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Immobilization of mercury and arsenic in a mine tailing from a typical Carlin-type gold mining site in southwestern part of China

Jianxu Wang [a, b], Xuecheng Sun [a], Ying Xing [c], Jicheng Xia [a, d], Xinbin Feng [a, *]

[a] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550082, PR China
[b] CAS Center for Excellence in Quaternary Science and Global Change, Xi'an, 710061, China
[c] School of Chemistry and Materials Science, Guizhou Normal University, Guiyang, 550001, China
[d] University of Chinese Academy of Sciences, Beijing, 100049, China

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ABSTRACT

The potential of chicken manure and ferrous sulfate in immobilizing both mercury (Hg) and arsenic (As) in a tailing collected from Danzhai gold (Au) mine in southwestern part of China was investigated. A laboratory leaching experiment was conducted firstly to study the effectiveness of 30 tons ha^{-1} to 90 tons ha^{-1} chicken manure, or its combination with 1.5 tons ha^{-1} to 15 tons ha^{-1} ferrous sulfate, on the mobilization of Hg and As in the tailing. Results showed that the sole application of chicken manure at dose of 30 tons ha^{-1} to 90 tons ha^{-1} to the tailing removed approximately 94%–99% and 49%–85% of Hg and As, respectively, from the leachate over three months. These reductions might be attributed to the formation of hardly soluble Hg sulfides (e.g., HgS) and As sulfides in the tailing. Addition of chicken manure and ferrous sulfate to the tailing decreased Hg and As concentrations by 60%–85% and 31%–60%, respectively, in the leachate as compared to sole treatment of chicken manure. The mechanisms of Hg and As removal might be linked to their adsorption by iron oxides and chicken manure. The co-application of 30 tons ha^{-1} chicken manure and 15 tons ha^{-1} ferrous sulfate to the tailing showed promising results in reducing Hg and As concentrations as compared to the other treatments. The results from field trial showed that about 73% of Hg and 82% of As were removed from the leachate by the selected amendments (30 tons ha^{-1} chicken manure and 15 tons ha^{-1} ferrous sulfate), as compared to the control. In summary, the co-application of 30 tons ha^{-1} chicken manure and 15 tons ha^{-1} ferrous sulfate was promising in mitigating risk of both Hg and As from tailings, and the knowledge provided might deepen our understanding of mobilization of Hg and As in the tailing in Carlin-type gold mining regions.

1. Introduction

The fast development of economy leads to a high demand of the metal(loid)s and their compounds, which are important components in a wide range of common products such as construction materials, ceramics, aeroplanes, computers, and paint (Kossoff et al., 2014). Therefore, there is a rapid development of mining industries in the last century. However, the discharge of metal(loid)s-containing water (Dudka and Adriano, 1997), exhaust (Li et al., 2014), and solid wastes by the mining and refining (or retorting) activities leads to serious environmental pollution problems (Wang et al., 2018).

Carlin-type gold deposit is a sediment-hosted disseminated gold deposit, and mainly distributed in southwest China. It is characterized by invisible gold in carbonate minerals. Generally, gold is accompanied by an accessory suite of elements such as arsenic (As), mercury (Hg), and others (Ilchik and Barton, 1997). The gold mining and refining activities leads to a widespread pollution of toxic elements (e.g., Hg and As) in the environment (Ning et al., 2011). Tailings produced by those activities usually contain high contents of toxic elements, and their improper management can result in the dispersion of these toxic elements, causing pollution (Sun et al., 2014). Mercury and As are common occurring elements in a Carlin–type gold deposit, and extremely toxic to human (Wang et al., 2018). Their transfers in terrestrial food web can threaten the health of the consumers (e.g., Feng et al., 2008; Liu J et al., 2019).

* Corresponding author.
E-mail address: fengxinbin@vip.skleg.cn (X. Feng).

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Mitigation of pollutant risks from tailings is important for ecological safety (Licskó et al., 1999). Methods, including riverine disposal, submarine disposal, wetland retention, backfilling, dry stacking and storage behind dammed impoundments, have been used to treat tailings (Lottermoser, 2010). The storage of tailings under water to avoid the formation of dusts in a “tailing pond” or “tailing dam” is popular in mining industries in the world (Mendez Monica and Maier Raina, 2008). However, a problem with this method is the maintenance of an approximate water level in a dam, in particular in semi-arid and arid areas. Currently, there is another solution that comprises of immobilization toxic elements and restoration of vegetation to avoid dusts from tailings (Gil-Loaiza et al., 2018). Unlike soils, tailings are poor in nutrients, rich in multiple toxic elements, and contain large particles (Sheoran and Sheoran, 2006). Both the immobilization of toxic elements to decrease their toxicities (Wang J et al., 2019) and the enhancement of nutrients levels to maintain the growth of plants are prerequisites for a successful vegetation restoration in a tailing.

However, the lack of approximate chemical/biological amendments remains a major barrier for immobilization of tailings. Many immobilization agents have been developed to treat As and Hg (Wang et al., 2012) polluted soils. Fe(II) (Seidel et al., 2005), Fe(III) (Kim et al., 2003), humic acid (Wang and Mulligan, 2009), zero valent iron (Kim et al., 2012), and the mixture of Al_2O_3, SiO_2 and CaO (Wang L et al., 2019), have been shown to be efficient in As immobilization. As for Hg, clay minerals and diammonium phosphate (Wang J et al., 2019), ammonium thiosulfate (Liu T et al., 2019, and biochar (Xing et al., 2019) have been used for this metal immobilization in soils. Nonetheless, none of these agents have been tested for tailings in Karst regions where their leachate are quite alkaline (pH > 9.9) due to the enrichment of carbonate (Khan and Jones, 2009). pH plays a crucial role in trace elements solubilization and precipitation in the environment. Mercury adsorption by soil particles was a consequence of an integrated effect of pH and organic matter; low pH (e.g., pH 3–5) favors the adsorption in organic matter-enriched soil; high pH enhances the adsorption in soil with poor organic matter content (Yin et al., 1996). Unlike Hg, As adsorption to soil was largely affected by its speciation and pH. For instance, maximum arsenite adsorption to soil occurs at pH of close to 9, under which arsenite occurred as H_3AsO_3 and soil was negatively charged. H_3AsO_3 can be strongly adsorbed by negatively charged soil particles via hydrogen bonding (Dias et al., 1997). In addition to immobilization, the improvement of biogeochemical environments of tailing is also crucial for the vegetation restoration.

Therefore, the aims of this study were to fit the knowledge gap of developing sustainable immobilizing agents to efficiently treat Hg–As tailing in Karst region in China. The $FeSO_4$ and chicken manure were used as sustainable amendments for both As and Hg in the tailing collected from Danzhai Au mine in China. The $FeSO_4$ is able to decrease pH, and has been demonstrated to be able to immobilize multiple toxic elements in soils and sediments (Serrano et al., 2012); chicken manure has been reported to be capable of reducing the mobility of multiple toxic elements through adsorption, complexation, and precipitation (Chen et al., 2010; Liu et al., 2009), and improving the nutrients levels of soils. This study mainly aimed to (1) study the effectiveness of different doses of $FeSO_4$ and/or chicken manure on the mobilization of Hg and As in the tailing collected from Danzhai Au mine, and screen the optimum doses of the amendments; (2) reveal the mechanisms of Hg and As immobilization by the amendments; and (3) study the performance of the selected amendments under field condition.

