Research Article

Impact of Redox Condition on Fractionation and Bioaccessibility of Arsenic in Arsenic-Contaminated Soils Remediated by Iron Amendments: A Long-Term Experiment

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Iron-bearing amendments, such as iron grit, are proved to be effective amendments for the remediation of arsenic- (As-) contaminated soils. In present study, the effect of redox condition on As fractions in As-contaminated soils remediated by iron grit was investigated, and the bioaccessibility of As in soils under anoxic condition was evaluated. Results showed that the labile fractions of As in soils decreased significantly after the addition of iron grit, while the unlabile fractions of As increased rapidly, and the bioaccessibility of As was negligible after 180 d incubation. More labile fractions of As in iron-amended soils were transformed into less mobilizable or unlabile fractions with the contact time. Correspondingly, the bioaccessibility of As in iron-amended soils under the aerobic condition was lower than that under the anoxic condition after 180 d incubation. The redistribution of loosely adsorbed fraction of As in soils occurred under the anoxic condition, which is likely ascribed to the reduction of As(V) to As(III) and the reductive dissolution of Fe-(hydr)oxides. The stabilization processes of As in iron-amended soils under the anoxic and aerobic conditions were characterized by two stages. The increase of crystallization of Fe oxides, decomposition of organic matter, molecular diffusion, and the occlusion within Fe-(hydr)oxides cocontrolled the transformation of As fractions and the stabilization process of As in iron-amended soils under different redox conditions. In terms of As bioaccessibility, the stabilization process of As in iron-amended soils was shortened under the aerobic condition in comparison with the anoxic condition.

1. Introduction

The widespread contamination of soils with arsenic (As), caused by human activities, is a high environmental and toxicological concern. Several technologies, such as in situ chemical stabilization, phytoremediation, and soil washing, can be used to decrease As pollution in soils [1, 2]. Among them, the in situ chemical stabilization implies the application of stabilizing amendments, which by chemical means reduces As mobility and bioaccessibility [3] and refer to the portion of As that can reach into groundwater [4]. Nowadays, iron-bearing amendments, such as iron oxides, iron salts, zero-valent irons, and iron-rich industrial byproducts, are proved to be effective in remediating As-contaminated soils [2, 5–7].

Among zero-valent irons, nanoscale zero-valent iron and iron grit are the two most prevalent soil remediation agents [4].

The remediation efficiency of iron amendments such as iron grit to As-contaminated soils is commonly dependent on the proportions of labile and unlabile As fractions accounting for the total As in soils. Thus, it is essential to understand the stabilization/aging processes of As related to the change of As fractions in soils. And the stabilization/aging processes of As in soils not only depended on the properties of amendments but also were affected by some environmental conditions, such as temperature and redox conditions [5, 8–10]. For instance, the crystallization of Fe (hydr)oxides and the diffusion of As in micropores from the outer layers into the inner complex are accelerated after the temperature

TABLE 1: Physicochemical properties of the experimental soil.

pH	7.67
Organic matter (g/kg)	28.7
Total Fe (g/kg)	43.1
Free iron oxides (g/kg)	17.0
Amorphous iron oxides (g/kg)	6.7
Total Al (g/kg)	93.7
Total Mn (g/kg)	0.627
CEC (cmol/kg)	12.7
Particle size (%)	
Sand (>0.05 mm)	31.8
Silt (0.002–0.05 mm)	55.6
Clay (<0.002 mm)	12.6
As fractions (mg/kg)	
F1	0.045
F2	0.082
F3	0.199
F4	2.59
F5	1.77
F6	16.5

elevated, resulting in the transformation of labile fractions of As into unlabile fractions [5, 9]. The forms of As and Fe-hydroxides in soils are commonly affected by the redox conditions [10, 11]. However, the effect of redox condition on the stabilization processes and bioaccessibility of As in soils amended by iron grit are little known.

The present study aims to (1) investigate the effect of redox condition on As fractions in As-contaminated soils remediated by iron grit in a long term and (2) evaluate the bioaccessibility of As in remediated (iron grit) soils and unremediated soils. Fractions of As in soils were extracted by the sequential extraction procedure (SEP). The results will be beneficial to understanding of the stabilization processes and remediation efficiency of As in soils amended by iron grit under different redox conditions.

