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# Effects of aerobic and anaerobic biological processes on leaching of heavy metals from soil amended with sewage sludge compost

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ABSTRACT

The risk from leaching of heavy metals is a major factor hindering land application of sewage sludge compost (SSC). Understanding the change in heavy metal leaching resulting from soil biological processes provides important information for assessing long-term behavior of heavy metals in the compost amended soil. In this paper, 180 days aerobic incubation and 240 days anaerobic incubation were conducted to investigate the effects of the aerobic and anaerobic biological processes on heavy metal leaching from soil amended with SSC, combined with chemical speciation modeling. Results showed that leaching concentrations of heavy metals at natural pH were similar before and after biological process. However, the major processes controlling heavy metals were influenced by the decrease of DOC with organic matter mineralization during biological processes. Mineralization of organic matter lowered the contribution of DOC-complexation to Ni and Zn leaching. Besides, the reducing condition produced by biological processes, particularly by the anaerobic biological process, resulted in the loss of sorption sites for As on Fe hydroxide, which increased the potential risk of As release at alkaline pH.



## 1. Introduction

Treatment and disposal of sewage sludge are important issues world-wide. Nowadays, land application, incineration, and landfilling are three main ways of sewage sludge disposal (Fytili and Zabaniotou, 2008). Since land application of sewage sludge can recycle the nutrient elements (such as N and P) as well as increase soil carbon sequestration, sewage sludge has been widely applied to soil in many countries after composting (Thangarajan et al., 2013). However, the heavy metals in sewage sludge compost (SSC) is a key factor limiting the land application of SSC. When SSC is applied to soil frequently, heavy metals may release to soil water, thereby potentially posing eco-toxicity for plants, animals, and human beings (Singh and Agrawal, 2008, 2010). To evaluate the environmental risk of heavy metals following land application of sewage sludge compost, the leaching behavior of heavy metals in the soil amended by SSC should be investigated.

Leaching of heavy metals is significantly influenced by various natural processes in the soil-water-plant system, such as microbial activities, weathering, drainage, and production of exudates by plant roots. All these processes are interactive with the potential to change properties of soil, thereby influencing the leaching behavior of heavy metals. When compost is applied to soil, it can increase the content of soil organic carbon and microbial activities (Aranda et al., 2015). The enhanced microbial activities resulting from SSC land apllication are important factors influencing leaching of heavy metals.

Both aerobic and anaerobic biological processes are common soil biological processes based on the content of water and dissolved oxygen in soils. The leaching behavior of heavy metals are likely to be different as a result of aerobic and anaerobic biological processes, due to the differences in microbial activities as well as soil conditions (such as redox). To date, a variety of studies have focused on the effects of aerobic biological processes on the leachability and speciation of heavy metals in the soil amended by organic amendments. Doelsch et al. (2010) determined the organic matter mineralization of the soil amended by pig slurry and composted green waste during a 28 day aerobic incubation and assessed its impact on the leachability of heavy metals. Rajaie et al. (2006) explored the change of Cd speciation in the compost amended soil during a 16 weeks aerobic incubation. Clemente et al. (2006) and Pardo et al. (2011) investigated how the speciation of Zn and Pb changed when compost and manure were applied to soil with 56 days aerobic incubation. Although the effects of aerobic biological processes have been well studied, the influence of

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anaerobic biological process on heavy metals' leaching behavior has been rarely investigated. The anaerobic process often dominates biological and chemical features of flooded and poorly drained soil, including when flooding is used as part of rice production. Besides, in well-drained soil, soil aggregation and water retention can lead to localized anaerobic conditions, but is restricted to small zones and to limited periods (Tiedje et al., 1984). Therefore, the impact of anaerobic biological processes on heavy metals leaching behavior should be a concern.

In this study, both aerobic and anaerobic incubations were conducted to investigate the effects of biological processes on leaching behavior of heavy metals in a soil amended with SSC. The pH-dependent leaching of heavy metals was determined to describe the leaching characteristics of heavy metals. In addition, the results of pH-dependent leaching tests were incorporated into geochemical modeling to illustrate chemical speciation of heavy metals across applicable pH domains and relevant geochemical conditions.

## 2. Materials and methods

### 2.1. Materials

The reference soil, type of silt loam, was sampled in the 0–25 cm horizon from a farmland area in Suzhou, Jiangsu, China (120°37′E, 31°19′N). The farmland has been used for wheat and rice cultivation for over 10 years. This region lies in the lower Yangtze River alluvial plain. The Quaternary strata are widely distributed and the thickness of the unconsolidated deposits increases generally from southwest to northeast. The SSC was sampled from composted municipal sewage sludge after a 30-day composting process with green waste.

The simulation of the compost amended soil was prepared by adding SSC to soil at 4% dry weight ratio, which corresponds to the compost application rate (10 cm tillage) of 48 ton/ha which is a common application rate currently adopted. The physicochemical properties of SSC, the reference soil, and amended soil were shown in Table 1 as previously reported by Fang et al. (2016).