2. Material and methods

Danzhai Au mine is located about 15 km southeast of Danzhai city in Guizhou province in China (107°58′E, 26 °09′N). The annual average air temperature is 12.6–17.2 °C, and precipitation is 1260 to 1508 mm for this region. Danzhai Au mine mainly contains Hg–As–Au minerals, and its mining and retorting activities can be dated back to 1950s, and ceased in 1980s. It is roughly estimated that about 186 million tons of wastes were produced, and piled up close to the mining site without any treatment (Su et al., 2010). The sampling site close to Wushui River, and the tailings remained untreated at the moment of sampling (Fig. 1).

2.1. Sample pretreatment

Tailing samples were transported to the laboratory, air-dried, and passed through a 4-mm nylon sieve prior to use. Both the ferrous sulfate ($FeSO_4 \cdot 7H_2O$, Agricultural grade, >99%) and chicken manure were purchased from a chemical company (Dongxin Chemical Co., Guiyang) in China. The chicken manure was air-dried, and passed through a 4-mm nylon sieve in the laboratory. The pH of chicken manure was 6.6, and its organic matter and total sulfur content was 41.6% and 0.1%, respectively. It contains 5.6 mg kg^{-1}As, 0.3 mg kg^{-1} Hg, 3.6 mg kg^{-1} Cr, and 6.3 mg kg^{-1} Pb, which were below their maximum allowable contents in fertilizers set by the Chinese government (GB, 18877-2002; As≤50 mg kg^{-1}, Hg ≤ 5 mg kg^{-1}, Cr ≤ 500 mg kg^{-1}, Pb ≤ 150 mg kg^{-1}).

2.2. Greenhouse experiment

A series of plastic containers (50 cm long × 20 cm wide × 14 cm height) filling with 3 kg of tailings were set in a greenhouse at Institute of Geochemistry, Chinese Academy of sciences. These pots were divided into 16 groups with each had three replicates. The tailing was treated with chicken manure (up to 90 tons ha^{-1}) and $FeSO_4 \cdot 7H_2O$ (1.5–15 tons ha^{-1}) with different combinations. The detailed experimental design is shown in Table 1. The tailing and chicken manure were manually mixed in a plastic bag, and thereafter the ferrous sulfate solution was spiked. The amount of chicken manure and ferrous sulfate spiked to the tailing was calculated on the basis of the weight of amendments per unit area of the pot. For instance, the surface area of the plastic container is 0.1 m^2, and the amount of chicken manure used is 900 g, meaning the spiking of this amendment at a dose of 9 Kg per meter square. Thus, 1 ha (1 ha = 10,000 m^2) of land received about 9×10^{-3} tons × 10,000 m^2/1 m^2 = 90 tons. The doses used in our study were within the recommended rates of 50–200 tons ha^{-1} for the land with hard rock by USEPA (1995), and the calculated amounts of toxic elements inputted into the tailing with chicken manure were 168 g ha^{-1} to 504 g ha^{-1} for As, 9 g ha^{-1} to 27 g ha^{-1} for Hg, 108 g ha^{-1} to 324 g ha^{-1} for Cr, 189 g ha^{-1} to 567 g ha^{-1} for Pb, respectively, which were far below the maximum values (41 kg ha^{-1} for As, 17 kg ha^{-1} for Hg, 3000 kg ha^{-1} for Cr, and 300 kg ha^{-1} for Pb) in the standard of land application pollutant limits for sewage sludge recommended by USEPA (1995).

To collect the leachate from the container, a silicone tube (2-cm in diameter) was connected between a hole at the bottom of the container and a polyethylene terephthalate (PET) bottle (500 mL). All the tubes and PET bottles were pre-cleaned by 5% HNO_3 prior to use. All the containers were carefully provided with approximate amount of deionized water (1.2–1.5 L) to avoid leaching, and then they were left for 30 days at room temperature to be in chemical equilibrium between tailing and amendments (Willard, 1979). After equilibration, about 150 mL of deionized water was provided to each pot every 2 days consecutively for three months. The leachate from each PET bottle was taken every month, and about 300 mL–500 mL of leachate was collected after one month of leaching. All the containers were covered by polyvinyl chloride

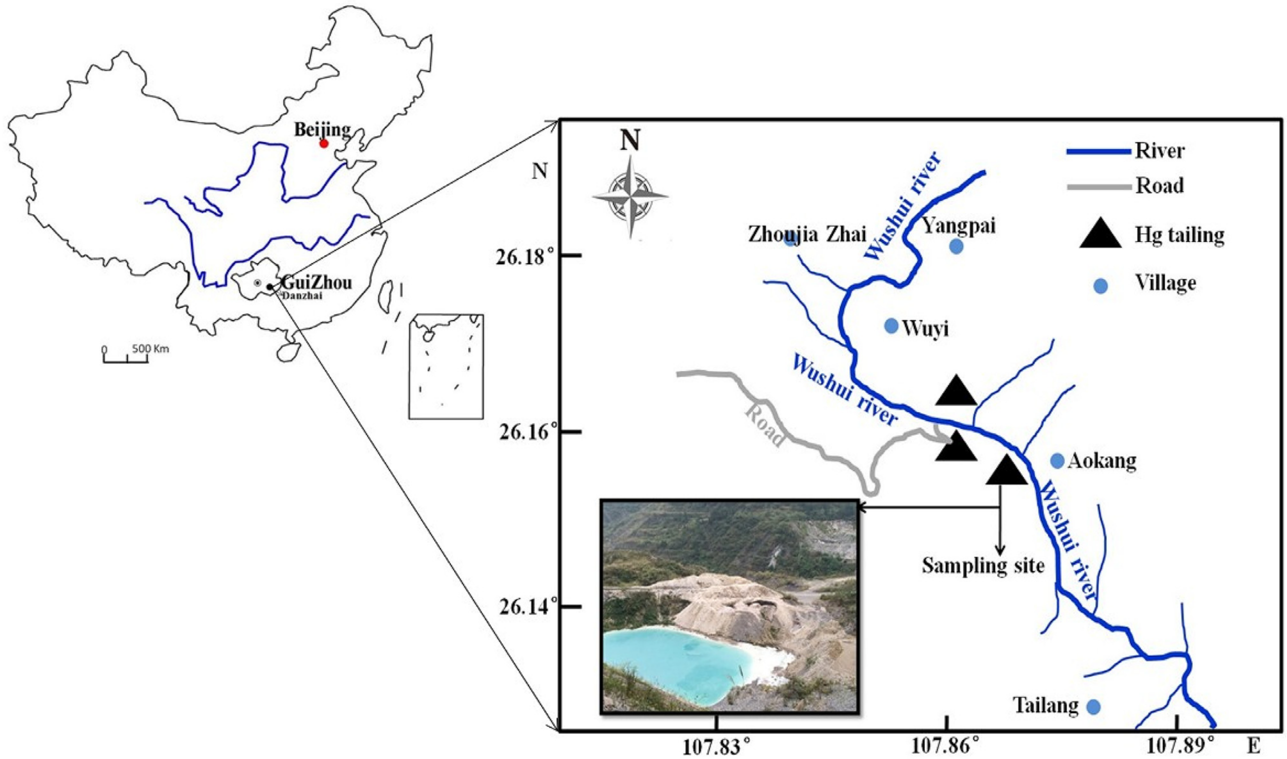

Fig. 1. Tailing sampling site.

