



## Environmental evaluation of the application of compost sewage sludge to landscaping as soil amendments: a field experiment on the grassland soils in Beijing

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### ABSTRACT

The improvement of landscaping soil fertility using organic wastes could be a feasible practice to avoid contaminants from entering the food chain but at the risk of polluting the soil, surface water, and groundwater. Currently, millions of tons of sewage sludge compost is used to improve the nutrient contents in the soil each year in China. To study the mobility of heavy metals, N, P, and EDCs in the soil, an experiment was conducted at a field site for over two years to study the application of sewage sludge compost to degraded grassland soils in Beijing. The grassland soil was amended with 6, 12, and 18 t/ha of compost sludge. The results indicate that the concentrations of Cu, Zn, and Cr in the 0–20 cm surface soil layer increased with an increase in the dosage of the compost sewage sludge, while the concentrations of Ni, Cd, As, Hg, and Pb did not change significantly. In the 0–100 cm soil profile, the concentrations of the DTPA-extractable Zn and Ni were significantly higher than that of the control, while the concentrations of the DTPA-extractable Ni, Cd, As, and Hg did not vary significantly. The concentrations of the DTPA-extractable Cu, Pb, and Ni were reduced as the depth of soil increased. The concentrations of heavy metals, such as Cd and Cr, in the surface run-off during the rainy season had exceeded the China surface water environmental quality standards (III). The estradiol toxic equivalency was below 10 and 0.1 ng/g in compost sludge and grassland soil, respectively. The soil phosphatase and catalase increased initially and then decreased with the increasing dosage of the compost sludge. The application of sewage sludge compost increased the content of phosphorus in the 0–20 cm surface soil layer by 100–200%, and the ammonia content was increased by 16–100%; the nitrate content was, however, lowered by approximately 35%. The concentrations of

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nitrogen and phosphorus in the surface run-off of content met the surface water environmental quality standards, while the content of heavy metals Cd and Cr did not. Therefore, the application of compost sludge should be avoided in rainy seasons to reduce the environmental risks of contamination from heavy metals.

*Keywords:* Compost sewage sludge; Heavy metals; Forestry application; Pollutant mobility; Soil enzymes

## 1. Introduction

Landscaping soil, sandy soil, and limestone quarries soil in Beijing are often subjected to severe degradation processes accompanied by a decline of the organic matter contents in soil, adversely affecting soil fertility and increasing the risks of erosion and desertification. In the European Union, approximately 4 million tons of dry sludge is applied yearly to the land. The sewage sludge production is expected to be 13 million tons in 2020, and 44% of this total is expected to be recycled for use on land [1]. Although the agricultural use of sewage sludge is regulated by legislation, there is no European framework for its application to nonagricultural lands; therefore, many countries use sludge for different purposes, such as poor land restoration, forestry, and degraded landscaping [2]. Wang et al. reported that sewage sludge demonstrated potential for use in the application of urban landscaping in China [3]. Yang and Zhao reported that the landscaping area in Beijing is appropriate for sludge disposal, with favorable economic and ecological benefits [4]. The landscaping industry in Beijing demands approximately 4.5 million tons of new soil and 0.18 million tons of peat soil each year. According to the ratio of 1:3 of compost to new soil, 3.75 million tons of 80% moisture content sludge can be used annually in forestry. If all of the sludge compost is used to replace peat soil, 0.6 million tons of 80% moisture content sludge can be consumed annually. That is, the annual consumptive use of wet sludge of the Beijing forestry sector can be 4.35 million tons. Because of unquestionable advantages in the soil utilization of compost sewage sludge, this disposal method is gaining increasing interest. High contents of organic matter and nutrients make sewage sludge a perfect material for use in the fertilization of landscaping soil [5,6]. Despite the obvious merits, compost sewage sludge may also lead to both environmental contamination and ecological deterioration, which require in-depth chemical and eco-toxicological evaluation. Heavy metals accumulated in sludge could be transferred to soils, which would appear to be a virtually permanent pollution to soils and consequently to groundwater and may result in adverse effects to

