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## Effect of chemical nitrogen fertilizer application on the release of arsenic from sediment to groundwater in Bangladesh

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### Abstract

The effect of chemical nitrogen fertilizer application on groundwater arsenic pollution was examined. Arsenic and N concentrations in peat sediment were high. In the groundwater, a high arsenic concentration was observed under a high ammonium-N concentration with reducing conditions. From  $\delta^{15}\text{N}$  analysis, the source of N in peat sediment and groundwater was possibly chemical N fertilizer. Thus, we assumed that a high N environment created by the fertilizer led the groundwater to reducing conditions through the enhancement of microbial activity, and the reducing condition promoted the arsenic release from peat sediment to the groundwater.

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Keywords: ammonium-N, cause, peat sediment, pollution, reducing condition

### 1. Introduction

Groundwater arsenic (As) pollution has been reported in large river delta regions in Asia, including the Ganges Delta of Bangladesh. As pollution is one of Bangladesh's most severe environmental problems. The main source of As in Bangladesh is of geological origin, since As is contained in the sediments of the shallow Holocene aquifers of the Ganges Delta [1]. Millions of shallow tube wells have been drilled in the Ganges Delta to supply drinking water, but the water contains a lot of As. According to the National Hydro-Chemical Survey of Bangladesh conducted in 1998-99, nearly 35 million people drink tube well water containing As levels that exceed the Bangladeshi drinking water standard ( $\leq 50 \mu\text{g/L}$ ). As taken into the human body causes various cancers [2]. Nearly 40 thousand arsenicosis patients have been identified in Bangladesh [3].

Much research has been conducted on groundwater As pollution in Bangladesh, but it has not produced outcomes that can satisfactorily mitigate the As poisoning. Year by year, the As poisoning problem becomes more severe. The mechanisms through which As is released from sediment into groundwater are still unknown. There are some hypotheses about the mechanism, including (i) the reduction of iron hydroxides and release of adsorbed As from the sediment [4, 5, 6], (ii) the release of As following the oxidation of As-rich pyrite in the sediment [7], and (iii) anion exchange of adsorbed As with phosphate from fertilizers [8]. Whether any of these hypotheses are right is still not clear, however, the most widely accepted mechanism is (i). The microbial activity is considered as a factor to create reducing conditions in groundwater through decomposition of organic matter in relation to the mechanism of (i) [9].

Intensive cropping is done in Bangladesh to produce food for 150 million people living in the small country of only 147 thousand  $\text{km}^2$ . The Bangladeshi government has subsidized chemical N fertilizer for farmers to use to grow crops and vegetables [10]. However, massive application of chemical N fertilizer is a possible cause of As pollution in groundwater. According to Kurosawa et al. [11], the ammonium-N concentration in groundwater was high in an As-contaminated area in Bangladesh, and the source of the ammonium-N would be chemical N fertilizers. The source and concentration of N in sediments, however, have not yet been examined.

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The intent of this paper is to clarify whether application of chemical N fertilizer causes As pollution of groundwater in Bangladesh by examining the following points: (1) how As concentration is correlated with ammonium-N and oxidation-reduction potential (ORP) for groundwater, (2) how the concentration of As is correlated with %N in sediments, and (3) whether the source of N in groundwater and sediments is chemical N fertilizer or not.

## 2. Materials and Methods

### 2.1 Study Area

The study area is Samta village in the Jessore district in southwestern Bangladesh. Figure 1 shows a location map of the area. This area is one of the most severely As-contaminated areas in the country [12]. The area belongs to the high Ganges river floodplain in the agro-ecological zoning of Bangladesh [13]. The soil in the area is calcareous dark grey/brown floodplain soil with low fertility [14]. According to meteorological data recorded at Jessore city from 1996-2003, the mean annual rainfall was 1,697 mm, the mean annual maximum and minimum temperatures were 33 °C and 17 °C, and the mean annual relative humidity was 80% [14]. Groundwater is used for domestic use and irrigation purposes. Rice-rice and sugarcane monoculture are the two dominant annual cropping patterns in the area. Although the amount of fertilizers applied is not available for the study area, the national average rates of chemical N fertilizer application are 72 kg/ha for rice and 85 kg/ha for sugarcane [15].