Table 1
The description of the treatments.

Group	Treatments	Treatments Abbreviation	Description
1	Control	Control	3 kg Tailing without any treatments
2	Tailing +30 tons ha^{-1} CM	CM1	3 kg Tailing +300 g chicken manure
3	Tailing+ 30 tons ha^{-1} CM +1.5 tons ha^{-1} FS	CM1+FS1	3 kg Tailing +300 g chicken manure +15 g $FeSO_4 \cdot 7H_2O$
4	Tailing +30 tons ha^{-1} CM +3 tons ha^{-1} FS	CM1+FS2	3 kg Tailing +300 g chicken manure +30 g $FeSO_4 \cdot 7H_2O$
5	Tailing +30 tons ha^{-1} CM +9 tons ha^{-1} FS	CM1+FS3	3 kg Tailing +300 g chicken manure +90 g $FeSO_4 \cdot 7H_2O$
6	Tailing +30 tons ha^{-1} CM +15 tons ha^{-1} FS	CM1+FS4	3 kg Tailing +300 g chicken manure +150 g $FeSO_4 \cdot 7H_2O$
7	Tailing +60 tons ha^{-1} CM	CM2	3 kg Tailing +600 g chicken manure
8	Tailing +60 tons ha^{-1} CM +1.5 tons ha^{-1} FS	CM2+FS1	3 kg Tailing +600 g chicken manure +15 g $FeSO_4 \cdot 7H_2O$
9	Tailing +60 tons ha^{-1} CM +3 tons ha^{-1} FS	CM2+FS2	3 kg Tailing +600 g chicken manure +30 g $FeSO_4 \cdot 7H_2O$
10	Tailing +60 tons ha^{-1} CM +9 tons ha^{-1} FS	CM2+FS3	3 kg Tailing +600 g chicken manure +90 g $FeSO_4 \cdot 7H_2O$
11	Tailing +60 tons ha^{-1} CM +15 tons ha^{-1} FS	CM2+FS4	3 kg Tailing +600 g chicken manure +150 g $FeSO_4 \cdot 7H_2O$
12	Tailing +90 tons ha^{-1} CM	CM3	3 kg Tailing +900 g chicken manure
13	Tailing +90 tons ha^{-1} CM +1.5 tons ha^{-1} FS	CM3+FS1	3 kg Tailing +900 g chicken manure +15 g $FeSO_4 \cdot 7H_2O$
14	Tailing +90 tons ha^{-1} CM +3 tons ha^{-1} FS	CM3+FS2	3 kg Tailing +900 g chicken manure +30 g $FeSO_4 \cdot 7H_2O$
15	Tailing +90 tons ha^{-1} CM +3 tons ha^{-1} FS	CM3+FS3	3 kg Tailing +900 g chicken manure +90 g $FeSO_4 \cdot 7H_2O$
16	Tailing +90 tons ha^{-1} CM +15 tons ha^{-1} FS	CM3+FS4	3 kg Tailing +900 g chicken manure +150 g $FeSO_4 \cdot 7H_2O$

CM, chicken manure; $FeSO_4 \cdot 7H_2O$, FS.

(PVC) films to avoid the contamination from dusts. The leachate were filtered with 0.45-μm cellulose filter membrane (Whatman, GE Healthcare Bio-Sciences, PA), and thereafter they were divided into four subgroups for total As, total Hg, dissolved organic carbon (DOC), and anions analyses. The subsamples for As and Hg determination were acidified with HNO_3 3% and HCl 3% (v/v), respectively.

2.3. Field trial

Two plots (2 m^2 for each) were established at Danzhai Au mine to investigate the performance of the amendments under field condition. One plot was designated as control, and the other plot was treated with chicken manure and ferrous sulfate. About 350 kg of the air-dried tailings were put on each plot. The chicken manure and ferrous sulfate were added to the treated plot at a dose of 30 t ha^{-1} and 15 t ha^{-1}, respectively. The chicken manure and tailing were mixed using a stainless steel shovel, and thereafter this mixture was equally divided into 10 subgroups. The ferrous sulfate, which was dissolved in the purified water, was also divided into 10 subgroups. Each subgroup of tailing and ferrous sulfate was mixed. Thereafter, all the homogenized subgroups were further mixed (Fig. S1). After mixing, the tailings were provided with purified water to reach a moisture content of 60% (w/w), and kept for 30 days to be in chemical equilibrium. The tailings were put on a high density polyethylene impermeable membrane to avoid the leaching of water to the underground, and set on a slope. A 2-L borosilicate glass bottle was set on the side of plot with lower topography to collect the leachate. A funnel covered with 1-mm nylon sieve was connected between the tailing and bottle. The borosilicate glass bottle was put in a plastic container to avoid the damage. The tailings mainly produced leachate during the raining episode. For the months when there were very few raining events, the deionized water was provided to the two plots with the same volumes to reach an annual precipitation of 1230 mm. The trial was conducted for five months, and the leachate was collected monthly. The leachate was filtered with 0.45-μm nitrocellulose filter membrane, divided into two subgroups, and acidified with HNO_3 3% and HCl 3% (v/v) respectively, for total As and total Hg analysis.

2.4. Solid sample analysis

The pH values of tailing or chicken manure were determined by a pH meter (Hanna HI3M, Hanna instruments®, USA) in its suspension with solid to deionized water ratio of 1:2.5 (w/w). The organic matter contents were determined using the method of Wang et al. (2014). The total Hg content in the samples was directly measured by pyrolysis of samples at 600 °C–700 °C using Lumex RA 915 + coupled with a Pyro 915 pyrolysis attachment, which has a determination limit of 5 ng g^{-1} (Sholupov et al., 2004). As for As, and other heavy metal determinations, about 0.05 g powdered tailing samples were placed in 15-mL Teflon tubes and digested with a mixture of hydrofluoric acid (HF) (48%) and HNO_3 (65%) and (1:15, v/v) in an oven at 180 °C for 24 h (Brenner et al., 1980). Thereafter, the Teflon tubes without caps were put on a MS7-HP550-S electric hotplate (SCILOGEX Co., CT, America), and the volume of digested solution was reduced to 0.5–1 mL by gradual increasing the temperature of electric hotplate. The residual in the Teflon tubes was reconstituted to 10 mL with 20% HNO_3 (65%). The trace elements concentration were determined using inductively coupled plasma–mass spectroscopy (ICP-MS; Element, Finnigan MAT Co.), and As concentration was measured by atomic fluorescence spectrometry (AFS-920; Beijing Jitian Instrument Co.). Mineralogical composition of the tailing was determined by X-ray diffraction (XRD) with a diffractometer (model: D/Max2200, Japan) using nickel filtered CuKα radiation (λ = 1.54178 Å). Working voltage and current were 40 kV and 30 mA, respectively. The pattern was recorded in the 2θ range of 2–60°, in the mode of step-scanning with 0.04° in step size and a counting time of 5s per step.