plants. Toxic metals in sludge can affect enzyme activity in the amended soils. Almendro-Candel et al. reported that Ni exhibited a high mobility due to sewage sludge application to soil and groundwater [7]. Contin et al. reported that soil receiving 10 times the allowed amount of sewage sludge was more resistant to the additions of Pb and Zn [8]. Metal extractability and availability from compost sludge and soil, which can be tested by DTPA extraction, are important when compost sludge is applied to soil [9]. In addition to chemical quantification of heavy metals, biological tests are the best and most sensitive method of evaluating the hazards of organic compounds related to the application of sewage sludge in soil. Zapusek and Lestan reported that soil mixtures containing sewage sludge from the Ljubljana and Maribor wastewater treatment plants (WWTPs) resulted in an increase of dehydrogenase activity and phosphomonoesterase activity, respectively [10]. Cheng and Grewal reported that compost amendment temporarily reduced the number of nematode genera [11]. Some trace organic pollutants in sewage sludge exhibit toxic effects of carcinogenicity, mutagenicity, genotoxicity, and endocrine disruptive activity.

Reclamation of degraded landscaping areas using compost sewage sludge may be subjected to less restrictive regulations than the application of these materials in agriculture. The five-year plans of Beijing indicated that until the year 2016, approximately 47% of sludge will be composted and recycled for use in nonagricultural lands, especially for landscaping soil, sandy soil and limestone quarries [12]. Different sludge properties may have a multidirectional influence on the overall toxicity of sewage sludge-amended landscaping soil and on the groundwater quality, e.g. heavy metals toxicity and distribution, endocrine disruptor toxicity, and nitrogen or phosphorus eutrophication to groundwater. This toxicity issue of sewage sludge-amended landscaping soil is relatively new and is poorly recognized in the literature. New Chinese national quality standards of the use of sludge in landscape use could also inhibit the use of compost sewage sludge for landscape purposes. Further studies are required to refine the knowledge of the use of

sewage sludge in landscaping before adopting a final decision.

To address this lack of knowledge, the aims of this work were to evaluate the possibility of applying compost sewage sludge to landscaping as soil amendments, the environmental risks to soil and groundwater of the use of sludge in landscaping soil, and the practical implications of the use of sludge for the improvement of landscaping soil fertility.

## 2. Materials and methods

### 2.1. Material characteristics

The compost sewage sludge was obtained from the Panggezhuang sludge composting plant in Beijing, China, using the aerobic windrow composting method. The sludge was from the Gaobeidian municipal WWTP, using primary and secondary treatments. The sludge originated from a gravity thickener (i.e. primary thickened sludge) and from a mechanical belt thickener (i.e. waste thickened sludge). A mixture of 60% sludge (of 80% moisture content) and 40% sawdust was produced, which, after 15 d of aerobic composting, was used as the material in our study. The characteristics of the compost sewage sludge from the Panggezhuang sludge composting plant are presented in Table 1.

The characteristics of the soil from the Mentougou area are presented in Table 2.

### 2.2. Experimental procedure

The experiment was performed in the Beijing Mentougou District over two years. The plant *Festuca arundinacea*, which is widely planted in China as turf-grass, was selected as the testing sample. *Festuca arundinacea* were planted in soil mixed with compost sludge. Each processed grassland area was 1 ha. Four types of experiments were conducted which consisted of using 0, 6, 12, and 18 t/ha of compost sludge. The lawn was manicured twice a year.

### 2.3. Analytical methods

Soil and sludge samples were first digested with aqua regia and  $\text{HClO}_4$  [13], and then the contents of

total heavy metals (Cu, Zn, Pb, Cr, Cd, As, Hg and Ni) were measured by ICP using a ICP-MS (X Series, Thermo) apparatus. The analysis of the sludge and soil samples was performed according to chemical analytical methods for soil [14]. The pH value was measured in a 1:2.5 (w/v)  $\text{CaCl}_2$  solution using a WTW-82362 pH meter (WTW, Germany). The analysis of nitrogen, phosphorus, and potassium was performed using the method proposed by Sempere et al. [15]. Total organic carbon (TOC) was determined using a solid TOC analyzer (SCSH, TOC-VCPH, Japan). The analytical precision of the TOC measurement was checked against a certified standard material, and all of the samples fell within  $\pm 10\%$  of the certified values. The endocrine disrupting effects were determined using a recombinant-gene yeast [16]. The enzymatic activity test was performed according to Tabatabai and Bremner [17].

