Arsenic Behavior in Paddy Fields during the Cycle of Flooded and Non-flooded Periods

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The behavior of As in paddy fields is of great interest considering high As contents of groundwater in several Asian countries where rice is the main staple. We determined the concentrations of Fe, Mn, and As in soil, soil water, and groundwater samples collected at different depths down to 2 m in an experimental paddy field in Japan during the cycle of flooded and non-flooded periods. In addition, we measured the oxidation states of Fe, Mn, and As in situ in soil samples using X-ray absorption near-edge structure (XANES) and conducted sequential extraction of the soil samples. The results show that Fe (hydr)oxide hosts As in soil. Arsenic in irrigation waters is incorporated in Fe (hydr)oxide in soil during the non-flooded period, and the As is quickly released from soil to water during the flooded period because of reductive dissolution of the Fe (hydr)oxide phase and reduction of As from As(V) to As(III). The enhancement of As dissolution by the reduction of As is supported by high As/Fe ratios of soil water during the flooded period and our laboratory experiments where As(III) concentrations and As(III)/As(V) ratios in submerged soil were monitored. Our work, primarily based on data from an actual paddy field, suggests that rice plants are enriched in As because the rice grows in flooded paddy fields when mobile As(III) is released to soil water.

Introduction

Recently, naturally high concentrations of As in groundwater in the Ganges delta plain (Bangladesh and West Bengal in India) have received significant concern (*1−3*). Groundwater is used not only for drinking, but also for irrigation of paddy fields, which produces the staple food in the region. Arsenic concentration is generally less than 10 mg/kg in the surface soil in Bangladesh, whereas the concentration is greater than 40 mg/kg in the soil that receives irrigation with the As-contaminated water (*4*). An incorporation of As from soils to rice would amplify the risk of As poisoning among people

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living in the area. Previous studies suggested that As concentration is greater in rice grains that grow in As-contaminated soils (*4−6*). Therefore, it is important to understand the behavior of As in paddy fields, but there are few systematic studies including the application of in-situ spectroscopic methods about the behavior of As in actual paddy fields. A paddy field has a distinct cycle of flooded and non-flooded periods that are accompanied by redox changes. The redox change should have a significant impact on the behavior of As in water, soil, and plants, since the mobility of As is redox-sensitive (*2, 7−10*).

This paper presents the results of a study on the behavior of As in a well-controlled experimental paddy field in NIAES (National Institute for Agro-Environmental Sciences) in Tsukuba, Japan. The paddy field is designed for sampling of soil water and groundwater at different depths, where the redox condition of the paddy field has been monitored for many years (*11*). We determined the concentrations of Fe, Mn, and As in the soil and water samples and also the oxidation states of these elements in the soil samples in situ using XANES (X-ray absorption near-edge structure) analysis. XANES has often been used to characterize the species of As in sorption studies in laboratory experiments (*12−15*, also see a review by Brown and Sturchio (*16*)) but not widely for As in natural samples except for As in mine tailings (*17−19*). This is the first study of As species in a paddy field using XANES technique.

The present results suggest that the As behavior is partly controlled by Fe (hydr)oxide, but that the reduction of As(V) to As(III) also contributes to the sharp increase of As concentration in soil water during the flooded period.

Experimental Setup and Sampling

The experimental paddy field (10 m × 50 m) is located in Tsukuba, Japan, which is specifically designed to study the behavior of trace elements in paddy soils (*11*). Rice plants have been planted every year in the field under a similar set up and water management since 1974. The area is on Tsukuba-Inashiki plateau, the diluvial upland, underlain by Kanto Loam formation. The site of the paddy field consists of fine-grained Gray Lowland soil. The layer at the depth between 0 and 0.60 m is alluvial soil dressed in 1974 (Figure 1). The Loam layer occurs at a depth between 0.60 and 1.33 m, and a transitional zone is located between 1.33 and 1.70 m depth. The deeper layer (>1.70 m) is Joso Clay layer and sandy soil of the Ryugasaki Formation (Figure 1). Soil horizons pertinent to this study are listed in Table 1 of the Supporting Information.

The air temperatures in the area are high (25−30 °C) in August and low (0−5 °C) in January. The paddy field is flooded in the beginning of May and drained at the end of August. The irrigation water is supplied from a large reservoir in NIAES (volume 5000 m³) whose water is introduced from the Sakuragawa River. Rice plants (*Oryza sativa* L. var japonica) are planted after the irrigation and harvested in the beginning of September. The groundwater table is at a depth between 0.90 and 1.60 m in the non-flooded period, since Joso Clay layer is impermeable. The flow rate of groundwater was measured to be 6.5×10^{-3} and 4.2×10^{-3} cm/s around 7−8.5 and 29−30 m depth.

