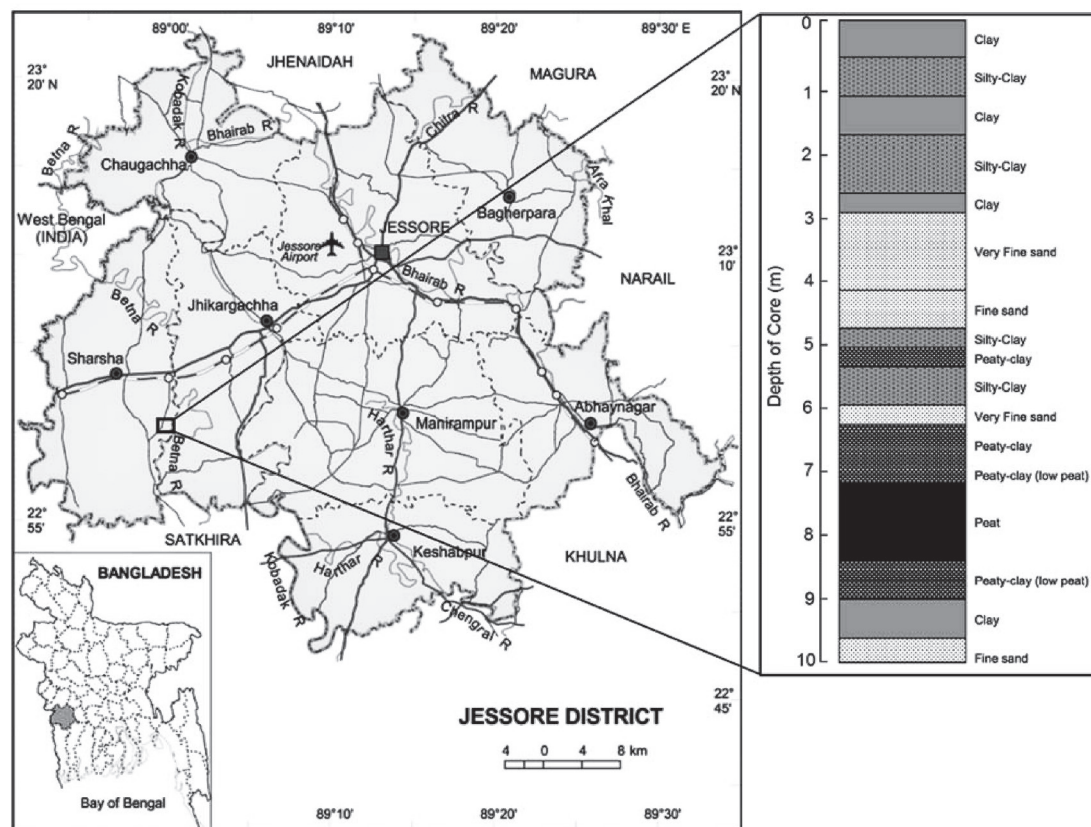


Fig. 1. Samta Village, Jessore District, Bangladesh (study area) and the diagram of the sediment layer from which peat samples were collected.

ing to the criterion given by Roy *et al.* (Roy *et al.*, 2005). The classification results indicated that two cores profiles were nearly the same with each other (Fig. 1). In the layers, three layers of peat/peaty clay in Cores 1 and 2 were targeted, which were the layers of peaty clay (L1 layer: 6.5–7 m), peat (L2 layer: 7.5–8.3 m) and peaty clay (low peat) (L3 layer: 8.5–9 m).

These samples collected were preserved in a plastic bag and then brought to Kyushu University, Japan for analysis with an import permission.

Analytical methods

Sediment samples were air-dried and ground to powder and sieved through a 2 mm sieve for further analysis. In order to determine the total concentration of heavy metals, the digestion method of the United

States Environmental Protection Agency 3050B method (USEPA, 1996) was applied. The digested solutions were diluted with pure water and filtered through 0.45 μm syringe filter to analyse the concentrations of arsenic (As), lead (Pb), copper (Cu), zinc (Zn), cadmium (Cd), chromium (Cr), iron (Fe), aluminum (Al) and manganese (Mn). Analysis was done by using Inductively coupled plasma – Mass spectrometry (ICP-MS, 7500ce, Agilent Tech, Inc., CA, USA). All determinations were repeated two times from digestion to analysis. The concentrations of C, N, H and ash in sediments were determined by using CHN Corder. For analysing $\text{NH}_4\text{-N}$, a colorimetric method was applied (Dorich and Nelson, 1983; SSSA, 1996). Available phosphorus (P_2O_5) concentration was analyzed using Olsen method (SSSA, 1996; Iatrou *et al.*, 2014).

