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Release of heavy metals during long-term land application of sewage sludge compost: Percolation leaching tests with repeated additions of compost

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HIGHLIGHTS

- Leaching of heavy metals during repetitive land application of SSC was evaluated.
- Percolation and pH-dependent leaching test were used to understand the leaching behavior.
- Repeated additions of SSC favored the formation of reducing condition.
- Establishment of reducing conditions can enhance the release of accumulated As.

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ABSTRACT

Leaching assessment procedures have been used to determine the leachability of heavy metals as input for evaluating the risk from sewage sludge compost land application. However, relatively little attention has been paid to understanding leaching from soils with repeated application of sewage sludge compost with elevated levels of heavy metals. In this paper, leaching assessment is extended to evaluate the potential leaching of heavy metals during repetitive application of composted sewage sludge to soils. Four cycling of compost additions and percolation leaching were conducted to investigate how leaching behavior of heavy metals changed with repeated additions of compost. Results showed that repetitive additions of compost to soil significantly increased the content of organic matter, which favored the formation of reducing condition due to improved microbial activities and oxygen consumption. Establishment of reducing conditions can enhance the leaching concentrations of As by approximately 1 order of magnitude, especially for the soil rich in organic matter. For Cd, Cr, Cu, and Pb, repeated additions of compost will cause accumulation in total contents but not enhancement in leaching concentrations. The infiltration following compost additions will leach out the mobile fraction and the residual fraction might not release in the next cycling of compost addition and infiltration. The cumulative release of Cd, Cr, Cu, and Pb accounted for less than 5% of the total contents during four times of compost applications.

1. Introduction

Since sewage sludge compost (SSC) contains high contents of plant nutrient elements and organic carbon, it can be used as soil amendment to improve soil characteristics. However, contaminants in SSC, such as heavy metals and toxic organic pollutants, are major limitations to land application of SSC (Clarke and Smith, 2011; Knoth et al., 2007; Wang et al., 2007). Related research revealed that sewage sludge in China had relatively higher contents of heavy metals when comparing with developed countries, such as EU and US (Dai et al., 2007; Hua et al., 2008; Jones et al., 2014). Therefore, the risk of heavy metals accumulation and leaching needs to be considered for repeated land application of SSC in China

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(Fang et al., 2016).

Leaching of heavy metals is a primary pathway for potential impact to human health and the environment. Prior studies and regulatory assessment have most frequently focused on total elemental content or single step leaching procedures as the basis for risk assessment (Jin et al., 2014; Qi et al., 2011). Robust leaching assessment procedures that facilitate more accurate estimates of heavy metals leaching have seldom been used to determine the leachability of heavy metals as input for evaluating the risk of SSC land application. Among various leaching assessment procedures, the percolation test can provide insight into leaching of heavy metals considering convection and diffusion, which are relevant to the application scenario. However, recently the percolation test is being used more often to evaluate the environment impact of various waste regarding to leaching of contaminants (Kalbe et al., 2014; Pantini et al., 2015; Tsiridis et al., 2015). Pantini et al. (2015) analyzed the release of contaminants from Mechanical Biological Treatment (MBT) wastes using batch and up-flow column tests. Tsiridis et al. (2015) assessed the interaction between soil and coal fly ash with regard to the final land disposal of fly ashes through column percolation tests. Kalbe et al. (2014) investigated the leaching behavior of contaminated soil and compared the results from column studies with lysimeter studies. Percolation test results also can be evaluated in conjunction with pH dependent leaching tests to provide insights into the changes in mechanism controlling leaching of heavy metals (Kosson et al., 2014a, 2014b).

Even though the percolation leaching test was used to determine the release of contaminates regarding beneficial use and land disposal of secondary materials, such as sewage sludge, compost, fly ash, little attention has been paid to the effect of repeated additions of secondary materials on the soils' leaching behavior. Traditionally, land application of SSC has been carried out continuously and annually with a certain loading rate. Multiple applications over many years can result in near-surface soil accumulation or long-term groundwater impact (van der Sloot, 2002).