## 3. Results and discussion

### 3.1. Effect of compost sludge on the heavy metals in topsoil

With the addition of increasing amounts of compost sludge, the contents of Cu, Zn, Cr, and Pb increased more significantly in the different topsoil treatments (Fig. 1). In contrast, the addition of the compost sludge did not significantly increase the concentrations of Cd and Hg, which may be because of the relatively low initial concentrations of Cd and Hg in the sludge combined with a certain absorption capacity of the turf grass to Cd and a certain volatility to Hg. The addition of compost sludge did not increase the heavy metals, such as Ni and As, because the contents of Ni and As were nearly the same in the compost sludge and the lawn topsoil.

Due to the relatively high content of zinc in the compost sludge, the concentration of zinc for the different topsoil treatments was the highest among the eight heavy metals and thus can determine the safety of the sludge application rate to control secondary sludge pollution. Similar ranges of heavy metal concentrations were reported in previous studies [18–20]. There is a linear relationship between the amount of zinc in the lawn topsoil and the amount of added

Table 1  
Characteristics of the compost sewage sludge

Total nutrient (g/kg)	Organic carbon (g/kg)	TN (g/kg)	TP (g/kg)	TK (g/kg)	Moisture (%)	pH	Boron (g/kg)
90.9	403.4	36.7	119.6	12.6	32	7.5–8.5	35.15
Pb (mg/kg)	Cd (mg/kg)	Cr (mg/kg)	Cu (mg/kg)	Zn (mg/kg)	Ni (mg/kg)	As (mg/kg)	Hg (mg/kg)
37.05	0.42	64.66	139.45	658.09	22.70	13.76	6.07

Table 2  
Analytical characteristics of grassland soil (0–20 cm) (units: mg/kg)

Organic carbon	TN	Nitrogen	AP	AK	As	Pb	Cd	Cr	Hg
26.2	0.65	94.8	226.1	151.5	3.51	14.8	0.35	51.2	0.88

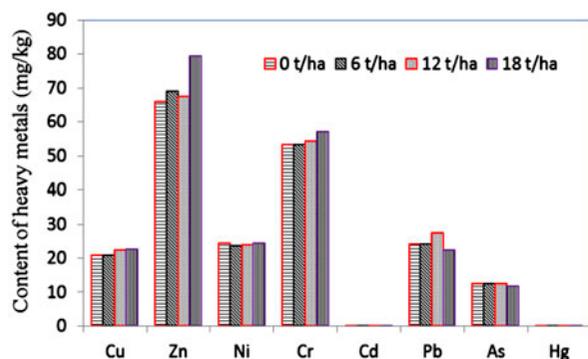


Fig. 1. Content of heavy metals in the 0–20 cm surface layer of grassland soil at different dosages of compost sewage sludge.

compost sludge, which can be expressed by the following formula:

$$y = 0.6417x + 64.65 \quad (R^2 = 0.8728) \quad (1)$$

where  $y$ —the zinc content of the topsoil, mg/kg,  $x$ —compost sludge application dosage, t/ha.

All of the heavy metals in the soil were lower than the national standard after two years of application. According to the Chinese soil environmental quality standards class II (GB15618-1995) of zinc (300 mg/kg, pH > 7.5) and based on an annual administered 12 t/ha compost sludge application rate in this study, the zinc content in the 0–20 cm topsoil would exceed the soil environmental quality standards after approximately 30 years of continuous application. If the application of this compost sludge soil to lawns is to be maintained over 100 years, the annual compost sludge application rate should be lower than 3.7 t/ha. Notably, the target value for zinc is 4,500 and 140 mg/kg in France and the Netherlands, respectively, and the intervention values for zinc are 9,000 and 720 mg/kg in France and the Netherlands, respectively [6]. The limited eco-screening value of zinc is 160 mg/kg in the USA [21]. The limited values of trace elements in soils depend on the national circumstances. If the lawn application of this compost sludge soil is to be maintained over 100 years, the annual compost sludge

application rate should be lower than 1.85, 8.88, and 1.11 t/ha in the USA, France, and the Netherlands, respectively.