Table 1. The main properties of the sediments in the two sediment cores

| Core | Layer | Depth (m) | H (%) | C (%) | N (%) | C/N ratio | Ash (%) | P_2O_5 ($\mu\text{g/g}$) | P_2O_5 (%) | $\text{NH}_4\text{-N}$ ($\mu\text{g/g}$) |
|------|-------|-----------|-------|-------|-------|-----------|---------|--|----------------------------|--|
| 1 | L1 | 6.5–7.0 | 1.38 | 1.71 | 0.13 | 13.2 | 85.1 | 341.5 | 0.09 | 46.2 |
| | L2 | 7.5–8.0 | 1.74 | 5.15 | 0.37 | 13.9 | 79.1 | 450.1 | 0.09 | 386.2 |
| | L3 | 8.5–9.0 | 1.02 | 0.91 | 0.06 | 15.2 | 87.7 | 485.3 | 0.08 | 36.3 |
| 2 | L1 | 6.5–7.0 | 1.36 | 0.59 | 0.07 | 8.4 | 85.7 | 444.1 | 0.09 | 77.4 |
| | L2 | 8.0–8.3 | 2.14 | 10.24 | 0.75 | 13.7 | 70.0 | 469.3 | 0.12 | 337.1 |
| | L3 | 8.5–9.0 | 1.3 | 1.23 | 0.11 | 11.2 | 83.2 | 604.4 | 0.12 | 262.6 |

RESULTS

Concentrations of trace toxic elements and soil properties**Table 2.** Mean values of the concentration ($\mu\text{g/g}$) of As, and other toxic heavy metals in the two sediment cores

| Core | Layer | Al | Fe | Mn | Cr | Cu | Zn | Cd | Pb | As |
|------|-------|--------|--------|-------|------|------|------|-----|------|------|
| 1 | L1 | 10,239 | 17,794 | 93.3 | 23.3 | 13.6 | 22.3 | 0.1 | 15.3 | 9.4 |
| | L2 | 9,244 | 13,470 | 78.6 | 20.7 | 18.3 | 18.8 | 0.1 | 17.2 | 54.7 |
| | L3 | 8,643 | 16,079 | 117.2 | 20.6 | 13.0 | 22.2 | 0.1 | 17.1 | 9.0 |
| 2 | L1 | 10,454 | 20,505 | 374.5 | 24.3 | 15.4 | 22.9 | 0.1 | 17.4 | 9.6 |
| | L2 | 7,873 | 10,998 | 71.2 | 18.7 | 29.5 | 14.2 | 0.3 | 24.9 | 79.1 |
| | L3 | 8,806 | 18,111 | 145.6 | 19.5 | 13.0 | 23.6 | 0.1 | 16.5 | 9.0 |

Correlations between the parameters**Table 3.** The Pearson's correlation coefficient values between the parameters measured for the peaty sediments