In this study, the leaching environmental assessment framework (LEAF) approach (Kosson et al., 2014a, 2014b) is extended to evaluate the potential leaching from repetitive application of composted sewage sludge to soils. Percolation tests with repeated additions of compost were conducted to evaluate the release of heavy metals during long-term land application of sewage sludge compost. In addition, two kinds of soils (a loamy soil with high organic matter content and a clay soil with low organic matter content) were used in the study and the effect of successive SSC additions on the two soil types were compared. SSC was added to the soil equivalent to the application rate of 48 t/ha/year for 4 times and after each addition the percolation test was carried out to evaluate leaching of heavy metals from infiltration. After the percolation test, pH-dependent leaching tests were conducted to compare the leaching characteristics of heavy metals in reference soils and the soils with up to four cycles of SSC addition and leaching.

2. Material and methods

2.1. Materials

The reference soil (NSG), a loamy soil, was sampled in the 0–25 cm horizon from a farmland area in Suzhou, Jiangsu, China (120°37′E, 31°19′N) (SI Text S1). The reference soil (NSO), a clay soil, was sampled in the 0–25 cm horizon from an urban landscape area in Kunming, Yunnan, China (102°42′E, 25°02′N) (SI Text S2). The sewage sludge compost (SSC) was sampled from composted municipal sewage sludge after a 30-day composting process with green waste (SI Text S3). The physicochemical properties of NSG, NSO, and SSC are shown in Table 1.

2.2. Column design and percolation leaching procedures

2.2.1. Control columns

There were two control columns designed to evaluate the leaching of heavy metals as a function of liquid-to-solid ratio (L/S) without compost application. In two control columns, approximately 360 g of reference soils (NSG and NSO) were compacted up to a height of approximately 15 cm, based on the soil density of about 1.35 g/cm^3 in the field application scenario. The percolation test was conducted based on the EPA method 1314 (US EPA, 2014a, 2014b). To prevent deflocculation of clay layers and moderate mobilization of dissolved organic matter, 1 mM $CaCl_2$ was used as eluent and fed upwards continuously through columns with the eluate production rate of 0.75 $\pm$ 0.25 L/S/per day, until a cumulative L/S of 40 L/kg was reached. Eluate samples were collected at the cumulative L/S of 0.2, 0.5, 1.0, 1.5, 2.0, 4.5, 5.0, 9.5, 10, 15, 20, 25, 30, 35, and 40 L/kg, respectively.

2.2.2. Columns with repeated additions of compost

Two columns with mixtures of SSC and a reference soil (NSG or NSO) were designed to evaluate leaching of heavy metals with repeated additions of SSC. The soil within each column experienced four cycles of first blending SSC with soil, followed by a percolation test, post-test soil characterization, and then starting a new cycle by blending additional SSC. In each cycle, blending SSC with soil simulated the addition of SSC to soil and the percolation leaching test was used to estimate the release of heavy metals during one year precipitation. Generally, the column test simulated the field scenario where SSC was added to soil in 4 successive years with the same annual application rate, and after each addition the percolation test was conducted to simulate the annual precipitation.

Initially, in these two columns, SSC was mixed into the reference soil at the dry weight ratio of 4%, corresponding to the 48 ton/ha compost application rate (10 cm tillage). Then, 360 g of the blended material was compacted up to a height of approximately 15 cm and the eluent (1 mM $CaCl_2$) was fed upwards continuously through columns until a cumulative L/S of 8 L/kg was reached. The L/S of approximately 8 corresponded to one year of average precipitation after compost was applied to the soil (SI Text S4). During the leaching procedure, nine eluate fractions were collected at the L/S ratio 0.2, 0.5, 1.0, 1.5, 2.0, 4.5, 5.0, 7.5 and 8 L/kg, respectively.

After each percolation test procedure finished, the blended material in each column was taken out, air dried, and homogenized. Approximately 30 g sample was taken for characterization. Then SSC was added to the blended material again at the dry weight ratio of 4% to start the next percolation procedure cycle.

To evaluate leaching behavior of heavy metals under the reducing condition imposed by biological reaction of the SSC residual in the soil within the column, an extension of the percolation procedure was conducted after the 4th SSC addition. After completion of the percolation procedure described above, eluant flow to the soil was stopped for 1 month, followed by resumption of column eluant flow. After resumption of eluant flow, eluate samples were collected at the cumulative L/S ratio 1.0, 2.0, 3.0, 4.0, 5.0, 6.0, 7.0, 8.0, and 10.0 L/kg, respectively. After the extension percolation procedure, all samples in the columns were taken out for physicochemical characterization and pH-dependent leaching test.

All eluate samples collected during percolation tests were filtered through 0.45 μm membrane before chemical analysis.

2.3. pH-dependent leaching test

A pH-dependent leaching test, US EPA method 1313 (US EPA,