2. Materials and Methods

2.1. Soil and Amendment Characteristics. The soils were collected from the upper 10 cm of an agricultural land irrigated by sewage in South China. Soil samples were air-dried, ground, and passed through a 0.9 mm sieve after discarding debris and finally homogenized in advance of analysis. The detailed physicochemical properties of the experiment soil are shown in Table 1, and the related analytical methods have been discussed in our previous publication [11]. The iron grit used for soil additive was purchased from Tianjin evergreen chemical reagent manufacturing Co., Ltd. (China).

2.2. Soil Preparation and Handling. Three samples were taken from the above composite soil with 300 g each. The first and the second ones were spiked by Na_3AsO_4 solution and made the concentrations of As in soils up to 100 mg/kg and then added with 6 g of iron grit (2%, w/w) and mixed immediately.

The first one was added with relatively small volume and high concentration of As solution to make the soil moisture equal to field capacity, which is labeled as S_{IA} . The second one was spiked with relatively big volume and low concentration of As solution to make the soil supersaturated, which is denoted as S_{IN} . The third one was treated with the same process of S_{IN} but with no addition of iron grit and is denoted as S_N . S_N and S_{IN} represent soils under the anoxic condition, and the latter one added iron grit. By contrast, S_{IA} with the addition of iron grit represents soils under the aerobic condition. All samples were incubated in an artificial climate chamber with a temperature of 25°C and a humidity of 65%. S_{IA} was opened and maintained at the soil moisture of field capacity, while S_N and S_{IN} were sealed and maintained a supersaturated condition. At different times (0 d, 3 d, 10 d, 30 d, 60 d, 120 d, and 180 d) after incubation, soil samples were collected and freeze-dried for 24 h in a vacuum before the extraction of As fractions [12].

2.3. Sequential Extraction Procedure and Quality Control. The dried samples were ground and mixed after being collected, and 1.00 g of samples was weighted into acid-washed 50 ml centrifuge tubes. All sample analyses were performed in duplicate. The fractionation of As in soils was determined by one modified Tessier's SPE [11]. Six fractions are denoted as F1, F2, F3, F4, F5, and F6 and are corresponding to water soluble fraction, exchangeable fraction, loosely adsorbed fraction, strongly adsorbed and organically bound fraction, Fe and Mn/Al oxides bound fraction, and the residual fraction. The details are shown in our previous study [11]. In this study, all reagents were analytical grade or better. The concentrations of As in the extractions were determined using an atomic fluorescence spectrophotometer (AFS-610A model, Beijing, China). A standard reference soil GSS-16 (National Center for Standard Reference Material, Beijing, China) was used for quality control of the acid digestion. The recovery for As in the standard reference soil GSS-16 was 98.7%. The errors for the sum of As measured in all six fractions to a single total As determination were within $\pm 18.5\%$.

3. Results and Discussion

3.1. Influence of Iron Grit Amendment on As Fractions in Soils. As shown in Figure 1, there was significant difference for all As fractions between iron grit amended soils (S_{IN}) and unamended soils (S_N) at initial time (0 d). It is worth mentioning that the determination of As fractions at initial time was after 24 h freeze-drying in this study. The labile fractions such as F1, F2, and F3 in S_N accounted for 32%, 14%, and 27% of the total As, respectively, and were far higher than that in S_{IN} . In contrast, the less mobilizable fractions such as F4 and F5 in S_N accounted for 11% and 4% of the total As, respectively, and were far lower than that in S_{IN} . This indicates that iron grit has a rapid effect on the decrease of labile fractions of As in soils and a large proportion of labile fractions of As in soils transformed rapidly into less mobilizable fractions after the addition of iron grit. This is likely to be ascribed to the rapidly increase of iron oxides in

FIGURE 1: The fractions of As in iron grit amended and unamended soils at different time under the anoxic condition ((a) S_N , (b) S_{IN} ; F1—water soluble fraction, F2—exchangeable fraction, F3—loosely adsorbed fraction, F4—strongly adsorbed and organically bound fraction, F5—Fe and Mn/Al oxides bound fraction, and F6—residual fraction).

