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Leaching characteristic of toxic trace elements in soils amended by sewage sludge compost: A comparison of field and laboratory investigations☆

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ABSTRACT

A 3-years field test and laboratory leaching test have been conducted to assess the environmental impact of land application of sewage sludge compost in conjunction with wheat and rice crops. Considering the complexity and variability of field conditions, we compared the result of laboratory test with the field test to understand the accuracy and uncertainty associated with using the laboratory test to evaluate the field scenario. The laboratory test with cycling of compost additions and water percolation was a high time-efficient and feasible method to simulate the annually repeated additions of compost in the field application scenario. The results of laboratory test were congruent to the 3-years field test regarding the leaching characteristics and geochemical speciation of toxic trace elements. Both the laboratory and the field test showed that repeated additions of compost to soils can increase leaching concentrations of toxic trace elements at neutral to alkaline pH. Increased toxic trace elements leaching was caused by the increase of organic matter from compost application and organic matter dissolution at alkaline pH. Uncertainties of the laboratory test mainly included the negligibility of crop growth and the strongly reducing condition formed with continuous percolation procedure.

1. Introduction

Treatment of sewage sludge is a very important issue all over the world (Ren et al., 2017). Particularly in China, the production of municipal sewage sludge reached up to approximately 35 million tons (98% water content) in 2015. After some stabilization methods, such as composting, sewage sludge potentially can be used as soil amendments. Sewage sludge compost (SSC) is rich in nutrient elements, which can pose advantage for plant growth as well as reduce fertilizer consumption (Cieślik et al., 2015; Charlton et al., 2016). However, SSC contains some contaminants (such as toxic trace elements) that maybe present in a range of concentrations depending on the sludge origin. It might result in near-surface soil accumulation, plant uptake or long-term groundwater impact during land application of SSC (van der Sloot, 2002; Wang et al., 2016). Related research revealed that sewage sludge in China had relatively higher contents of toxic trace elements (TTEs) when compared to other regions, such as EU and US (Dai et al., 2007; Fuentes et al., 2006; Hua et al., 2008; Jones et al., 2014). Therefore, the environmental risk of TTEs should be evaluated when determining the feasibility of long-term land application of SSC.

Field experiments can be useful to evaluate the environmental impact of SSC land application (Bhogal et al., 2003; McGrath and Cegarra, 1992; McGrath et al., 2000; Parat et al., 2005; Wu et al., 2012a, 2012b). Since SSC is applied to soil repeatedly and annually with a certain loading rate, the field experiment should be carried out over multiple cycles with on-going field monitoring to assess the long-term environmental impact. McGrath and Cegarra (1992) and McGrath et al. (2000) conducted the field test for 20 years and the results showed that land application of sewage and SSC will significantly increase the total contents of TTEs in soils. The

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40-years field test reported by Udom et al. (2004) indicated that Pb and Cu tended to migrate to soils at deeper depth. Considering that field experiments are time-consuming and difficult to control, use of laboratory leaching tests to simulate the field application scenario and estimate the long-term environmental impact would be advantageous.

Leaching tests are commonly used in the laboratory to estimate release of TTEs during beneficial reuse of secondary materials (such as fly ash, biochar, compost etc.) on land. Leaching, the transfer of TTEs from the solid phase of soil to soil water, is considered as an initial and essential step in both plant uptake of TTEs and release of TTEs to water resources (Fang et al., 2016a, 2017; Venegas et al., 2016). Results from various laboratory leaching tests have been conducted to determine the bioavailable and mobile fractions of TTEs (Jalali and Arfania, 2011; Toribio and Romanyà, 2006).

However, the relationships between laboratory testing and field behavior must be known to verify the application of laboratory test to predict the behavior of TTEs in the real scenario. In the real field scenario, there are multiple biogeochemical factors that may influence TTEs behavior including irreversible sequestration in soil components and partitioning between adsorption phases (Fang et al., 2016b; Honma et al., 2016). These factors (such as plant exudates and redox conditions) are difficult to control in the laboratory leaching test. The complexity and variability of field conditions imparts the need to compare laboratory and field results to understand the accuracy and uncertainty associated with using laboratory testing to estimate mobility and bioavailability of TTEs under real field scenarios.