2.5. Liquid sample analysis

The pH was determined by a pH meter (Hanna HI3M, Hanna instruments®, USA). As for Total Hg analysis, a 0.5 mL of BrCl (0.5%, v/v) was added to the solution for 24 h to convert all Hg species to Hg(II). Prior to analysis, excessive BrCl was eliminated using 0.2 mL of 20% $NH_2OH \cdot HCl$ (w/w) solution. Mercury in the solution was determined by cold vapor atomic fluorescence spectrometry (CVAFS) (Tekran 2500, Canada) (USEPA, 1999). The As concentration was measured by atomic fluorescence spectrometry (AFS-920; Beijing Jitian Instrument Co., China) (Cai, 2000). The DOC and anions were measured using an total organic carbon analyzer (Elementar High TOC/TN analyzer, Langenselbold, Germany) and ion chromatograph (ICS90, Dionex Co, CA, USA), respectively.

2.6. Quality assurance and quality control

Quality assurance and control analysis for total Hg and As in solid samples were performed using certified reference materials (RTC-CRM023 and GBW07405) and triplicates. The measured total As and Hg contents in the standards RTC-CRM023 and GBW07405 were 386.5 mg kg^{-1} and 76.8 mg kg^{-1}, which were close to the certificated value of 380 mg kg^{-1} and 77.8 mg kg^{-1}, respectively. The variability between the triplicate samples was less than 11% for total As and Hg analysis for solid samples. For liquid samples, recoveries for spiked matrix were 84%–103% for total As (Trace CERT®, 1 mg L^{-1} As in nitric acid) analysis, and 89%–107% for total Hg (ICP-MS Standard, 10 mg L^{-1} Hg in nitric acid) analysis, and the variability between the triplicate samples was less than 8% for all the analysis. Statistical analysis was performed with SPSS 17.0 software, and the figures were created using Origin 8.0.

3. Results and discussion

The studied tailing was alkaline (pH = 9.87), and contained low organic matter content (9 mg g^{-1}) (Table S1). The main mineral compositions included SiO_2 an $CaMg(CO_3)_2$. The parental rock of Danzhai mine was dolomite ($CaMg(CO_3)_2$), and the processing of Au ores could have lead to the mixture of dolomite into the tailing. The appearance of minor organic matter in the tailing might be attributed to the mixture of soil particles nearby the tailing piles by wind. The high pH of the tailing may be favorable for mobilization of As, as indicated by Sun et al. (2014).

The average total As, Hg, lead, zinc, chromium, copper, and manganese content was 523, 21, 304, 221, 29, 15, and 610 mg kg^{-1} (Table S1). No limitations for toxic elements contents in solid wastes were set by the Chinese government. Mercury and As were primary environmental concerns in the tailing in Danzhai, as demonstrated by previous laboratory investigations (Sun et al., 2014; Table S2). For instance, Hg (12 ng mL^{-1}) and As concentrations (9,929 ng mL^{-1}) in the leachate exceeded the maximum allowable Hg and As concentrations in the environmental quality for surface water set by the Chinese government (GB 3838-2002), when the 0.1 mol L^{-1} CH_3COONH_4, and the mixture of NaH_2PO_4 (0.1 mol L^{-1}): Na_2HPO_4 (0.1 mol L^{-1}) (3 : 2 v/v) were used as leaching agents (Sun et al., 2014).

3.1. Impact of sole application of chicken manure on Hg and As leaching from the tailing-greenhouse study

The average concentration of Hg in the leachate of the non-treated tailing was 11,000, 10,000 and 5,800 ng L^{-1} after leaching for 30, 60 and 90 days, respectively (Fig. 2). The noticeable mobilization of Hg in the non-treated tailing was likely due to its high proportions of Hg^{2+} (2–5%) and Hg^0 (15–35%), as indicated by Li et al. (2013) and Yin et al. (2016). As for chicken manure

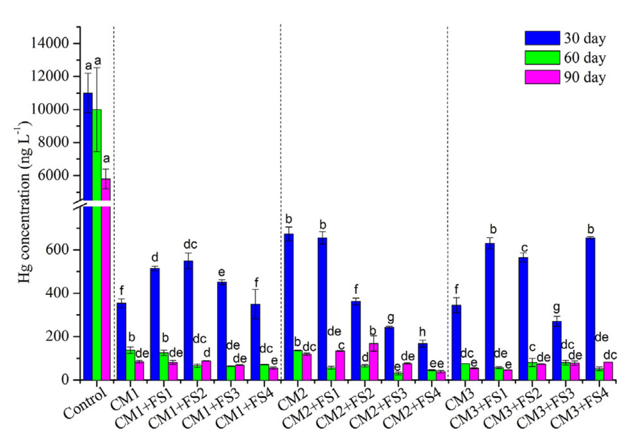

Fig. 2. The concentration of Hg in the leachate in the non-treated tailing, and the tailing treated with 30 tons ha^{-1} (CM1) to 90 tons ha^{-1} (CM3) or their combinations with 1.5 tons ha^{-1} (FS1) to 15 tons ha^{-1} (FS3). The different small letters above each column means the difference in Hg concentrations between sixteen treatments at each sampling time is significant at $P < 0.05$. CM, chicken manure; FS, ferrous sulfate.

treatment, the average concentrations of Hg in the leachate in the 30 t ha^{-1}, 60 t ha^{-1}, and 90 t ha^{-1} treatments were 354 ± 20 ng L^{-1}, 673 ± 32 ng L^{-1}, and 345 ± 35 ng L^{-1}, and they were further deceased to 84 ± 6.1 ng L^{-1}, 118 ± 5.2 ng L^{-1}, and 54 ± 3.0 ng L^{-1} after three months of leaching. The application of chicken manure at three doses led to a reduction of total Hg (94%–99%) in the leachate as compared to the non-treated tailing (Table 2).