soils after the addition of iron grit, because iron oxides have strong adsorbed ability on water soluble As [13, 14]. After the incubation, F1 in both of S_N and S_{IN} decreased significantly within 120 d and were from 32% and 9% of the total As to 4% and less than 1%, respectively. F2 in S_N decreased from 14% to 3% and reached a steady state within 60 d, while it in S_{IN} was negligible during the whole incubation period. F3 in S_N decreased from 27% to about 10% and reached a steady state within 30 d, while it was increased firstly from 4% to 7% within 3 d and then decreased to 2% within 60 d in S_{IN} . F4 increased from 11% to about 49% within 30 d and then decreased slowly to 44% in the later incubation (60 d–180 d) in S_N . By contrast, F4 in S_{IN} was shown a remarkable decrease from 63% to 31% within 180 d. F5 in both of S_N and S_{IN} increased within 180 d and were from 4% and 11% of the total As to 22% and 18%, respectively. F6 in S_N increased from 12% to 17% and reached a steady state within 60 d, while in S_{IN} a remarkable increase from 13% to 48% within 180 d was exhibited. Compared to As fractions in S_N , the proportions of F1, F2, F3, F4, and F5 in S_{IN} were lower, while the proportion of F6 in S_{IN} was higher after the incubation. This indicates that iron grit is an effective amendment to stabilize As fractions in soils.

The sum of weakly bound fractions of As after amended is usually used to assess the effectiveness of amendments reducing As bioaccessibility in soils [15, 16]. In this study, the sum of F1, F2, and F3 to the total As, denoted as F_M , represents the bioaccessibility of As in soils [9]. F_M in both of S_N and S_{IN} showed a significant decrease from 73% and 13% to 17% and 2%, respectively, and reached a steady state within 60 d (Figure 2). Therefore, in terms of the As bioaccessibility,

FIGURE 2: Changes of As bioaccessibility in soils under anoxic conditions (unamended soil, S_N ; iron grit amended, S_{IN}).

the stabilization/aging processes of As in both of S_N and S_{IN} occur and end within 60 d, and the addition of iron grit is more effective on the stabilization of As in soils.

3.2. Effect of Redox Conditions on As Fractions in Iron-Amended Soils. F1 in both of S_{IA} and S_{IN} decreased markedly within the first 10 d following the addition of water soluble As and iron grit (Figure 3). By contrast, in the incubation

FIGURE 3: Changes of As fractions in iron grit amended soils under different redox conditions over time (anoxic condition, S_{IN} ; aerobic condition, S_{IA}).

time of 60 d to 180 d, the changes of $F1$ in these two soils were distinct; in this period, $F1$ exhibited further decreases at slow rates and the tendency was not complete at the end of incubation in S_{IN} , while $F1$ in S_{IA} had no significant change in 60 d–180 d. This indicates that the transformation of $F1$ into less mobilizable fractions in iron-amended soils under the anoxic condition is a long-term (>180 d) process, while it is a short-term (<60 d) process in iron-amended soils under the aerobic condition. Moreover, in the whole incubation period, the proportion of $F1$ in S_{IN} was higher than that in S_{IA} , indicating that more soluble As in iron-amended soils transformed into less mobilizable fractions under aerobic condition in comparison with under anoxic condition. This is likely to be attribute to the ongoing reduction of As(V) to As(III) in soils under anoxic condition and the higher adsorption of As(V) than As(III) onto soil components [10, 11, 17, 18].

Different from $F1$, there was no significant difference of $F2$ in both S_{IA} and S_{IN} , and the proportion of $F2$ exhibited a negligible level during the whole incubation time (Figure 3). So it could be speculated that $F2$ was converted to less mobilizable fractions at an extremely fast rate following the addition of iron grit to soils whether in an oxidizing environment or in a reducing environment. This may be ascribed to the rapidly formed Fe oxides after the addition of iron grit into soils, because the rapid adsorption of added As on the surface of newly formed amorphous iron oxides has been found [19].

$F3$ in S_{IA} showed a marked decrease and reached a steady state quickly within the first 10 d (Figure 3). By contrast, $F3$ in S_{IN} exhibited a significant increase within the first 3 d and then decreased, indicating that the redistribution of $F3$ in iron-amended soils under anoxic conditions occurred. The reason for the increase of $F3$ in S_{IN} within the first 3 d may be as follows: on the one hand, pH in S_{IN} may increase within the first 3 d, because soil pH increased slightly after the flood while pH in nonflooded soil showed little change [20]; on the other hand, the reduction of As(V) to As(III) often occurs in soils under anoxic conditions [12], both leading to the increase of $F3$ in S_{IN} within the first 3 d, because As(III) is preferentially nonspecific adsorbed to Fe-hydroxides at alkaline condition [2]. As on external surface of soil minerals gradually diffuses into inner surface and mineral lattice and forms relatively immobilizable surface complexes and insoluble secondary solid phases [10, 21], which may be responsible for the decrease of $F3$ in iron-amended soils with the contact time. The change of $F3$ in S_{IA} ended in the first 10 d while in S_{IN} it ended within 60 d, indicating that the transformation of $F3$ into other fractions in iron-amended soils under the anoxic condition is more lasting than that under the aerobic condition.