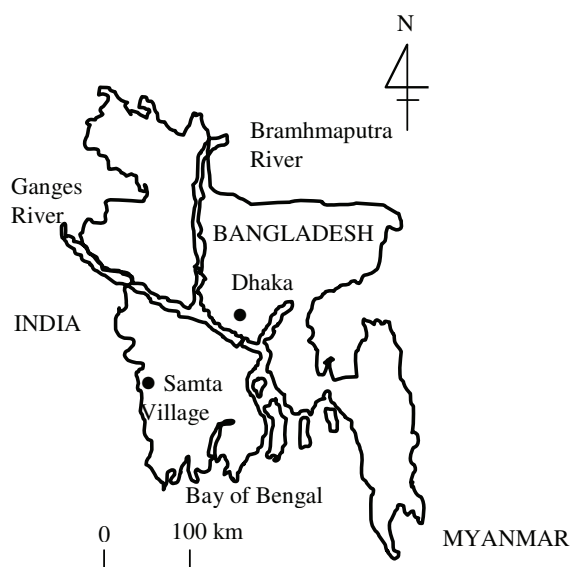


Fig. 1 Location map of the study area

### 2.2 Sampling of Groundwater, Sediments and Fertilizer

Groundwater was sampled from shallow tube wells to measure the water quality. The depths of the wells were determined based on interviews with local people. Sediments were sampled using a hand boring method from ground level to 60 m deep at intervals of 0.6 m. The chemical N fertilizer used in the study area was sampled from a local bazaar nearby. The sampling was done in February and March of 2009 at the beginning of the dry season, and the collected samples were brought to Japan for analysis.

### 2.3 Measurement of Groundwater, Sediment and Fertilizer Parameters

The ORP of the groundwater was measured immediately after sampling at each site using a portable meter (Horiba D53, Horiba Co. Ltd). The recorded ORP value was then converted to a value corresponding to the standard hydrogen electrode. The ammonium-N ( $\text{NH}_4\text{-N}$ ), nitrate-N ( $\text{NO}_3\text{-N}$ ) and nitrite-N ( $\text{NO}_2\text{-N}$ ) concentrations of groundwater were measured at each site using a field kit (Aqua tester, KRK). The As concentration of groundwater was measured by inductively coupled plasma mass spectroscopy (ICP-MS) (Agilent 7500 series, Agilent Technologies) at Kyushu University.  $\delta^{15}\text{N}$  analysis of groundwater to identify the source of inorganic-N (ammonium-N plus nitrate-N plus nitrite-N) in the water was performed for four selected samples. The analysis was contracted to Shoko Co. Ltd., Japan, due to lack of equipment in our laboratory.

The sediments were air-dried at room temperature for 3 to 4 days and then ground to a powder. These sediments were

classified as peat, peaty clay, clay, silty clay and sand according to the criterion given by Roy et al. [16]. Sediment samples were first digested using USEPA 3050B method [17], and then the digestive solution was analyzed using ICP-MS as noted above. Sediment samples were freeze-dried overnight, and then powdered with a porcelain mortar and pestle. The meshed samples were placed into tin capsules and an ANCA mass-spectrometer (SL/20-20, SerCon) at Kyushu University was used to measure the %N and  $\delta^{15}\text{N}$  values of the sediments. The  $\delta^{15}\text{N}$  values of the chemical N fertilizer were measured in the same way as for the sediment samples.

## 2.4 Laboratory Experiment on the Release of As from Sediment

To elucidate the effect of chemical N fertilizer application on the release of As from sediment, two cases of an experiment were conducted in the laboratory at Kyushu University. In the first, 10 g of peat sediment and 1 g of chemical N fertilizer (urea) were mixed into 100 ml of pure water, and in the second, 10 g of peat sediment was mixed into 100 ml of pure water. These mixtures were done at the beginning of experiment, i.e. on day 1. The urea fertilizer contained 46% of N according to its specification, and the fertilizer did not contain As by our measurements. In the experiment, the ratio of the fertilizer to the sediment in amount was far higher than that in the natural sites. This ratio was set on purpose to identify the release mechanism.