Soil water was collected at depths of 0.20, 0.50, 1.0, 1.5, and 2.0 m by placing porous ceramic cups connected to glass filters and vacuum pumps (−62 kPa). Groundwater was collected at wells at depths of 2.6 and 12 m. Seven Pt electrodes are buried at depths of 0.10, 0.20, and 1.0 m to
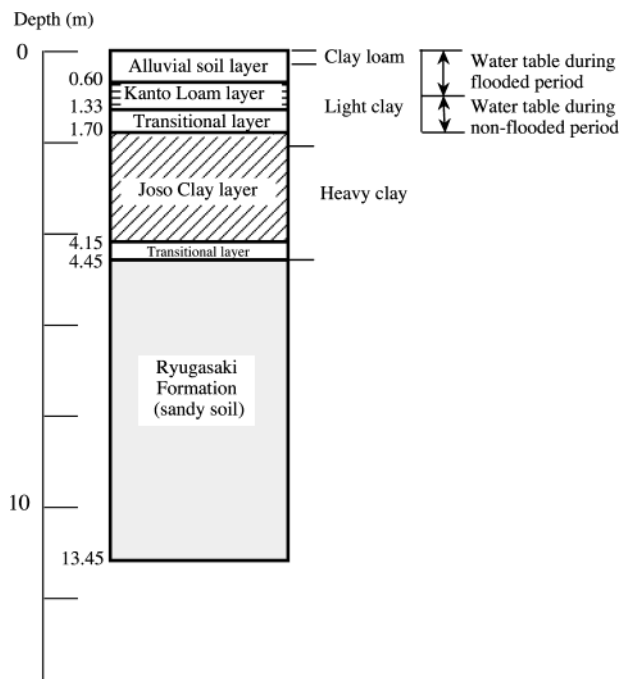
FIGURE 1. Schematic column section of layered soil horizons in the paddy field used in this study.

monitor Eh values in the area covering 10 m × 5 m. Our samples were obtained in this well-calibrated area.

Water samples were collected approximately every 3 months from August 2000 to October 2001. The collection of water (2 L) took less than a day during the flooded period but more than several days during the non-flooded period. Water samples were acidified to 2% HNO_3 solution by adding concentrated HNO_3. Surface soil sample at approximately 0.10 m depth (plowed horizon) was collected during flooded (August 21, 2000) and non-flooded periods (April 26, 2001) for XANES and sequential extraction studies. The samples for XANES measurement were kept frozen in a plastic bag filled with Ar gas, and the XANES measurement was conducted directly on the frozen samples in the plastic bag. The bag contains interstitial water as well as soil, but the contribution of As in interstitial water, $\leq 5 \mu g/kg$, to the result is negligible considering the much higher concentration of As in soil (≥ 10 mg/kg).

Experimental Procedures

Water Analyses. Iron contents in water sample were measured by inductively coupled plasma emission spectrometry (ICP-AES; Yanaco, UOP-1S). Manganese, Zn, and As contents were measured by inductively coupled plasma-mass spectrometry (ICP-MS; VG, PQ-3) using Ge and In as internal standards. The mass interference to ^{75}As by $^{40}Ar^{35}Cl$ was evaluated from the peaks of 77 ($^{40}Ar^{37}Cl$ and ^{77}Se) and 78 (^{78}Se), but the contribution of $^{40}Ar^{35}Cl$ to mass 75 was negligible during the study. The precision of the measurement was less than 10% for Fe and less than 5% for Mn, Zn, and As. The standard river water sample (JAC0031) from the Japan Society for Analytical Chemistry was used to check the accuracy of the analysis. The differences between our results and the recommended values for all elements were less than 10%. Phosphate concentration was analyzed by ion-exchange chromatography (pump, JASCO PU-1580; oven, JASCO CO-1565; conductivity detector, Shodex CD-5; column, Shodex IC I-524A).

Soil Analyses. Concentrations of major elements and Zn for soil samples were measured on fused disks made from a mixture of sample (2 g) and flux (0.8 g of $Li_2B_4O_7$ + 3.2 g

of $LiBO_2$: Johnson Matthey Spectro flux 100B and 0.60 g of $LiNO_3$) using X-ray fluorescence spectrometry (Rigaku ZSX-101e) after drying overnight at 110 °C, following the procedure described in Kanazawa et al. (20). Arsenic concentration in soil was determined by the ICP-MS. Approximately 0.1 g of soil sample was digested with a mixture of concentrated HNO_3 (1.5 mL) and 1:1 H_2SO_4 (1.5 mL) overnight, and the solution was redissolved into a mixture of HNO_3 (0.5 mL) and $HClO_4$ (0.3 mL) after overnight dryness. The solution was filtered, and the filtrate was diluted by 2% HNO_3 for measurement with ICP-MS. The As concentration for JSO-1, a reference soil sample from the Geological Survey of Japan, yielded 7.9 mg/kg, while the recommended value is 8.1 mg/kg (21).