Similar to $F1$ in soils, $F4$ also showed a significant decrease in both S_{IA} and S_{IN} , and the decrease of $F4$ was not complete within the incubation time in both S_{IA} and S_{IN} (Figure 3). This indicates that effect of redox conditions on the tendency of $F4$ in iron-amended soils is negligible. The

decomposition of organic matter and the increase of crystallization of Fe oxides with the contact time may be responsible for the decrease of F_4 in iron-amended soils [10, 22, 23]. In addition, the proportion of F_4 in S_{IN} was significantly higher than that in S_{IA} during the whole incubation period. This may be attributed to the more As(V) reduction to As(III) and less decomposition of dissolved organic matters in soils under anoxic condition than that under aerobic condition [8, 11].

Contrary to the four former fractions, F_5 in both S_{IA} and S_{IN} increased promptly within the first 10 d after the addition of water soluble As and iron grit (Figure 3). In contrast, during the later incubation period (10 d–180 d), the changes of F_5 in these two soils were distinct: F_5 in S_{IA} reached the equilibrium within the first 10 d, while in S_{IN} it exhibited a further increase in the later incubation period (10 d–180 d). This indicates that the transformation of F_5 into other fractions in iron-amended soils under the anoxic condition is a long-term (>180 d) process, while it is a short-term (<10 d) process in iron-amended soils under the aerobic condition. The reductive dissolution and reproduction of As-Fe phases in soils occur under reducing conditions, but it is inhibited at oxidative conditions, which may be responsible for the different tendency of F_5 in S_{IA} and S_{IN} [17, 24]. It is worth mentioning that the proportion of F_5 in S_{IN} was significantly lower than that in S_{IA} during the incubation period of 3 d–60 d. This may be ascribed to the reduction of As(V) into As(III) and the poor crystallization of Fe oxides in soils under the anoxic condition [10].

F_6 in both S_{IA} and S_{IN} increased markedly within the first 30 d following the addition of As and iron grit (Figure 3). The changes of F_6 in these two soils in the later incubation period (30 d–180 d) were distinct: F_6 in S_{IN} reached the equilibrium within the first 30 d, while in S_{IA} it showed a further increase at slow rates and the tendency was not complete within 180 d. This indicates that aerobic condition is likely to prolong the formation of F_6 in iron-amended soils in comparison with anoxic condition. This may be attributed to the lasting well crystallization of Fe oxides in iron-amended soils under the aerobic condition. The proportion of F_6 in S_{IN} was significantly lower than that in S_{IA} during the whole incubation period. This indicates that the aerobic condition likely leads to the more increase of F_6 in iron-amended soils over time in comparison with the anoxic condition. Reduction of As(V) to As(III) under the anoxic condition over time may be also responsible for the above effect, because a higher proportion of labile fraction of As(V) commonly transforms into less mobilizable fractions, such as F_6 , in soils over time in comparison with As(III) [10].

3.3. Impact of Redox Conditions on As Bioaccessibility in Iron-Amended Soils. As shown in Figure 4, F_M in both S_{IA} and S_{IN} decreased rapidly within the first 10 d following the addition of As(V) and iron grit into the soils. In contrast, the changes of F_M in these two soils in the later incubation period (10 d–180 d) were distinct: F_M in S_{IA} reached the equilibrium within the first 10 d, while in S_{IN} it showed a further decrease and reached the equilibrium within 120 d. This indicates that anoxic condition would lead to the tendency of F_M in iron-amended soils reaching equilibrium

FIGURE 4: Changes of As bioaccessibility in soils under different redox conditions after the addition of iron grit over time (anoxic condition, S_{IN} ; aerobic condition, S_{IA}).

later in comparison with aerobic condition. In terms of As bioaccessibility, the stabilization process ended within 10 d in S_{IA} , but it was complete within 120 d in S_{IN} , indicating that the anoxic condition would prolong the stabilization process of As in iron-amended soils. In addition, over the whole incubation period, the proportion of F_M in S_{IN} was higher than that in S_{IA} , which demonstrates that the aerobic condition tended to be more conducive to reduced bioaccessibility of As than anoxic condition. It is likely to be coaffected by the reductive dissolution of Fe (hydr)oxides and the reduction of As(V) to As(III) at anoxic condition [25, 26], because As which was coprecipitated and adsorbed on Fe-(hydr)oxides surface could be released due to the reductive dissolution of the Fe oxides and the lower adsorption of As(III) onto the surface of soil minerals than that of As(V) [27].