The above mixtures were combined in an airtight container on day 1, and the container was shaken mechanically. The containers were shaken for one hour every day, after which the solution was sampled. This solution was centrifuged and filtered through a 0.2  $\mu\text{m}$  opening filter that was the finest filter commercially available. Though fine clay and nano-scale oxyhydroxides could pass the filter, the amount passed would have been very small judging from that the filtered solution was transparent. The concentrations of As and ammonium-N, and the ORP of the filtered solution were measured using the same methods as discussed above. From the As concentrations, the amount of As released from the peat sediment into the water was calculated.

**Table 1. Depth of sampled groundwater, ORP and concentrations of As, ammonium-N, nitrate-N and Nitrite-N in groundwater for each well at sites A through E**

Site	Well No.	Depth of the sampled water (m)	ORP (mV)	As ( $\mu\text{g/l}$ )	Ammonium-N (mg/l)	Nitrate-N (mg/l)	Nitrite-N (mg/l)
A	1	34	77	432	4.0	0.01	0.01
	2	34	76	507	3.0	0.01	0.01
	3	40	81	240	3.0	0.01	0.01
	4	37	95	195	3.0	0.01	0.00
	5	23	75	200	3.0	0.01	0.01
	6	17	116	223	4.0	0.01	0.02
B	7	37	94	124	1.5	0.01	0.01
	8	40	97	130	1.5	0.01	0.01
	9	30	87	105	1.5	0.01	0.01
	10	32	70	189	2.0	0.01	0.01
	11	40	73	73	1.5	0.01	0.02
	12	12	95	569	3.0	0.01	0.02
C	13	27	79	129	2.0	0.01	0.02
	14	46	70	296	3.0	0.01	0.02
	15	21	88	410	3.0	0.01	0.02
	16	27	78	906	4.0	0.01	0.01
	17	18	92	166	3.0	0.01	0.02
	18	15	153	187	2.0	0.01	0.02
D	19	34	83	827	4.0	0.01	0.01
	20	34	86	853	4.0	0.01	0.01
	21	40	88	827	4.0	0.01	0.01
	22	34	85	561	4.0	0.01	0.01
	23	23	83	1480	4.0	0.05	0.01
	24	13	192	77	1.2	0.02	0.00
E	25	35	38	136	2.0	0.01	0.01
	26	35	84	352	2.0	0.01	0.01
	27	40	88	157	2.0	0.01	0.01
	28	27	94	133	2.0	0.01	0.02
	29	37	93	164	2.0	0.01	0.01
	30	14	155	104	5.0	0.02	0.01

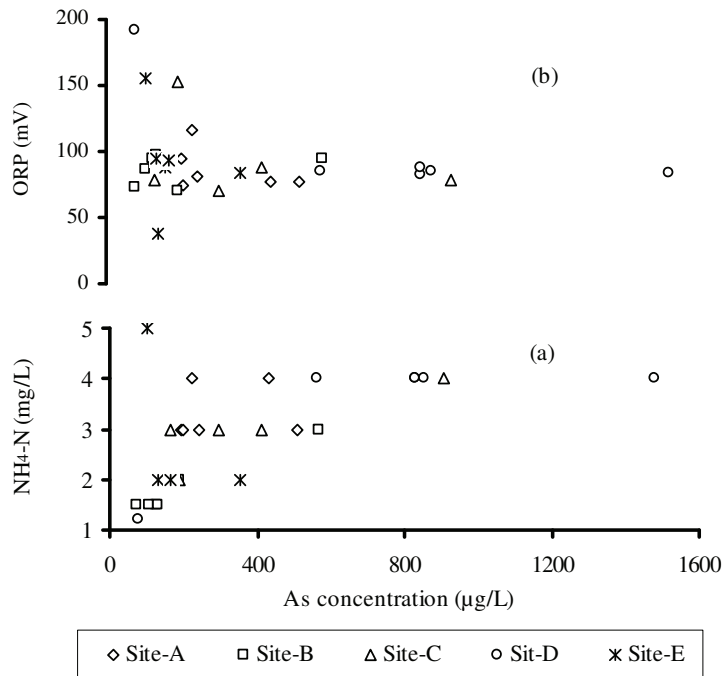


Fig. 2 As concentration in relation to ammonium-N (NH<sub>4</sub>-N) concentration and ORP for groundwater

The experiment continued for ten days. Since measurements were taken seven times during that period, seven identical samples were drawn for measurement in each case, because each sample was disposed of after measurement.