| Core | H | C | N | Ash | P ₂ O ₅ (Avail) | P ₂ O ₅ (Total) | NH ₄ -N | As | Al | Fe | Mn | Cr | Cu | Zn | Cd |
|---|----------------|----------------|----------------|----------------|--|--|--------------------|----------------|-------|----------------|-------|-------|----------------|----------------|--------------|
| C | 0.95** | 1. | | | | | | | | | | | | | |
| N | 0.95** | 1.** | 1. | | | | | | | | | | | | |
| Ash | -0.97** | -0.98** | -0.98** | 1. | | | | | | | | | | | |
| P ₂ O ₅ - Avail. | -0.12 | -0.05 | -0.04 | -0.07 | 1. | | | | | | | | | | |
| P ₂ O ₅ - Total | 0.55 | 0.53 | 0.56 | -0.68 | 0.62 | 1. | | | | | | | | | |
| NH ₄ -N | 0.76 | 0.72 | 0.73 | -0.79 | 0.37 | 0.59 | 1. | | | | | | | | |
| As | 0.94** | 0.98** | 0.97** | -0.95** | -0.04 | 0.45 | 0.79 | 1. | | | | | | | |
| Al | -0.41 | -0.64 | -0.63 | 0.62 | -0.54 | -0.58 | -0.54 | -0.6 | 1. | | | | | | |
| Fe | -0.76 | -0.91 | -0.89 | 0.83 | 0. | -0.32 | -0.69 | -0.91 | 0.77 | 1. | | | | | |
| Mn | -0.34 | -0.51 | -0.48 | 0.43 | 0.04 | -0.09 | -0.42 | -0.47 | 0.61 | 0.75 | 1. | | | | |
| Cr | -0.41 | -0.61 | -0.6 | 0.62 | -0.62 | -0.64 | -0.67 | -0.58 | 0.97 | 0.74 | 0.67 | 1. | | | |
| Cu | 0.93** | 0.97** | 0.97** | -0.96** | -0.05 | 0.57 | 0.62 | 0.93** | -0.58 | -0.8 | -0.3 | -0.5 | 1. | | |
| Zn | -0.91* | -0.99** | -0.98** | 0.94** | 0.13 | -0.42 | -0.65 | -0.97** | 0.62 | 0.92** | 0.48 | 0.55 | -0.97** | 1. | |
| Cd | 0.89* | 0.97** | 0.97** | -0.92** | -0.23 | 0.41 | 0.55 | 0.92** | -0.58 | -0.88** | -0.53 | -0.51 | 0.95** | -0.98** | 1. |
| Pb | 0.8 | 0.88* | 0.89* | -0.88** | 0.11 | 0.65 | 0.50 | 0.83* | -0.69 | -0.73 | -0.21 | -0.57 | 0.96** | -0.89** | 0.87* |

(*, ** significant at 0.05 and 0.01 levels; P₂O₅ (Avail): P₂O₅ Available)

DISCUSSION

According to Table 1, N, NH₄-N and C concentrations were far larger in L2 layer than in the layers. While, according to Table 2, As concentration was also far larger in L2 layer than in the layers. The As concentration in L2 layer was 54.7–79.1 $\mu\text{g/g}$. Therefore, As may be related to N, NH₄-N and C in concentrations. Concentration of H was also large in L2 layers.

Ash occupied a large portion of 70% or more, showing a lot of inorganic residue (minerals) left in the sediments. P₂O₅ has a possibility to be sourced from phosphate fertilizer, because P₂O₅ is a usual form of phos-

phate fertilizers. In Table 2, P₂O₅ was always higher than NH₄-N. N concentration was low in L1 and L3 layers (< 0.2) but medium or high in L2 layers. The C/N ratio, an indicator for the degree of decomposition, ranged medium or low of 8.4–15.1.

According to Table 2, Fe concentration was the largest with more than 10,000 $\mu\text{g/g}$, followed by Al (7,873–10,454 $\mu\text{g/g}$) and then by Mn (71.2–374.5 $\mu\text{g/g}$) concentrations. Concentrations of Cr, Cu, Zn and Pb did not differ with each other and ranged from 13.6–29.5 $\mu\text{g/g}$. Cd concentration was a low of 0.1–0.3 $\mu\text{g/g}$.

According to Table 3, significant correlations were observed between the concentration of As, C–H–N and

ash. The correlation coefficient of As with C–H–N was positive, and that with ash was negative. Thus, the higher As may be related to higher C, N, H in concentrations. Here, organic substances may be related to the higher As concentration. While, neither correlation of P_2O_5 nor NH_4 –N with the As concentration was observed. Further examination is necessary.

Significant positive correlations were also observed between As and Cu, Zn, Cd and Pb concentrations. It was not clear why the As was related to other toxic heavy metals of Cu, Zn, Cd and Pb in concentrations. On the other hand, significant correlation was not observed between As and Fe, and between As and Mn. For which, further examination is necessary.

AUTHOR CONTRIBUTIONS

Nguyen Van Thinh performed the chemical analyses and wrote the first draft of the manuscript. Masaru Matsumo and Kiyoshi Kurosawa conducted the data analyses and provided laboratory equipment in Japan. Shamin Uddin sampled the sediment cores in Bangladesh and sent the samples to Japan. Yiping Xie assisted the chemical analysis in laboratory.

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