XANES Spectroscopy. The K-edge XANES spectra of Fe, Mn, and As were measured at BL-12C in Photon Factory, KEK, Tsukuba, Japan (22). Incident X-rays were monochromatized with a Si(111) double-crystal monochromator and focused to 1 × 0.5 mm² with a bent cylindrical mirror, which also reduces higher orders of X-rays. All spectra were collected in the fluorescence mode using a 19-element Ge semiconductor detector (SSD) (23). Energy calibration was made by preedge peak maximum of hematite fixed at 7.113 keV for Fe, preedge peak maximum of δ-MnO_2 at 6.539 keV for Mn, white line peak maximum of As_2O_3 at 11.866 keV for As. The relative energy to each peak used for the calibration was employed to display XANES spectra in this paper. The energy step was typically 0.25 eV in the XANES region. The energy of the peak did not shift more than 0.25 eV throughout all measurements.

Sequential Extraction. The sequential extraction procedure is essentially identical to the method by Tissier et al. (24). A 1.0 g amount of air-dried soil sample was used, and the concentration for this experiment refers to the original weight. Exchangeable cations were extracted into Leachate 1 using sodium acetate solution (1.0 M, pH 8.2, 8.0 mL). Leachate 2 contains elements in carbonates that were dissolved in 8.0 mL of sodium acetate solution (1.0 M, pH 5.0). Elements bound to Fe—Mn (hydr)oxide (Leachate 3) were extracted by 25 mL of hydroxylammonium chloride solution (0.10 M, pH 2.0) followed by 25 mL of sodium oxalate solution (0.20 M). Elements bound to organic matter were leached by a mixture of 3.0 mL of 0.020 M HNO_3 and 5.0 mL of 30% H_2O_2 (pH 2.0) at 85 °C for 2 h and for an additional 2 h after adding 3.0 mL of 30% H_2O_2 solution (Leachate 4). After cooling to room temperature, 5.0 mL of ammonium acetate solution (3.2 M) was added. The residue was digested with a 3:1 mixture of 38% HF and 70% $HClO_4$ except for the digestion for As determination.

Laboratory Experiments of the Interaction between Soil and Water. A laboratory experiment was designed to simulate the condition of soil in the paddy field (25, 26). A 20.0 g amount of the surface soil at 0.10 m depth was placed in a plastic columnar bottle (inner diameter, 2.67 cm), which has 19.04 g dried weight at 110 °C. The soil was mixed with 11.98 g of Milli-Q water to produce flooded conditions. The amount of water was selected to attain 180% of water saturation (W_s) defined as W_s (%) = 100 V_w/V_s, where V_w is the total volume of water and V_s is the volume of soil. Five identical samples were prepared and kept at 25 °C. Eh and pH were measured by a Pt electrode and a glass electrode, respectively, by inserting them at a depth of 2 cm from the surface of the soil after 1, 6, 11, 16, and 21 days. The soil solution was sampled using a plastic syringe equipped with the subsurface glass filters. The water samples were immediately filtered by membrane filters (0.45 μm) for the measurement of As(III)/As(V) ratio with HPLC—ICP-MS, as described in Takahashi et al. (26). The soil samples after the extraction of water were kept frozen in the plastic bottle for the in-situ measurements of As(III)/As(V) using XANES as described before.
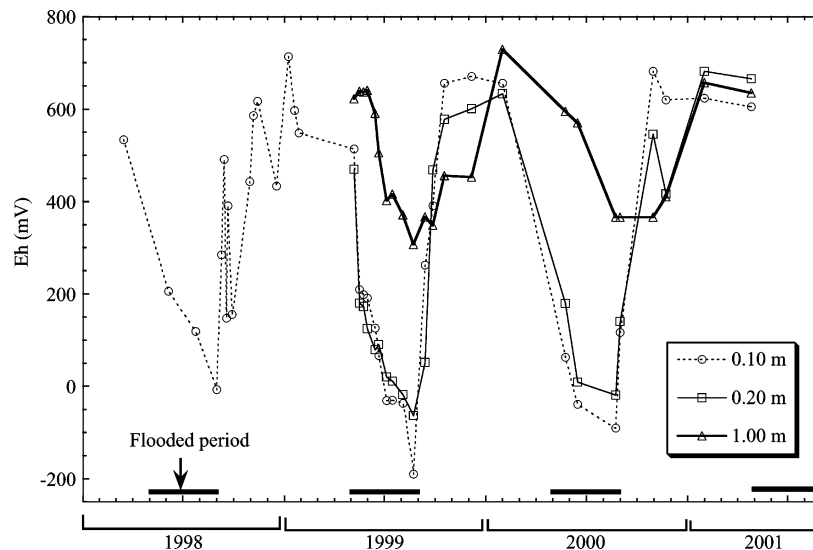
FIGURE 2. Eh variation during the period from March 1998 to May 2001 at depths of 0.10, 0.20, and 1.00 m in the paddy field.
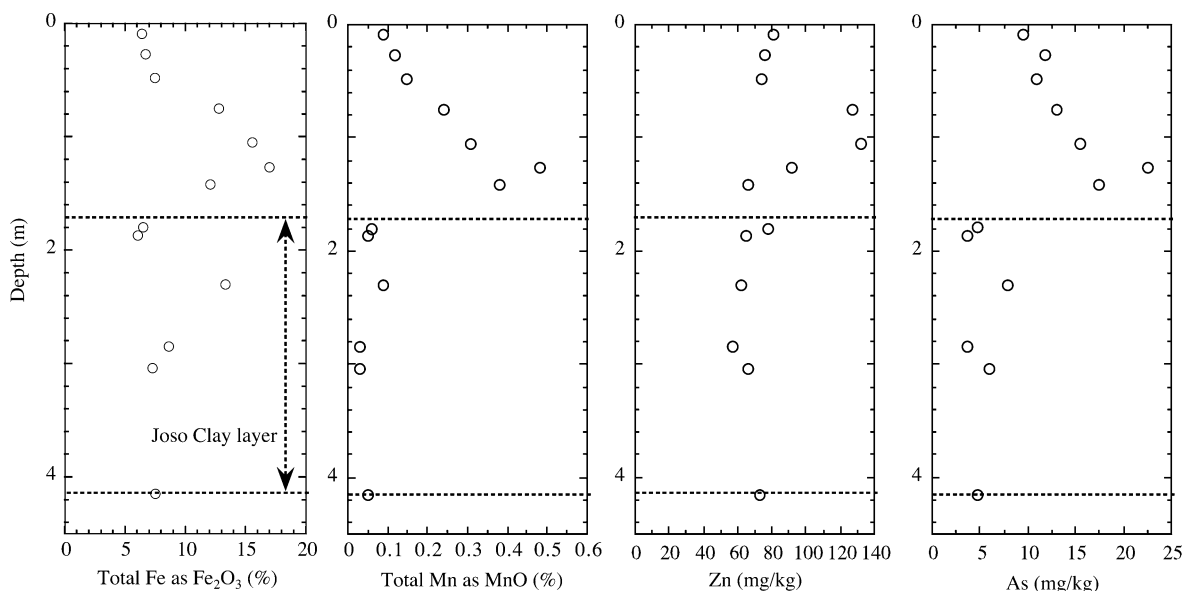