### 3. Results

#### 3.1 Groundwater Quality

Table 1 shows the depth of sampled groundwater, the ORP, and the ammonium-N, nitrate-N and nitrite-N concentrations of the groundwater for each well at each site. Six wells were targeted at each site. The depth of the sampled groundwater was 12 m to 46 m. The ORP value ranged from 0 mV to 200 mV, which was categorized as reduced condition [18, 19]. The As concentrations were higher than the Bangladeshi drinking water standard, and the ammonium-N concentration was high, more than 1 mg/L, but concentrations of nitrate-N and nitrite-N were low, each being less than 0.05 mg/L at each site.

#### 3.2 Relationship between As Concentration, ORP and Ammonium-N Concentration in Groundwater

Figure 2 shows As concentration in relation to ammonium-N concentration and ORP of the groundwater for sites from A to E. From Fig. 2(a), it appears that the As concentration increases as the ammonium-N concentration increased, but there was a wide variation in the As concentration when the ammonium-N concentration was a high (3 mg/L to 4 mg/L). When the ammonium-N concentration was 5 mg/L, the As concentration was at a low of 104 µg/L. The tube well from which this sample was taken was shallow (14 m) and located quite near to an abandoned 10 m deep dug-well that was supposed to have a high ammonium-N concentration affected by waste dumping. The ammonium-N concentration in the tube well may have been affected by existence of the dug well.

In Fig. 2(b) it can be seen that an As concentration higher than 400 µg/L was observed only when the ORP was between 50 mV to 100 mV indicating that the As was released from sediments largely in reduced groundwater conditions.

#### 3.3 Geological Profile of the Sediments

Figure 3 shows the geological profile of the sediments from the ground surface to 60 m deep at sites A through E. From Fig. 3, we see that silty clay, clay, peaty clay and peat sediments are mainly located within 9 m of the surface at each site, followed by sandy sediment proceeding deeper. The deeper, sandy sediment was more than 10 m thick at most of the sites, whereas the peat and peaty clay sediments were less than 2 m thick. There were peat or peaty clay sediments located 36 m to 48 m deep at sites A, B, C and E, but neither peat nor peaty clay sediment was observed at site D at those depths.

Figure 4 shows the As concentration of each sediment by depth at site A. The As contamination in each sediment

ranged from 34.9 mg/kg to 75.3 mg/kg for peat, 16.5 mg/kg to 19.8 mg/kg for peaty clay, 5.9 mg/kg to 15.6 mg/kg for clay, 1.7 mg/kg to 3.2 mg/kg for silty clay and 0.7 mg/kg to 2.8 mg/kg for sand. The As concentration of the peat sediment was very high, being twice or more than that of the peaty clay sediment, which had the second highest As concentration.

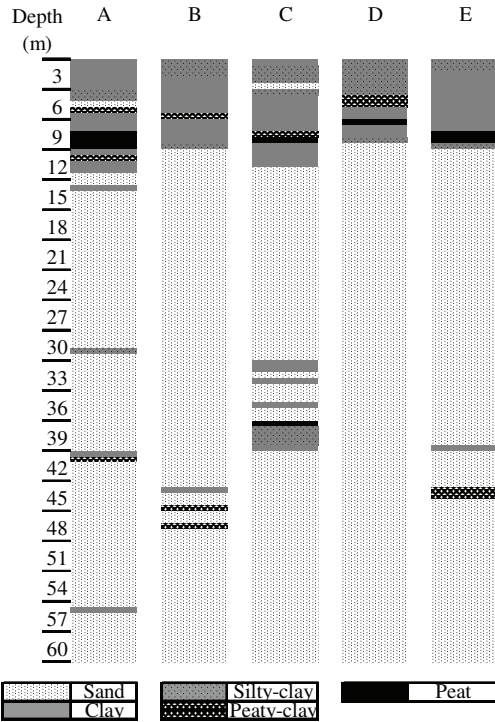


Fig. 3 Geological profiles of sediments from the ground surface to 60 m deep at sites A to E