### 2.2. Aerobic and anaerobic incubation

The aerobic and anaerobic incubation were conducted to simulate the aerobic and anaerobic biological processes in the system considering only soil and water. The aerobic and anaerobic incubations of the reference soil and the compost amended soil were carried out in triplicate in 300 mL glass jars. The water contents of aerobic and anaerobic treatments were equal to 37.5% and 60.0%, respectively, corresponding to the field water capacity of the soil under poorly drained and well-drained conditions. All the jars were incubated in the dark at 28 °C. For the aerobic incubation, the headspace of each jar was filled with air and headspace gas was sampled and analyzed every 2 days initially and the at 7 day intervals by GC (Agilent 7820A) to determine the emission of $CO_2$. After each sampling, the headspace was evacuated with a vacuum pump (10–15 min) and filled with air in 1 min. This was repeated four times to remove most of the background gas. For the anaerobic incubation, the headspaces was filled with $N_2$ to maintain anaerobic conditions and a similar gas sampling procedure was used to determine the emission of $CH_4$. According to the mineralization rate of organic matter, the aerobic and anaerobic incubation were conducted for 180 and 240 days, respectively.

### 2.3. Physicochemical analysis of the compost amended soil

The physicochemical properties of samples comprised measurements of the pH, electrical conductivity (EC), dissolved organic carbon (DOC), total organic carbon (TOC), humic substances (humic acids (HAs) and fulvic acids (FAs)), Al (hydr)oxides and Fe (hydr) oxides. Sample pH, EC, and DOC were determined through extracting samples with deionized water at a liquid to solid ratio (L/S) of 10 L/kg-dry. 20 g samples and 200 mL deionized water were added to the bottle and tumbled for 48 h at 28 ± 2 rpm under room temperature. The bottles were then centrifuged at 10,000 rpm for 10 min and the pH and EC of the supernatant were tested. Later, the supernatant was filtered through a 0.45 μm filter membrane and the filtrate was collected for DOC measurement (TOC-cvph: Shimadzu, Kyoto, Japan). TOC in the solid matrix was determined by the high-temperature combustion method with a Shimadzu solid sample module coupled to a TOC analyzer (TOC-cvph: Shimadzu, Kyoto, Japan). Two separate furnaces were set at 900 °C for TC and 200 °C for TIC respectively. The content of TOC was calculated by subtracting TIC from TC.

The contents of FAs and HAs were obtained using the method introduced by the International Humic Substance Society (IHSS) (ISO/TC 190/SC 7/WG 6 N 193, 2008). The extraction containing 0.1 M $Na_4P_2O_7$ and 0.1 M NaOH was used to extract FAs and HAs in the solid phase. 1 g sample, along with 25 mL extraction solution were mixed and shaked in a 50 mL centrifuge tube at 90 °C for 1 h, followed by a centrifugation procedure at 5000 rpm for 5 min to collect the supernatant. This extraction procedure was repeated until the supernatant became colorless, indicating that there was no FAs and HAs remained in the solid phase. The extract of each procedure was combined to determine the total contents of FAs and HAs. The FAs and HAs in the extracts were separated according to their solubility difference at low pH, where HAs were precipitated and FAs were soluble. To separate FAs and HAs, 20 mL extract was added to the 50 mL centrifuge tube and the pH of the extract was adjusted to pH 1 with 6 M HCl. After overnight standing, the tube was centrifuged at 10,000 rpm at 15 °C to separate the precipitated HAs from the dissolved FAs. The FAs in the supernatant was purified through a DAX-8 resin absorption procedure and the precipitated HAs was then re-dissolved by 0.1 M NaOH. Finally, HAs and FAs contents in the liquid phases were measured by TOC analyzer (TOC-cvph: Shimadzu, Kyoto, Japan).

Al (hydr)oxides and Fe (hydr)oxides were extracted by ascorbic acid and ammonium oxalate-oxalic acid, respectively (ISO/TC 190/SC 7/WG 6 N 189, 2008; ISO/TC 190/SC 7/WG 6 N 191, 2008). The extraction solution for Al (hydr)oxides was prepared by adding 9.68 g of di-ammonium oxalate monohydrate, 6.51 g of oxalic acid dehydrate, and 600 mL deionized water to 1 L PE bottle. During the extracting procedure, 3 g sample and 300 mL extraction solution were added to the 500 mL PE bottle, which was then shaked in the dark at room temperature for 4 h. After that, the bottle was centrifuged at 5000 rpm for 30 min and the supernatant was filtrated through 0.45 μm filter to collect the extract of Al. To prepare

**Table 1**
Physicochemical properties of sewage sludge compost, the reference soil, and the amended soil.

| Samples | EC (μs/cm) | DOC (mg/L) | TOC (g C $kg^{-1}$) | Humic substances (g C $kg^{-1}$) | Fe (hydr)oxides (g Fe/kg) | Al (hydr)oxides (g Al/kg) | pH | Clay (%) |
|---|---|---|---|---|---|---|---|---|
| SSC | 749,000 | 243 | 139 | 87.1 | 31.8 | 4.2 | 6.75 | 1.5 |
| Reference soil | 773 | 18 | 18.4 | 12.9 | 2.7 | 1.9 | 7.35 | 13.0 |
| Amended soil | 1106 | 42 | 24.6 | 16.6 | 2.9 | 1.8 | 6.91 | 13.0 |

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