FIGURE 3. Depth profiles of Fe (total Fe as Fe$_2$O$_3$), Mn (total Mn as MnO), As, and Zn concentrations of soil. Note that the soil layer above Joso Clay layer is permeable.

Results and Discussion

Eh Variation in the Cycle of Flooded and Non-flooded Periods. The Eh values are constantly high around 600 mV during the non-flooded period and decrease after the introduction of water at the beginning of May (Figure 2). The Eh values continuously decrease until the end of August, probably because of increased bacterial activity during summer months. The Eh variation is profound at a depth of 0.10−0.20 m compared to the depth of 1.0 m. Although the values are not monitored after April 2001, the Eh variation in 2001 is expected to be similar to that of previous years. The pH values of the soil water show a narrow spread between 6.5 and 7 without any significant change at different depths.

Depth Profiles of Fe, Mn, and As in Soil Samples. Vertical variations of Fe and Mn concentrations in soil, especially above 2 m (Figure 3), are similar to those commonly observed in paddy soils (*27*). Iron and Mn are fixed in soil as Fe(III), Mn(IV), and Mn(III), and their concentrations are lower at shallower depths in the range from 0 to 1.3 m because they dissolve as Fe(II) and Mn(II) under highly reducing condition. High contents of organic matter between 0 and 0.6 m depths (see Supporting Information for the content of organics) result

in reductive conditions at shallower depths during the flooded period. The reductive conditions of the soil at shallow depths do not extend to the deeper levels during the flooded period, as shown by the relatively high Eh condition at a depth of 1.0 m (Figure 2). The presence of Fe and Mn oxides maintained relatively oxic conditions around 1.3 m depth. Soil deeper than a depth of ca. 1.5 m is always below the water table and under reduced conditions, which leads to the low Fe and Mn contents in soil at a depth around 1.80 m. The depth profile for As is very similar to those of Fe and Mn (Figure 3), suggesting that the behavior of As is closely related to those of Fe and Mn in the paddy soil. The depth profile of Zn is also comparable to those of Fe, Mn, and As. This is consistent with earlier studies showing that Zn behavior is primarily governed by Fe and Mn (hydr)oxides (*28, 29*). Full chemical data of soils are listed in Table 1 in the Supporting Information.