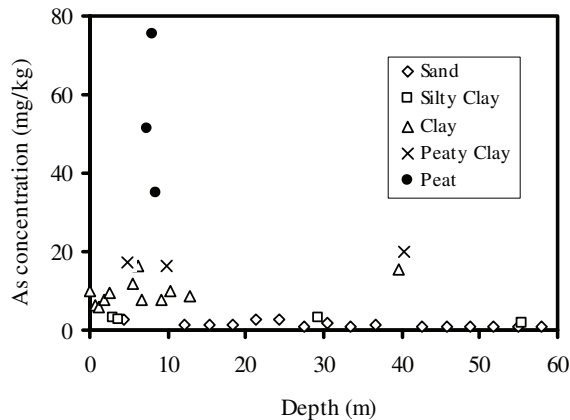


Fig. 4 As concentration of each sediment by depth at site A

**3.4 Relationship between As Concentration and %N in Sediments**

Figure 5 shows the relationship between As concentration and %N in sediments at site A. Here, %N is positively correlated with As concentration. The peat sediment had the highest %N (0.3% to 0.9%) and As concentration (35 mg/kg to 75 mg/kg) of all the sediments. In the peaty clay sediment, %N (0.1% to 0.2%) and As concentration (17 mg/kg to 20 mg/kg) were the second highest among the sediments, but were much lower than those of the peat sediment.

**3.5  $\delta^{15}N$  Values of Groundwater, Sediment and Fertilizer**

The  $\delta^{15}\text{N}$  value of the groundwater sampled from four representative wells at sites A and C is shown in Table 2. We targeted these samples, which had a high ammonium-N concentration. The  $\delta^{15}\text{N}$  value ranged from 1.83‰ to 2.54‰, a low value with a minor variation. Since the nitrate-N and nitrite-N concentrations were much lower than the ammonium-N concentration, the  $\delta^{15}\text{N}$  value represents ammonium-N. The  $\delta^{15}\text{N}$  values of the peat sediment at site A and the N fertilizer collected near the study area are also shown in Table 2. The value for the sediment ranged from 2.81‰ to 5.52‰, while that of the N fertilizer was 1.81‰.

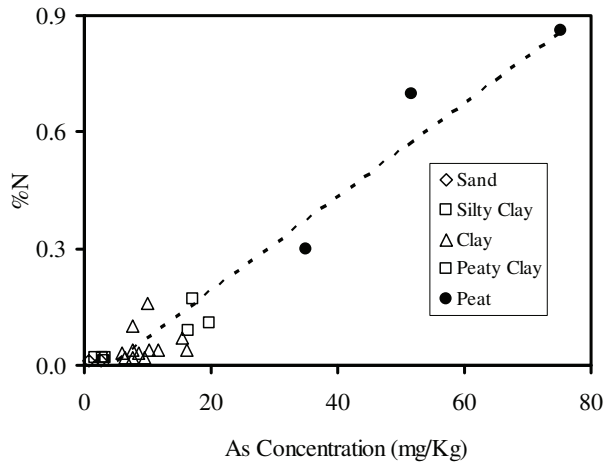


Fig. 5 Relationship between As concentration and %N in sediments at site A

Table 2.  $\delta^{15}\text{N}$  value of groundwater, sediment and fertilizer for representative samples

Sample	Groundwater		Peat sediment		N fertilizer		
	$\delta^{15}\text{N}$ (‰)	Ammonium-N (mg/L)	Sample	$\delta^{15}\text{N}$ (‰)	N (%)	Sample	$\delta^{15}\text{N}$ (‰)
1	2.39	4.0	I	5.52	0.93	i	1.81
2	2.54	3.0	II	2.81	0.39		
3	2.28	2.0					
4	1.83	4.0					