The average concentration of As in the leachate of the non-treated tailing was 140, 135 and 120 ng mL^{-1} after leaching for 30, 60 and 90 days (Fig. 3). Application of chicken manure at three doses decreased As concentration in the leachate compared to the control, and the extent of decrease was greater in higher dose treatment than the lower one (Fig. 3). Also, the concentration of As decreased noticeably in the leachate with the extension of time. For instance, the concentrations of As in the leachate in 30 t ha^{-1} CM, 60 t ha^{-1} CM, and 90 t ha^{-1} CM treatments were 100 ± 3.8 ng mL^{-1}, 68 ± 2.5 ng mL^{-1}, and 54 ± 4.7 ng mL^{-1}, after one month of leaching, and they decreased to 62 ± 5.2 ng mL^{-1}, 38 ± 3.3 ng mL^{-1}, and 18 ± 0.9 ng mL^{-1} (About 49%–85% of total As were removed; Table 3), after three months of leaching (Fig. 3).

3.1.1. The possible mechanisms of Hg immobilization by chicken manure-greenhouse study

Chicken manure contained functional groups such as hydroxyl, carboxyl, and amine (Merlin et al., 2014), which might inhibit the mobilization of toxic elements in wastes through adsorption, complexation, and precipitation (Li et al., 2018; Ngah and Hanafiah, 2008). The chicken manure used in this study was enriched with sulfur (about 0.1%), which might play a key role in affecting Hg mobilization in the tailing. The relationships between Hg and pH/sulfate/DOC of the leachate might provide the evidences of Hg/As immobilization by the chicken manure (Fig. 4, -A, -B, -C). There was a negatively linear correlation between the sulfate and Hg in the leachate (Fig. 4, -B), meaning the Hg decreased with the increase of sulfate. Reduction of sulfate to sulfide by sulfate reducing bacteria (SRB) was the predominant reaction leading to the reduction of sulfate in the environment (Widdel and Bak, 1992). The presence of more sulfates might enhance SRB activities, producing more sulfides to bind with Hg^{2+} to form hardly soluble Hg sulfides (e.g., HgS) (Benoit et al., 1999).

The pH noticeably affected Hg speciation and its mobilization in the environment (Haitzer et al., 2003). Application of chicken manure at three doses decreased pH significantly, in particular after the incubation for three months when the pH was decreased by 2 units compared to the control (Fig. S2). This acidification might be related to microbe activities after chicken manure amendment to the tailing. For instance, the microbe (e.g., acetogenic bacteria) could use simple organic compounds decomposed from chicken manure as carbon food sources to produce acids (e.g., acetate) to acidify their growth medium (Inglett et al., 2005). The pH of the leachate in the control remained relatively stable during the experiment (Fig. S2), perhaps because the lack of carbon sources to enhance the microorganism activities (Moynahan et al., 2002). Generally, the low pH favors the mobilization of Hg since the metal associated with hydroxides, carbonates, and phosphate precipitates could be dissolved under acidic conditions (Draszawka-Bołzan, 2017), as well as the number of negative sites for cation adsorption decreased with the decrease of pH (Rieuwerts et al., 1998). In contrast to this phenomenon, there was a positive correlation between pH and Hg concentration in the leachate (Fig. 4, -A), which indicates that effect of pH decrease on Hg mobilization was minor as compared to its immobilization by chicken manure (e.g., Hg was likely immobilized by sulfide sulfur produced by the decomposition

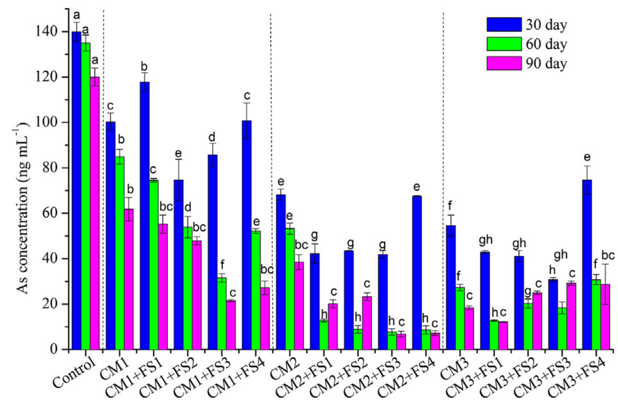

Fig. 3. The concentration of As in the leachate in the non-treated tailing, and the tailing treated with 30 tons ha^{-1} (CM1) to 90 tons ha^{-1} (CM3) or their combinations with 1.5 tons ha^{-1} (FS1) to 15 tons ha^{-1} (FS3). The different small letters above each column means the difference in As concentrations between sixteen treatments at each sampling time is significant at P < 0.05. CM, chicken manure; FS, ferrous sulfate.

Table 2
Impact of chicken manure (CM) and/or ferrous sulfate (FS) applications on the changes (%) of the Hg concentration in the leachate in the treated tailing as compared to the control.

Treatments	30 days	60 days	90 days
30 t ha^{-1} CM	−97	−99	−99
30 t ha^{-1} CM+1.5 t ha^{-1} FS	−95	−99	−99
30 t ha^{-1} CM+3 t ha^{-1} FS	−95	−99	−98
30 t ha^{-1} CM+9 t ha^{-1} FS	−96	−99	−99
30 t ha^{-1} CM+15 t ha^{-1} FS	−97	−99	−99
60 t ha^{-1} CM	−94	−99	−98
60 t ha^{-1} CM+1.5 t ha^{-1} FS	−94	−99	−98
60 t ha^{-1} CM+3 t ha^{-1} FS	−97	−99	−97
60 t ha^{-1} CM+9 t ha^{-1} FS	−98	−99.5	−99
60 t ha^{-1} CM+15 t ha^{-1} FS	−98	−99.6	−99
90 t ha^{-1} CM	−97	−99	−99
90 t ha^{-1} CM+1.5 t ha^{-1} FS	−94	−99	−99
90 t ha^{-1} CM+3 t ha^{-1} FS	−95	−99	−99
90 t ha^{-1} CM+9 t ha^{-1} FS	−98	−99	−99
90 t ha^{-1} CM+15 t ha^{-1} FS	−94	−99	−99

Increasing/decreasing percentage = (Treatment-control)/control*100% (data was calculated from Fig. 2).
(+): increasing; (−): decreasing.

Table 3
Impact of chicken manure (CM) and/or ferrous sulfate (FS) applications on the changes (%) of the As concentration in the leachate in the treated tailing as compared to the control.

Treatments	30 days	60 days	90 days
30 t ha^{-1} CM	−28	−37	−49
30 t ha^{-1} CM+1.5 t ha^{-1} FS	−16	−45	−54
30 t ha^{-1} CM+3 t ha^{-1} FS	−47	−60	−60
30 t ha^{-1} CM+9 t ha^{-1} FS	−39	−77	−82
30 t ha^{-1} CM+15 t ha^{-1} FS	−28	−61	−77
60 t ha^{-1} CM	−51	−61	−68
60 t ha^{-1} CM+1.5 t ha^{-1} FS	−70	−90	−83
60 t ha^{-1} CM+3 t ha^{-1} FS	−69	−93	−81
60 t ha^{-1} CM+9 t ha^{-1} FS	−70	−94	−94
60 t ha^{-1} CM+15 t ha^{-1} FS	−52	−94	−94
90 t ha^{-1} CM	−61	−80	−85
90 t ha^{-1} CM+1.5 t ha^{-1} FS	−69	−91	−90
90 t ha^{-1} CM+3 t ha^{-1} FS	−71	−85	−79
90 t ha^{-1} CM+9 t ha^{-1} FS	−78	−86	−76
90 t ha^{-1} CM+15 t ha^{-1} FS	−47	−77	−76

Increasing/decreasing percentage = (Treatment-control)/control*100% (data was calculated from Fig. 3).
(+): increasing; (−): decreasing.