To identify the phases hosting As, sequential extraction was employed for surface soils (0.10 m depth) collected during flooded and non-flooded periods. The fractions of Fe, Mn, Zn, and As in leachates are shown in Figure 4. The elements retained in the residues are not included in the calculation
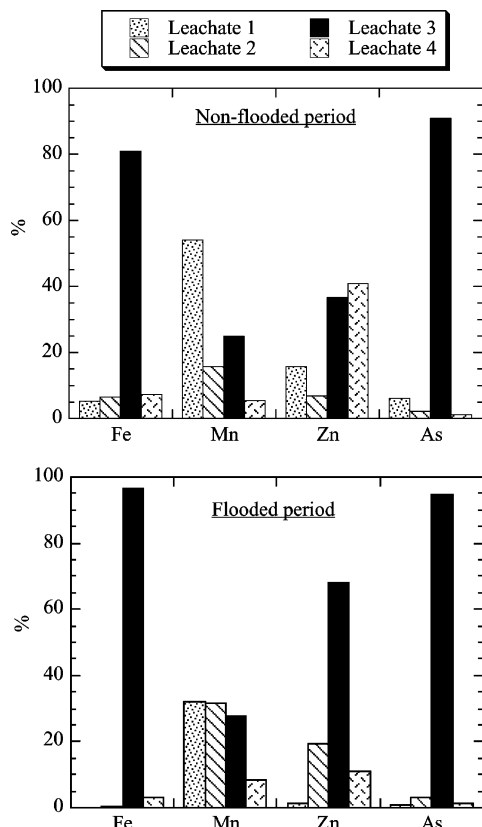
FIGURE 4. Fractions of Fe, Mn, As, and Zn extracted from soil during the sequential extraction experiments. Leachate 1 elements are exchangeable cations in clays, Leachate 2 cations in carbonates, Leachate 3 elements in Fe and Mn (hydr)oxides, and Leachate 4 elements bound to organic matter. Note that the amount of elements retained in the residue after the sequential leaching was not included in the diagram. The residues contained 68.4% of total Fe, 93.9% of total Mn, 73.6% of total As, and 68.8% of total Zn during the non-flooded period and 85.1% of total Fe, 92.1% of total Mn, 71.9% of total As, and 59.9% of total Zn during the flooded period.

because those do not participate to the soil−water interactions (see the caption of Figure 4 for the fraction in the residue). Iron is mainly extracted in Leachate 3 (Fe and Mn (hydr)oxides). Arsenic also is mostly extracted in Leachate 3 under both flooded and non-flooded conditions. The results confirm the interpretation based on the depth profile of elements (Figure 3) that As is hosted by Fe (hydr)oxide in soil.

Dissolved Fe, Mn, and As Concentrations in Soil Water and Groundwater. Irrigation water was collected in August 2000 and January 2001 for the comparison with the soil water and groundwater. The concentrations of Fe and As in the irrigation water are greater than their concentrations in most of the soil water samples during the non-flooded period (Figure 5), suggesting that Fe and As in irrigation water may be incorporated into soil during the non-flooded period. In contrast, soil water shallower than a depth of 1.0 m contains higher concentrations of Fe, Mn, and As than irrigation water during the flooded period, suggesting the release of these ions to the water from the submerged soil. These results suggest an important role of soil−water interactions for the concentrations of these elements in soil water.

The depth profiles of dissolved Fe and Mn concentrations in soil water and groundwater show little Fe and Mn dissolved in water under oxic conditions (Figure 5). In contrast, the reducing water carries high concentrations of these ions during the flooded period. The difference is profound at a depth of 0.20−0.50 m, where the most reducing condition

is attained (Figure 2). This is consistent with the enrichment of Fe and Mn in the soil between 0.6 and 1.30 m (Figure 3). The data suggest reductive dissolution of Fe and Mn from soil during the flooded period.

Temporal variation of Fe concentration shows less dissolved Fe in June 2001 than August 2001. This is most likely explained by the progressive reduction of soil during the flooded period, lowering Eh from May to August (Figure 2).

To understand the different behaviors of Fe and Mn, XANES was applied to soil samples collected under flooded and non-flooded conditions (Figures 6 and 7). Their oxidation states were evaluated using the shift of the absorption edge because it shifts to a higher energy level at higher oxidation states (30−33). The similarity between the spectra of soil samples and those of reference materials suggests that Mn in soil is mostly tetravalent during the non-flooded period and that it is reduced to lower valency during the flooded period (Figure 6). The average oxidation state of Mn is quantitatively determined using the position of the preedge peak defined as the centroid of Gaussian function (23) (Figure 6). According to the method, the average oxidation state of Mn is estimated to be 3.8 during the non-flooded period and 2.2 during the flooded period. The flooding in the paddy field causes the reduction of Mn(IV) to Mn(II) or Mn(III) in the soil.

The soil samples under flooded and non-flooded conditions show similar Fe K-edge XANES spectra (Figure 7). This suggests that Fe has a similar oxidation state during the flooded and non-flooded periods because the preedge position of the spectra reflects the average oxidation state of Fe (32, 33). Comparison of our soil sample data with XANES spectra of reference materials in Figure 7 shows that the Fe species in the two soils are present as ferrihydrite, which exhibits little change during the flooding and nonflooding cycle. The results suggest that Fe(II) formed during the flooding period does not remain in the soil but is dissolved into soil water.

Under flooded conditions, As concentrations increase together with Fe and Mn in the soil water (Figure 5). This confirms earlier laboratory studies showing the release of As from submerged soil to water (7−9, 26). The concentrations of dissolved As and Fe in June 2001 are lower than those in the month of August in 2000 and 2001. This too proves the control of As concentrations by Fe (hydr)oxide and is consistent with the previous studies showing that the effect of Fe redox chemistry is the predominant factor regulating the As behavior in aqueous systems including the high abundance of As in the groundwater of Bangladesh (2). Earlier workers suggested that phosphate enhances the mobility of As in some cases (2, 34). The concentrations of dissolved P in our water samples are lower than the detection limit of 1.0 mg/kg. Recent studies show that P has little influence on the soil−water distribution of As even at higher concentrations of P than those in our study (13). Therefore, it is concluded that P has little effect on the behavior of As in this study.