### 3.6 Experiment on the Release of As

Figure 6 shows how the release of As from peat sediment to water changes from day to day during the experiment. Figure 6(a) shows the case where N fertilizer was added, and Fig. 6(b) shows the case where no fertilizer was added. The As release was 157  $\mu\text{g}/\text{kg}$  and 175  $\mu\text{g}/\text{kg}$  in case (a) and (b) on day 1, and increased rapidly to 196  $\mu\text{g}/\text{kg}$  in case (a) but decreased a little to 169  $\mu\text{g}/\text{kg}$  in case (b) on day 2. The As release was highest on day 3, being 208  $\mu\text{g}/\text{kg}$  and 181  $\mu\text{g}/\text{kg}$  in case (a) and (b), respectively. After that it decreased with day, but the decrease was slower in case (a) than in case (b). The As release on day 10 was 171  $\mu\text{g}/\text{kg}$  in case (a), but 108  $\mu\text{g}/\text{kg}$  in case (b). The As release was consistently higher in case (a) than in case (b) from day 2 to day 10.

Table 3 shows the ORP and ammonium-N concentration of the water each day. In case (a), the ORP increased on day 2, and then showed minor changes between 390 mV and 416 mV each day while the ammonium-N concentration changed between 60 mg/L and 200 mg/L each day. In case (b), the ORP increased on day 2, and then showed minor changes between 435 mV and 486 mV each day. The ammonium-N concentration was very low, always no greater than 1.2 mg/L.

The ORP in the experiment was not so low as that in the field. The experiment was conducted at a room, where light was not shut off. Sealing for airtight on the container, which was a beaker, might not be enough because the beaker was a usual type. These conditions might hinder the microbial activity even some extent, resulting in comparatively higher ORP in case (a) and small difference of ORP between cases (a) and (b), respectively.

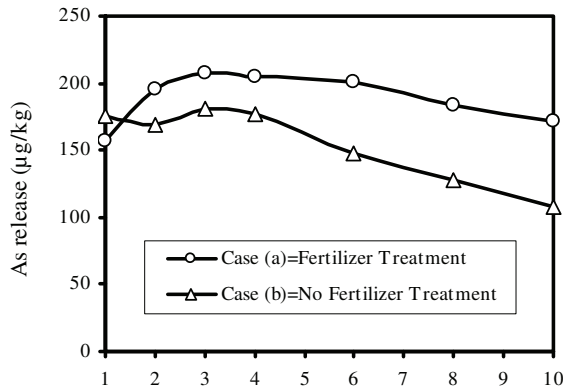


Fig. 6 Changes of As released from peat sediment into water by day

Table 3. ORP and ammonium-N concentration in the water on each day of the experiment

Day	ORP (mV)		Ammonium-N (mg/L)	
	Case (a)	Case (b)	Case (a)	Case (b)
1	200	311	60	1.2
2	414	454	70	0.8
3	407	472	80	1.0
4	416	453	120	0.8
6	390	492	140	0.8
8	413	435	160	0.8
10	403	486	200	1.2

## 4. Discussion

### 4.1 Mechanism of As Release from Sediment to Groundwater

Arsenic was released in a reduced groundwater environment (Fig. 2(b)). Of the three hypotheses noted earlier, the release mechanism corresponds to the “reduction of iron hydroxides and release of adsorbed As from the sediment” hypothesis. The release of As from peat sediment contributes greatly to the As concentration of the groundwater under the hypothesis. Because, as the reducing condition of groundwater become stronger, the As release occurs intensively with no limits from the peat sediment that had abundant As (Fig. 4). The amount released from peaty clay sediment may also contribute, but not greatly because its As concentration is lower. A very high As groundwater concentration was observed at site D (Table 1), where the peat or peaty clay sediment layer was not very thick compared to the other sites (Fig. 3). For this reason, it appears that not only the thickness of the sediment but other factors may affect the release, and further study is necessary.

The As concentration became higher as the concentration of ammonium-N increased in the groundwater (Fig. 2(a)), and when the ammonium-N concentration was high, the groundwater ORP showed a reduced condition to be present (Fig. 2(b)). From these relationships, it was assumed that as the ammonium-N concentration increased, oxygen in the water was consumed by enhanced microbial activity, where the ammonium-N served as a nutrient for microorganisms. The groundwater conditions thus developed into a reducing environment as suggested by Kurosawa et al. [11].

If the lateral flow exists in addition to the vertical flow in groundwater in the study area, the effect of the flow from outside the area on a groundwater sampling site will be small, because the lateral flow could have slow velocity due to the flat topography and the fertilizer application is done widely in the area.