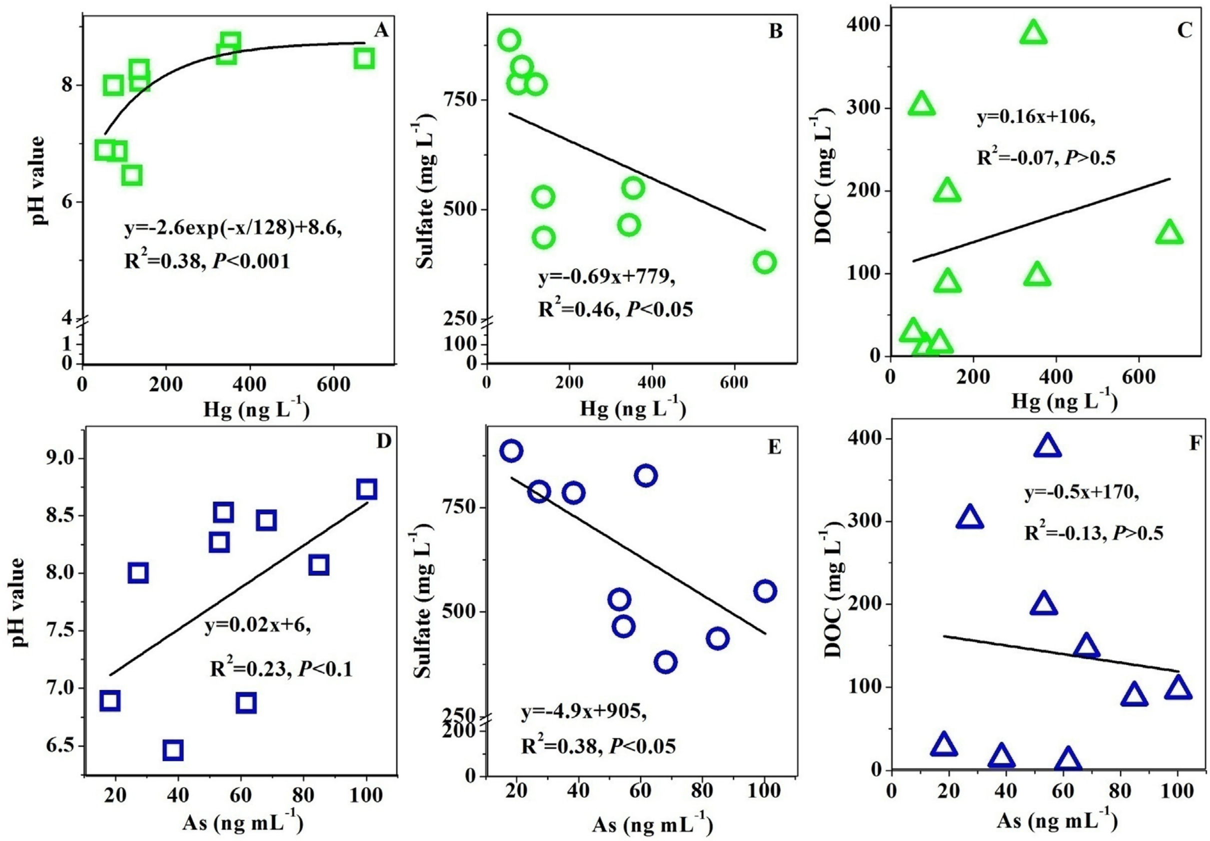

Fig. 4. The relationships between concentration of Hg/As and the levels of different environmental parameters in the leachate in the tailing (control and chicken manure-treated tailing). (A): Hg concentration *vs* pH; (B) Hg concentration *vs* sulfate concentration; (C) Hg concentration *vs* dissolved organic carbon (DOC) concentration; (D): As concentration *vs* pH values; (E) As concentration *vs* sulfate concentration; (F) As concentration *vs* DOC concentration.

of chicken manure). Dissolved organic carbon (DOC) is well-known for its high binding affinity to Hg, and considered as a key factor controlling Hg mobilization in the environment (Ravichandran, 2004). It can promote the mobilization of Hg by breaking Hg–S bond of HgS (Waples et al., 2005), inhibiting the nucleation of meta-cinnabar (Ravichandran et al., 1999), and forming soluble Hg-DOC complexes (Chen et al., 2017). In this study, there was no clear relationship between DOC and Hg in the leachate (Fig. 4, -C), meaning a subordinate role of DOC derived from chicken manure in affecting Hg mobilization in the tailing.

3.1.2. The possible mechanisms of As immobilization by chicken manure

There was a tendency of the increase of concentration of As with the increase of pH in the leachate (Fig. 4, -D), indicating a high leachability of As in the alkaline environment. The result is consistent with a previous observation that the promotion of As mobility by the desorption of negatively charged As ions (e.g., arsenate) as a result of the reduction of positive surface charge sites in mineral or soil particles under alkaline conditions (Masscheleyn et al., 1991). Application of chicken manure decreased pH noticeably, which could partially lead to the decrease of As concentrations in the leachate. Apart from pH, there was also a significantly negative correlation between the As and sulfate in the leachate (Fig. 4, -E), which indicates that the decrease of sulfate was companied with the diminishing of As. This observation might be attributed to the formation of poorly soluble arsenic sulfide minerals (e.g., As_2S_3, AsS, etc.) in the tailing by the reduction of both sulfate and arsenate. It is reported that the presence of sulfate and arsenate, as well as approximate amount of electron donors could lead to the microbial reduction of both sulfate and arsenate to form arsenic sulfides (Rodriguez-Freire et al., 2016). A similar reduction process might have happened in the tailing spiked with chicken manure. Arsenic mainly might present as As(V) in the tailing, as indicated by previous studies (e.g., Paktunc et al., 2004; Ono et al., 2016). The small organic moleculars produced by the decomposition of chicken manure might serve as electron donors to be used for sulfate and arsenate reduction by microorganisms. Therefore, it is possible for the formation of arsenic sulfide minerals in the tailing treated with chicken manure.

Dissolved organic carbon (DOC) could bind with As to affect its mobilization in the environment (Chen et al., 2016). However, the interaction between DOC and As was affected greatly by the As speciation, pH, type of humic acid, and other cations (Liu and Cai, 2010). There was no noticeable relationship between As and DOC (Fig. 4, -F and Fig. S3) in the leachate, likely due to the low binding affinity of DOC to As under alkaline condition ($pH > 7$) (Buschmann et al., 2006).