Although the reductive dissolution of Fe (hydr)oxide is important for the dissolution of As, Zn does not show similar behavior as As in the depth profiles in Figure 5, despite that Zn is likely to be present in Fe (hydr)oxide based on the data of sequential extraction (Figure 4). Decoupling between As and Zn suggests another factor for the large concentration of dissolved As in the soil water. It must be noted that Zn does not change the oxidation state, whereas As itself may also be reduced under flooded conditions. After the release of Zn in the aqueous phase by the reductive dissolution of Fe (hydr)oxide phase, it is likely that Zn is re-adsorbed on other phases such as metal oxides (e.g., aluminum (hydr)oxides and remaining Fe (hydr)oxides) and clay minerals and complexed with organic substances in the solid phase. It is
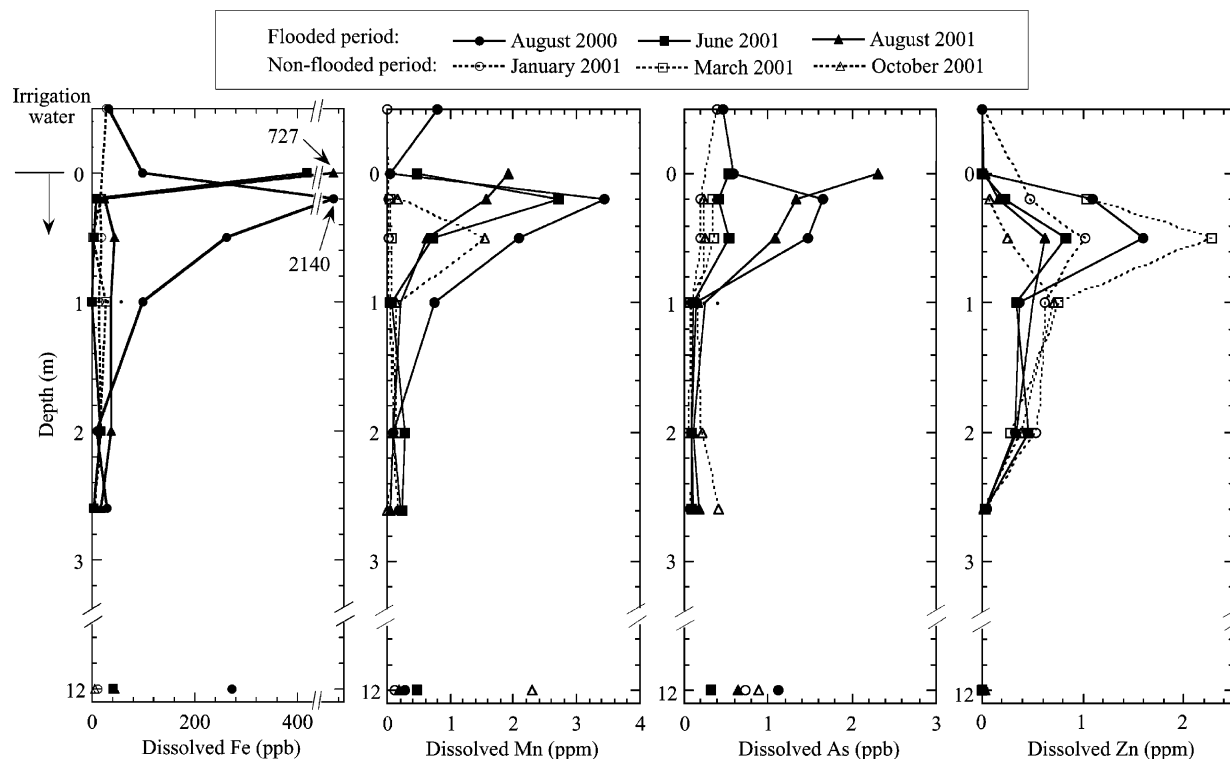
FIGURE 5. Depth profiles of dissolved Fe, Mn, As, and Zn in soil water and groundwater during the flooded (solid lines and filled symbols: August 2000, June 2001, and August 2001) and non-flooded periods (dotted lines and open symbols: January 2001, March 2001, and October 2001). The data on surface waters are plotted at 0 m depth during the flooded period. The data of irrigation water are also shown. Full chemical data of soil water and groundwater are listed in Table 2 in Supporting Information.
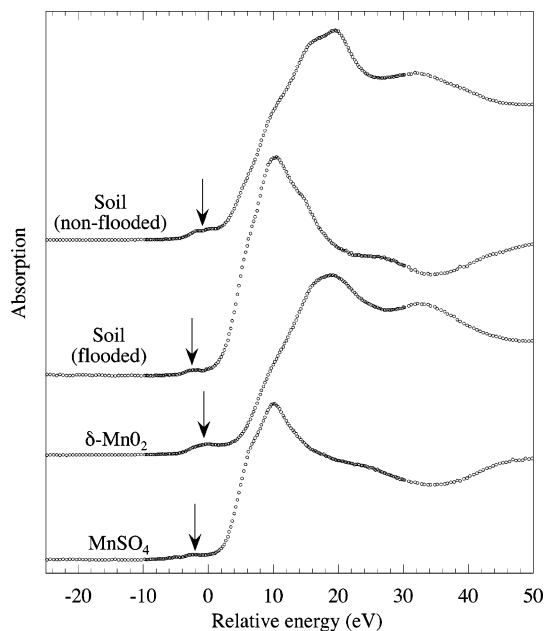