### 4.2 Source of N in Groundwater and Sediment

Seiler [20] showed that the  $\delta^{15}\text{N}$  value of chemical N fertilizer was -4 to +4 ‰, soil organic N was +4 to +9 ‰, and animal or human wastes was more than +10 ‰, respectively. According to this criterion, the source of N of a peat sediment ( $\delta^{15}\text{N}$

of 2.81 ‰) was chemical N fertilizer. That of another peat sediment ( $\delta^{15}\text{N}$  of 5.52 ‰) was the soil organic N. However, the source of the latter peat sediment can be a combination of the chemical N fertilizer and some others for instance of soil organic N, considering from the magnitude of  $\delta^{15}\text{N}$ . Even for the former peat sediment, the combination is conceivable, because its  $\delta^{15}\text{N}$  was a little higher than that for the fertilizer measured by us (1.81‰ in Table 2).

While, the source of N of a groundwater was considered to be the chemical N fertilizer from the  $\delta^{15}\text{N}$  (1.83‰ in Table 2), which was almost the same with the  $\delta^{15}\text{N}$  of the fertilizer measured (Table 2). The source of another groundwater ( $\delta^{15}\text{N}$  was 2.54 ‰ in Table 2) was probably the chemical N fertilizer or the combination of the fertilizer N and some others as mentioned above. The groundwater is thought to be contacting or closely positioned to the peat sediment, and the source of N in peat sediment would not be different from that of groundwater. Since the peat sediment is located mainly at shallow depths of 10 m or less (Fig. 3), N, probably originating in fertilizer or the combination of the fertilizer and others that soaked into the ground, can be adsorbed onto the sediment. For this reason, the peat sediment had a high %N (Fig. 5).

It is not clear why peat sediment had the highest %N, despite some peaty clay and clay sediments being located shallower than the peat sediment. Since peat contains 12 - 60 % of organic carbon [21], further research should examine organic carbon's ability to raise %N.

The N in the sediments is considered to be released to groundwater simultaneously with the As, probably producing a high ammonium-N concentration in the groundwater.

### 4.3 Effect of Chemical N Fertilizer Application on the Release of As from Sediment

The experiment on the release of As showed that the release was larger in case (a) than in case (b) during the 10-day experiment (Fig. 6). In the experiment, all conditions for the two cases were identical except for chemical N fertilizer application.

ORP was not technically in a reduced condition of 0 mV to 200 mV in either case, but the ORP was consistently lower in case (a) than in case (b) during the experiment, and the ammonium-N concentration increased with day in case (a), while remaining very low in case (b) (Table 3). These characteristics show that the ORP and ammonium-N concentrations are related, to some extent, to the release of As from the sediment.

The average release of As during the experiment was 22% higher in case (a) than in case (b). Therefore, the N fertilizer was identified as playing a role in promoting the release of As from the sediment.

Here, the releases of other elements than As, i.e. Fe, Al and Mn, which are the significant adsorbents of arsenic [22], were not examined. Because, the effects of microbial activities on the Fe, Al and Mn release were the same as those on the As mobilization [23], suggesting that the release of As could not be interfered by the releases of Fe, Al and Mn, respectively.

## 5. Conclusions

The following conclusions were drawn from the present study. The concentration of As is positively related to the ammonium-N concentration in groundwater and the %N in sediments.  $\delta^{15}\text{N}$  analysis showed chemical N fertilizer including the combination of the fertilizer and other sources to be a plausible source of N in peat sediments and groundwater. The As release from sediments was done with the mechanism of the “reduction of iron hydroxides and release of adsorbed As from the sediment” based on the negative relationship between the As concentration and ORP of groundwater in the area. The peat sediment, containing a high amount of As, was thought to contribute greatly to the amount of As released under the mechanism. The reducing condition of groundwater was thought to become stronger by the microbial activity, where the increased ammonium-N served as a nutrient for microorganisms. Therefore, the application of N fertilizer seems to have a positive effect on the As concentration in groundwater. From the laboratory experiment, N fertilizer was confirmed to play a positive role in promoting the release of As from peat sediments into water even some extent.

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