In this paper, influence of SSC land application on leaching characteristics and plant uptake of TTEs were investigated using a field test for 3 years. Results of the field test were compared with laboratory leaching test results (including column test and pH-dependent leaching test results, reported by Fang et al. (2017)) to verify the applicability of using laboratory testing as a tool to simulate the real scenario and evaluate long-term environmental risk. The experiment design protocol and uncertainties associated with the laboratory test were also discussed.

2. Material and methods

2.1. Field test description

The field test was conducted for 3 years from Dec. 2012 to Dec. 2015 to investigate the impact of SSC application on plant growth and soil characteristics in Suzhou, Jiangsu, China (120°37′ E, 31°19′ N). The soil properties and detailed information about the field site were shown in Supplementary Text 1. There were six treatments in the field test site as shown in Table 1. Each treatment was carried out in triplicate plots (24 m^2 each) (Fig. S1). The field site was used for plant growth, including the growing season of winter wheat from December to April and the growing season of rice from May to November. Agricultural activities for the field test were shown in Table 2. During the field test, soils were sampled four times each year at December, March, May, and September. On each plot (24 m^2), top soils at depth of 0–10 cm and subsoils at depth of 10–20 cm were sampled. Three soil-cores at each depth were taken and mixed to form a representative sample which was air-dried for further analysis. Samples of plant (wheat and rice) straw and grain were collected during plant harvest. On each plot, an area of 1 m^2 was selected randomly and plants in that area were collected as the representative samples. Then the straw and grain of plants were separated and oven-dried at 70 °C.

2.2. Physicochemical properties analysis

Physicochemical properties analysis of SSC and soils included pH, electrical conductivity (EC), dissolved organic carbon (DOC), total organic carbon (TOC), humic substances (HAs and FAs), Fe (hydr)oxides, Al (hydr)oxides, and total content of TTEs (As, Cd, Cr, Cu, Ni, and Pb) (Supplementary Text 3).

2.3. Measurement of TTEs total contents

Total contents of TTEs in SSC, soils, plant straw, and plant grain were determined after acid digestion. The acid used was lefort aqua regia (12 mL) with HNO_3: HCl of 3:1.

Bio-concentration factor (BCF) was calculated according to Eq. (1) and used to the reveal abilities of various plant species to take up and accumulate TTEs from soils.

$$BCF = \frac{Heavy\ metal\ concentration\ in\ plant\ tissues\ (mg/kg)}{Heavy\ metal\ concentration\ in\ soils\ (mg/kg)} \quad (1)$$

2.4. Statistical analysis

One-way analysis of variance (ANOVA) was used to determine the main effect of treatment, sampling time, and SSC addition times on total contents of TTEs in soils. For plants, effects of treatment and SSC addition times were investigated. Regarding the total contents of TTEs in soils, there are 432 data in total (72 data for each treatment, 108 data for each sampling time, and 144 data for each SSC addition times). Regarding the total contents of TTEs in plants straw or rice, there are 54 data in total (9 data for each treatment and 18 data for each SSC addition times). Tukey's test was performed to group the TTEs content based on the significance level (p) at 0.05. The results of Tukey's test can show the ordering of TTEs content with the change of variables (treatment, sampling time, and SSC addition times).

2.5. Laboratory column experiment to simulate the field scenario

Cycling of SSC additions and water percolation were conducted to simulate the annually repeated additions of SSC in the real field

Table 1
Soil treatments in the field test.

No.	Treatment[a]
1	Control, no application of SSC and no initial application of chemical fertilizer
2	Application of chemical fertilizer at common rate
3	Application of SSC at loading rate of about 33 ton (dry matter)/ha, 2 times each year
4	Application of SSC at loading rate of about 16 ton (dry matter)/ha, 2 times each year
5	Application of SSC at loading rate of about 7.5 ton (dry matter)/ha, 2 times each year
6	Application of SSC at loading rate of about 2 ton (dry matter)/ha, 2 times each year

[a] This is the treatment during initial planting. Then for all the six treatments, chemical fertilizer was added once or twice during the growing season at a common time and application rate. SSC was applied to the soils at 0–10 cm.