FIGURE 6. Manganese K-edge XANES spectra of soil samples collected during flooded and non-flooded periods, and reference material of $MnSO_4$ and δ-MnO_2. The arrow indicates the position of preedge peak of each spectrum, defined as the centroid of Gaussian function where the preedge structure is determined by the combination of a Gaussian and a quadratic function (*31*).

well-known that K_d (sorption distribution coefficient between sorbent and water) values of As(III) are less than those of As(V) (*2, 26, 35, 36*). Arsenic(III) released from Fe (hydr)oxide is likely retained in the water

Oxidation State of As in Soil and Water. The change of redox state of As in soil was monitored using in-situ
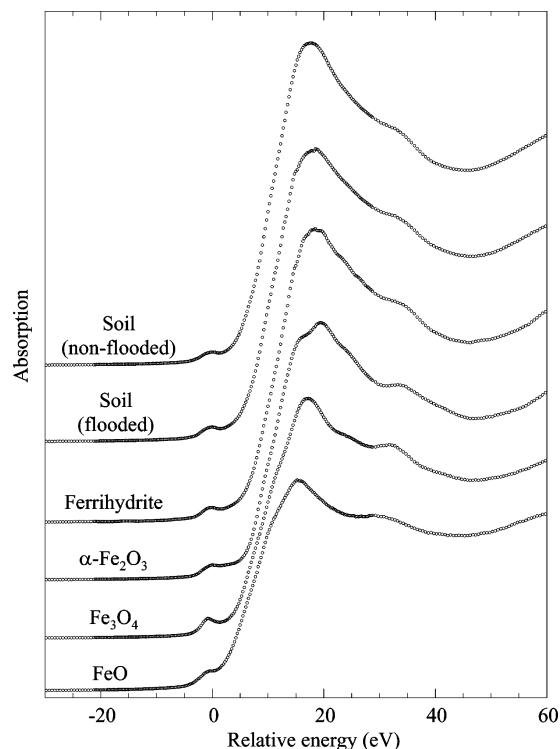

FIGURE 7. Iron K-edge XANES spectra of soil samples collected during the flooded and non-flooded periods with reference material of α-Fe_2O_3, Fe_3O_4, and FeO.

spectroscopic method (*26*). The simulation of the soil sample by the reference materials ($NaAs(III)O_2$ and $NaH_2As(V)O_4$) shows that 30% of As is As(III) under non-flooded conditions and up to 70% under flooded conditions (Figure 8). Preferential partition of As(III) into the aqueous phase compared
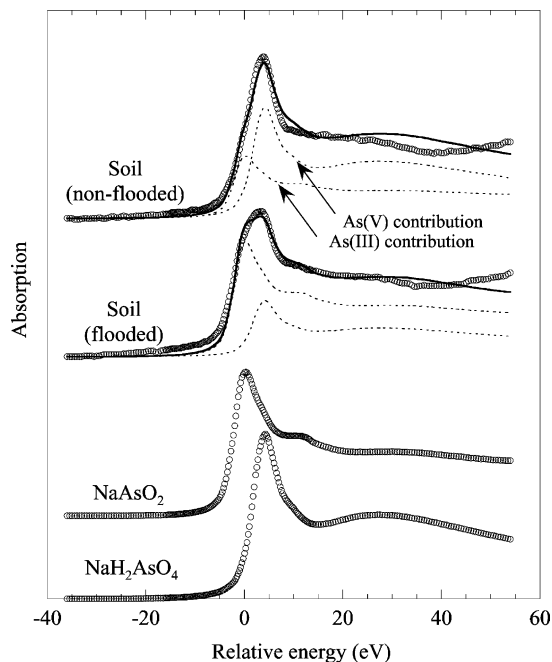
FIGURE 8. Arsenic K-edge XANES spectra of soil samples collected during the flooded and non-flooded periods and reference materials of NaAs(III)O₂ and NaH₂As(V)O₄. Fitted results are shown by solid line, while dotted lines are deconvoluted spectra of As(III) and As(V) species.
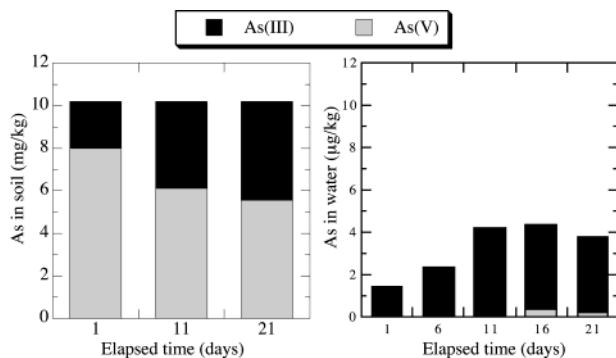

FIGURE 9. Concentrations of total As, As(III), and As(V) in soil and water in the laboratory experiments of As distribution between submerged soil and water.

with As(V) (*2, 26, 35−37*) suggests that this reduction of As in soil explains the increased As concentration in the soil water.