3.4. Effect of Redox Conditions on the Stabilization Processes of As in Iron-Amended Soils. The stabilization process of As in S_{IN} had two distinct stages in the whole incubation period (Figure 5(a)). The first stage was from 0 d to 3 d, where F_1 and F_4 decreased and, in contrast, F_3 , F_5 , and F_6 increased, while other fractions exhibited no marked change, indicating that F_1 and F_4 transformed into F_3 , F_5 , and F_6 in S_{IN} within the first 3 d, and the main transformation was from F_1 and F_4 to F_6 due to their large variations. In this stage, transformation of F_1 to F_6 in S_{IN} was likely to be affected by the molecular diffusion in micropores of the soil minerals [9, 10]. Transformation of F_4 to F_6 in S_{IN} was coaffected by the decomposition of organic matter and molecular diffusion in the surface of soil minerals [22], because As occluded within organic matters would be released again after the decomposition of arganic matter by microbial respiration under the anoxic condition and then diffused into the mineral

FIGURE 5: The stabilization processes of As in soils at different redox conditions ((a) S_{IN} and (b) S_{IA}).

lattice [10]. The second stage in S_{IN} was from 3 d to 180 d; $F1$, $F3$, and $F4$ decreased and, in contrast, $F5$ and $F6$ increased, while $F2$ was without significant change. This indicates that three more mobilizable fractions transformed into two less mobilizable or immobilizable fractions in the later incubation time in S_{IN} . The main transformation in S_{IN} in the second stage was also from $F4$ to $F6$ due to their large variations, and the decomposition of organic matter and molecular diffusion in the surface of soil minerals should be responsible for this transformation.

Similar to the iron-amended soils under anoxic condition, the stabilization process of As in iron-amended soils under aerobic condition also had two distinct stages in the whole incubation period (Figure 5(b)). The first stage was also within the first 3 d, where $F1$, $F3$, and $F4$ decreased and, in contrast, $F5$ and $F6$ increased, while $F2$ was without significant change. This indicates that three more labile fractions transformed into two unlabile fractions in S_{IA} within the first 3 d. The main transformation in S_{IA} in the first stage was from $F1$ to $F5$ and $F4$ to $F6$ due to their large variations. In this stage, the transformation of $F1$ to $F5$ in S_{IA} may be ascribed to the well crystallization of Fe oxides and As occluded within Fe-(hydr)oxides, because the crystallization of Fe oxides in soils under aerobic conditions would occur rapidly [28]. The transformation of $F4$ to $F6$ in S_{IA} is also likely to be attributed to the decomposition of organic matter and molecular diffusion in the surface of soil minerals. The second stage in S_{IA} was also from 3 d to 180 d; $F1$ and $F4$ decreased and, in contrast, $F6$ increased, while other fractions showed no significant change, indicating the transformation of $F1$ and $F4$ into $F6$ in S_{IA} in the later incubation period. In this stage, the main transformation in S_{IA} was $F4$ to $F6$, and the reason was discussed as in the above.

4. Conclusions

The present study demonstrated that water soluble and loosely adsorbed As was the primary fractions in soils spiked As(V). After the addition of iron grit, the primary fractions of As in soils were characterized by strongly adsorbed and organically bound fractions (~63%), while the water soluble and loosely adsorbed As only account for a small proportion. More labile fractions transformed into immobilizable fractions in iron-amended soils under aerobic conditions in comparison with under anoxic conditions. The redistribution of $F3$ in iron-amended soils under anoxic conditions occurs, as a result of the reduction of As(V) to As(III) and the reductive dissolution of Fe-(hydr)oxides.

The stabilization processes of As in iron-amended soils under the anoxic and aerobic conditions were characterized by two stages. The increase of crystallization of Fe oxides, decomposition of organic matter, molecular diffusion, and the occlusion within Fe-(hydr)oxides cocontrolled the transformation of As fractions and the stabilization process of As in iron-amended soils under different redox conditions. In terms of As bioaccessibility, the stabilization process of As in iron-amended soils was shortened under the aerobic condition in comparison with under the anoxic condition.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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