3.2. Impact of co-application of chicken manure and ferrous sulfate on the leaching of Hg and As from the tailing

Ferrous sulfate was often used to lower soil pH, and recommended as an amendment to improve the biogeochemical environment of soils and tailings (Zou et al., 2018). The application of the mixture of chicken manure and ferrous sulfate on the mobilization of Hg and As in the tailing was investigated.

3.2.1. Hg and As

The concentrations of Hg and As showed large variations in the leachate of the tailings spiked with 30 tons ha^{-1} to 90 tons ha^{-1} chicken manure and 1.5 tons ha^{-1} to 15 tons ha^{-1} ferrous sulfate as compared to the sole chicken manure treatments during the first month of leaching (Figs. 2 and 3). These variations should be caused by the adsorption/desorption of Hg and As by tailing and amendments (chicken manure and ferrous sulfate) when they were over-saturated (e.g., Roussiez et al., 2013). The concentrations of Hg decreased about 3.3%–53%, and 35%–77% in the 30 tons ha^{-1} and 60 tons ha^{-1} chicken manure coupled with different doses of ferrous sulfate treatments (except for 30 tons ha^{-1} CM+3 tons ha^{-1} Fe in the third month, 60 tons ha^{-1} CM+3 tons ha^{-1} Fe in the third month), respectively, as compared to the sole chicken manure treatment in the second and third month of leaching (Table S3 and Fig. 2). The concentration of As decreased about 10%–65% and 38%–85%, in the 30 tons ha^{-1} and 60 tons ha^{-1} chicken manure coupled with different doses of ferrous sulfate treatments, respectively, as compared to the sole chicken manure treatments in the second and third month of leaching (Table S4 and Fig. 3). The combination of ferrous sulfate with higher treatment dose of chicken manure (90 tons ha^{-1}) seems to be less efficient in decreasing Hg and As mobilization as compared to the other treatments. It appears that the amendment of both chicken manure and ferrous sulfate to the tailing added an additional immobilization effect to both Hg and As (except for 90 tons ha^{-1} treatments).

3.2.2. The possible mechanisms of Hg and As immobilization by chicken manure and ferrous sulfate

As we discussed in section 3.1, the predominant mechanisms of immobilization of Hg and As by chicken manure might be linked to the formation of Hg and As sulfide complexes, as indicated by their close relationships with sulfate (Fig. 4, -B, -E). There were no clear relationships between As/Hg and sulfate/DOC/pH (Fig. 5, -A, -B, -C, -D, -E, except for As *vs* pH) in the leachate in the tailing treated with both ferrous sulfate and chicken manure, suggesting that the immobilization of Hg and As by ferrous sulfate and chicken manure might through mechanisms being different from the sole chicken manure. However, if we took a close look at the data in Fig. 5, there was a very small variation in Hg concentrations in the leachate collected at 60 and 90 days, indicating a minor influence of pH on Hg mobilization. The result is in line with a prior study which reported that addition of Fe(III) into Hg solid wastes collected from Sulfur Bank Mercury Mine in USA led to an reduction of Hg concentration in the leachate, and this reduction was relatively independent of pH (Randall et al., 2004). In this study, Hg might be immobilized by the amendments (ferrous sulfate and chicken manure) through the iron hydr(oxides) adsorption (e.g., α-FeOOH). A previously mechanic investigation on using goethite (α-FeOOH) to remove Hg^{2+} from solution showed an stable adsorption of Hg^{2+} by goethite under alkaline conditions (Barrow and Cox, 1992). The ferrous sulfate in alkaline solution could fast form ferrous hydroxide, which could be further converted to α-FeOOH (O'Connor et al., 1992). In this study, the tailing's pH was 9.8 and was amended with ferrous sulfate. It may be favorable for the formation of Fe hydr(oxides) for Hg adsorption. Additionally, the presence of chicken manure might further enhance the Hg removal by Fe hydr(oxides) because the organic materials had been reported to be able to enhance Hg adsorption by metal oxides under varying pH conditions (Xu and Allard, 1991). Bäckström et al. (2003) documented a positive effect of organic matter (e.g., 20 mg L^{-1} fulvic acid) on Hg adsorption by Fe oxides.

The concentration of As was correlated negatively with pH in the leachate in the chicken manure and ferrous sulfate treatment, which was opposite of the result from sole chicken manure treatment (Fig. 5, -D). Ferric (oxyhydr)oxides are demonstrated to be efficient adsorbents for As in the environment (Dixit and Hering, 2003). Arsenic adsorption by ferrous sulfate should play a significant role in As immobilization in this study. The increase of pH could lead to the decrease of As adsorption by iron oxides (Dixit and