Our proposed interpretation is further supported by our laboratory experiment. It was designed to measure the temporal change of As concentrations and As(III)/As(V) ratios in both soil water and submerged soil following the method described by Yuita et al. (*25*) (Figure 9). The Eh value decreased from 362 mV at the start of the experiment to 135 mV on the 21st day, where pH values were in the range between pH 5.5 and 6.2. The As(III)/As(V) ratio in soil increased with time due to the reducing environment (Figure 9), while the As(III) is predominant in the aqueous phase. The total concentration of As in the water increased with increasing time (except for the sample incubated for 21 days). The results show that As(III) that formed in the soil is released to the water because of its strong tendency to desorb from Fe (hydr)oxide. The experimental results confirm our proposed interpretation for the behavior of As in paddy fields that an increase in dissolved As under the reduced condition during the flooded period results from the desorption property of As(III).

Comparison of Two Factors Controlling the Behavior of As. The dissolution of As in the paddy field is controlled by the reductive dissolution of Fe (hydr)oxide and reduction of As(V) to As(III). These two factors are semiquantitatively compared using the concentrations of Fe and As extracted during the sequential extraction experiments. Leachate 3, which contained dissolved Fe (hydr)oxide, contained 1.27% Fe and 3.28 mg/kg of As under non-flooded conditions. This corresponds to a molar ratio of As/Fe of 1.92×10^{-4}. The ratios of As/Fe in the soil waters during the flooded period should be comparable to this value if the reductive dissolution of Fe (hydr)oxide is responsible for the release of As. The soil water samples show much higher ratios, 4.48×10^{-3}, 9.55×10^{-4}, and 2.37×10^{-3}, during the flooded period collected in August 2000, June 2001, and August 2001, respectively. High ratios suggest either (i) heterogeneous distribution of As in Fe (hydr)oxide phases or (ii) the reduction of As(V) into As-(III) in the solution. The former implies that Fe (hydr)oxide phases dissolved during the flooded period contained much greater amounts of As, a greater capacity of As sorption by less crystallized Fe (hydr)oxide (*37*). This is, however, discounted by the Fe K-edge XANES, which shows no change in the Fe (hydr)oxide phase between flooded and non-flooded periods. Therefore, we propose that the reduction of As(V) to mobile As(III) causes high concentrations of As in soil waters. This is supported by a high proportion of As(III) even in soil samples determined by As K-edge XANES.

We evaluated how the change in the distribution coefficient of total As by reduction affects the concentration of dissolved As assuming that there are sufficient sorption sites of Fe (hydr)oxide for As. The concentrations of dissolved As, [As]$_{dis}$, is the sum of dissolved As(III) and As(V).

$$[As]_{dis} = [As(III)]_{dis} + [As(V)]_{dis}$$

$$= [As(III)]_{soil}/K_{d_{As(III)}} + [As(V)]_{soil}/K_{d_{As(V)}}$$

$$= [As]_{soil}\{r_{III}/K_{d_{As(III)}} + (1 - r_{III})/K_{d_{As(V)}}\} \quad (1)$$

where r_{II} is the fraction of As(III) in soil with values of 0.7 and 0.3 in the flooded and non-flooded periods, respectively. The $K_{d_{As(III)}}$ and $K_{d_{As(V)}}$ values determined from our laboratory experiments were 1.31 and 23.4 (mL/g), respectively. The values of [As]$_{soil}$ do not change very much between flooded and non-flooded periods. The term $\{r_{III}/K_{d_{As(III)}} + (1 - r_{III})/K_{d_{As(V)}}\}$ changes from 0.259 during the non-flooded period to 0.547 during the flooded period. Therefore, the reduction of As(V) to As(III) accounts for the 2.11-fold increase of dissolved As. Our data of soil water at depths of 0.20 and 0.50 m in the paddy field show an increase of dissolved As by a factor of about 4. Therefore, one-half of the increased concentration of As is explained by the large desorption property of As(III).

Rice farming involves planting rice seedlings in flooded paddy fields. Irrigation maintains the flooded conditions during the main growth period. Our study suggests that As in water is incorporated as As(V) in the soil of paddy fields during the non-flooded period and that it is released to water as As(III) during the flooded period. This implies that rice plants likely incorporate the mobile As(III) from soil water because rice plants grow during the flooded period.

Supporting Information Available

Three tables. This material is available free of charge via the Internet at http://pubs.acs.org.

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