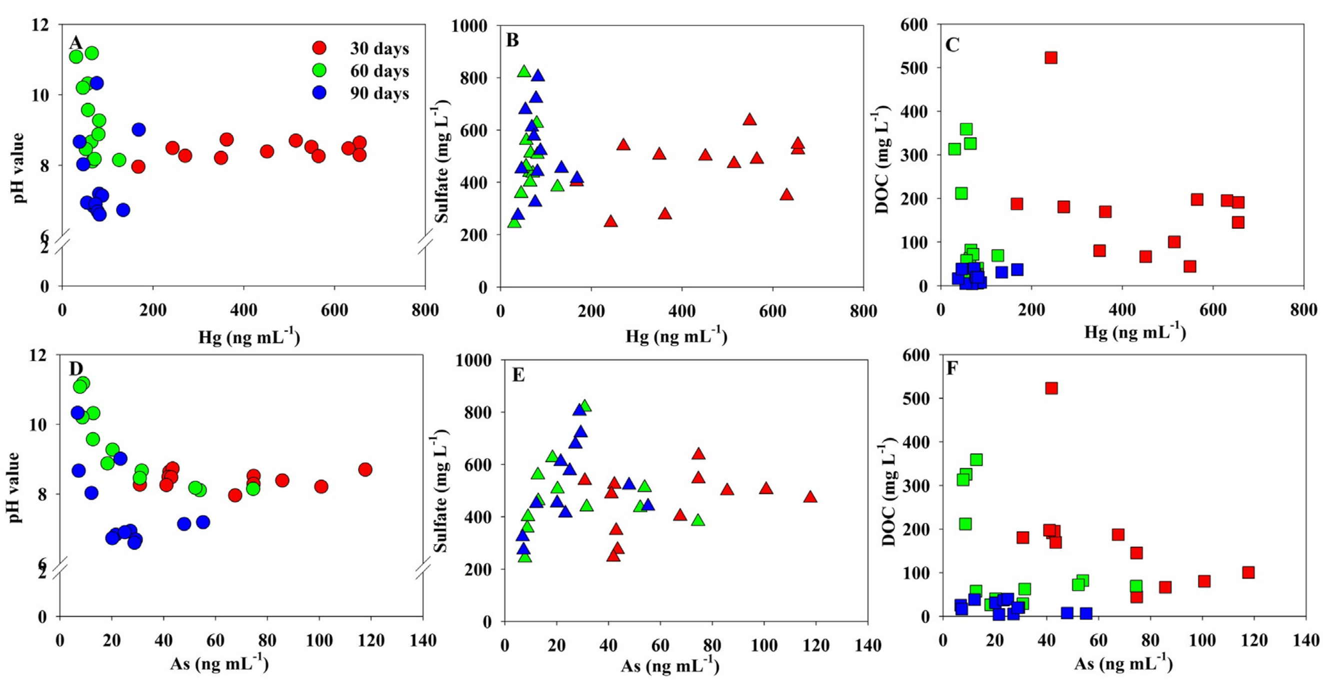

Fig. 5. The relationships between concentration of Hg/As and the levels of different environmental parameters in the leachate in the tailing treated with 30 tons ha^{-1} to 90 tons ha^{-1} chicken manure and 1.5 tons ha^{-1} to 15 tons ha^{-1} ferrous sulfate. (A): Hg concentration *vs* pH values; (B) Hg concentration *vs* sulfate concentration; (C) Hg concentration *vs* DOC concentration; (D): As concentration *vs* pH values; (E) As concentration *vs* sulfate concentration; (F) As concentration *vs* DOC concentration.

Hering, 2003). In this study, the concentration of As in the leachate in the tailing decreased with the increase of pH (Fig. 5, -D), which indirectly support the hypothesis of As adsorption by iron oxides. The poor correlations between As and sulfate/DOC suggests a minor role of these factors in affecting As mobilization in the tailing (Fig. 5, -E, -F).

3.3. Field trial

The performance of the amendments (30 tons ha^{-1} chicken manure and 15 tons ha^{-1} ferrous sulfate) on the removal of Hg and As from the leachate in the tailing under field condition was investigated. The 60 tons ha^{-1} chicken manure and 15 tons ha^{-1} ferrous sulfate showed a relatively higher removal rate than 30 tons ha^{-1} chicken manure and 15 tons ha^{-1} ferrous sulfate, but the former's capital cost should be higher than the latter. Thus the 30 tons ha^{-1} chicken manure and 15 tons ha^{-1} ferrous sulfate is recommended. The main aim was to investigate whether the application of the selected amendments to tailing could reduce the concentrations of Hg and As in the leachate from the tailing. The average monthly precipitation at Danzhai County was about 92 mm during the period of our trial, which only accounted for about 7% of its annual precipitation (1230 mm). This means that we were not able to collect enough amount of leachate with this limited precipitation. As an alternative, we provided deionized water to each treatment to reach an annual precipitation of 1230 mm, by which we were able to investigate the effect of high flooding on the performance of amendments. As shown in Fig. 6 and Table S5, the concentrations of Hg and As in the leachate in the non-treated tailings were 2,970 ng L^{-1} to 5,130 ng L^{-1}, and 330 ng mL^{-1} to 450 ng mL^{-1}, respectively, during the trial, which were at the same magnitudes with the results from the greenhouse study, and exceeded their maximum allowable concentrations in surface water set by the Chinese government. Application of the amendments removed about 37%–73% of total Hg, and 75%–82% of total As from the leachate, respectively. These results were close to that from the greenhouse study, further demonstrating that the selected amendments were efficient in reducing the mobility of Hg and As in the tailing. The mechanism of Hg and As removal by the chicken manure and ferrous sulfate should be the one that we proposed in previous section (section 3.2). The major anions and pH were determined to understand the impact of the chicken manure and ferrous sulfate amendments on the change of chemical compositions of the leachate. As shown in Table S5, sulfate concentration in the treated leachate was generally higher than the control because of the input of sulfate; DOC concentrations in the treated leachate were similar to the control, suggesting that chicken manure and ferrous sulfate amendments did not significantly change this biogeochemical parameter under field conditions. It should be noted that chicken manure might contain trace antibiotics (Yang et al., 2014). More studies should be conducted to explore the risk of antibiotics in chicken manure after its amendment to soil, and to monitor the emission of ammonia and the dispersing of particles from chicken manure-amended tailing. This knowledge could be helpful for further improving the performance of the amendments in tailings.

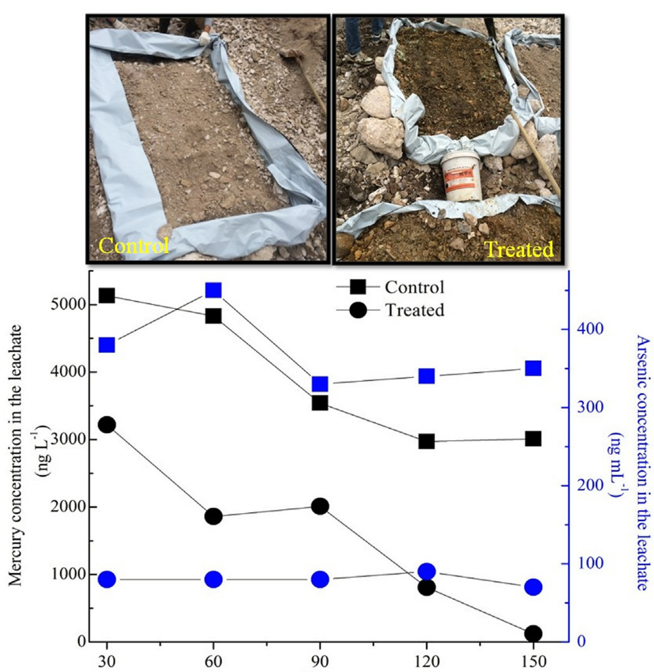

Fig. 6. The concentration of Hg/As in the leachate in the non-treated tailing, and the tailing treated with 30 tons ha^{-1} chicken manure (CM) and 15 tons ha^{-1} ferrous sulfate (FS) over the five months in the field. The two photos above the figure were taken from the field prior to collecting the leachate. Non-treated control (left) and the treated tailing (right). The solid square symbol indicates non-treated control; the solid circle symbol indicates the treated tailing. The blue symbols indicate As and the black symbols indicate Hg. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

4. Conclusions

The results showed that application of both 30 tons ha^{-1} chicken manure and 15 tons ha^{-1} ferrous sulfate as sustainable amendments to the tailing immobilized Hg and As. Both the results from greenhouse and field studies showed that about 37%–73% of total Hg, and 75%–82% of total As were removed from the leachate in the tailing, and the pH was decreased from 9.7 to slightly alkaline. Therefore, it may be an good option to use chicken manure and ferrous sulfate to treat tailings in Karst regions of China. Also, the results suggest that iron and organic matter play an crucial role in both Hg and As immobilization. Future work may focus on the interaction of organic components in chicken manure and iron and Hg/As in tailings to elucidate how these factors control Hg and As mobilization, as well as long-term performance of the amendments under field conditions. Also, the monitoring of antibiotics, ammonia emission, and particles dispersion is necessary.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2019.118171.

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