### 3.2. Effect of compost sludge on the DTPA-extractable heavy metals at different depths of the soil

In the two years of the field experiment, the grass was harvested four times. The DTPA-extractable heavy metals within the different depths of the soil for all treatments are shown in Fig. 2. With an increasing dosage of the compost sludge, the contents of DTPA-extractable Zn and Ni increased significantly for the different treatments and at different soil depths, especially in the topsoil, while the results were different for Cu, Cr, Pb, and Cd. The measurement of the DTPA-extractable heavy metals may reflect the heavy metals in the soil that can be effectively absorbed by living creatures. The DTPA extraction techniques are used to establish relationships between the concentrations of heavy metals in soil and in plants, which could simulate the metal-uptake process, and were found to be useful in estimating the amounts accumulated in plants [22–24].

In each layer of the soil profile, the content of DTPA-extractable Zn was higher than in the control sample because Zn is a type of heavy metal that is easy to transfer and leach [25]. The concentration of DTPA-extractable Zn in the application of 18 t/ha of compost sludge in the 0–20 cm topsoil was three times higher than that for the control treatment (Fig. 2(a)). The DTPA-extractable Zn content in the 20–100 cm layer of soil was 1–2 times higher than that of the control treatment, indicating that the application of compost sludge not only increased the surface-soil Zn accumulation but also increased the Zn that moved downward into the deeper layer soil. For the higher amount of the compost sludge addition, the content of DTPA-extractable Zn was also higher in the different soil layers. Although Zn is a necessary element for plants, the continuous accumulation of excess zinc not only produces toxic effects to plants but also increases human health risks as a result of contaminated drinking water extracted from the groundwater. The DTPA-extractable Zn results agreed well with the

reported results by Méndez (2012), which indicated an increased risk of heavy metals leaching after sewage sludge application [26].

Ni, whose transfer ability is lower than that of Zn, is a type of heavy metal necessary for plants. It can be observed in Fig. 2(b) that with an increase in the amount of compost sludge, the DTPA-extractable Ni content of the soil profile layers also increased significantly. However, DTPA-extractable Ni was mainly accumulated in the 20–40 cm layer. With an increase of soil depth, the DTPA-extractable Ni content decreased due to its limited transfer ability.

The concentrations of the heavy metals of Cu, Cr, Pb, and Cd did not significantly increase with the addition of compost sludge. Cr, Cd, and Pb are toxic

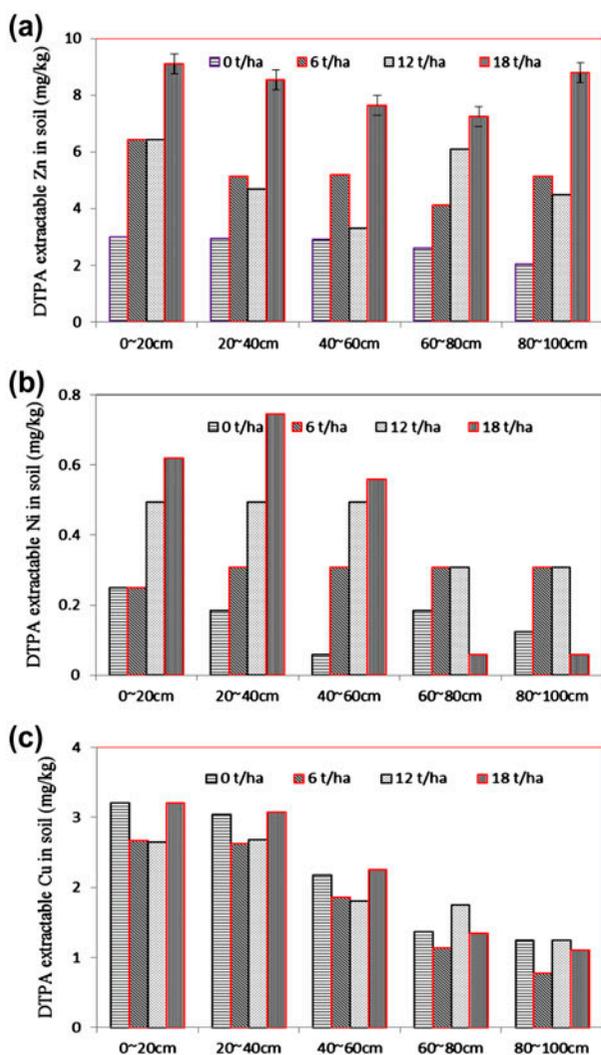


Fig. 2. DTPA extractable contents of heavy metals in the 0–100 cm grassland soil profile at different dosages of compost sewage sludge.

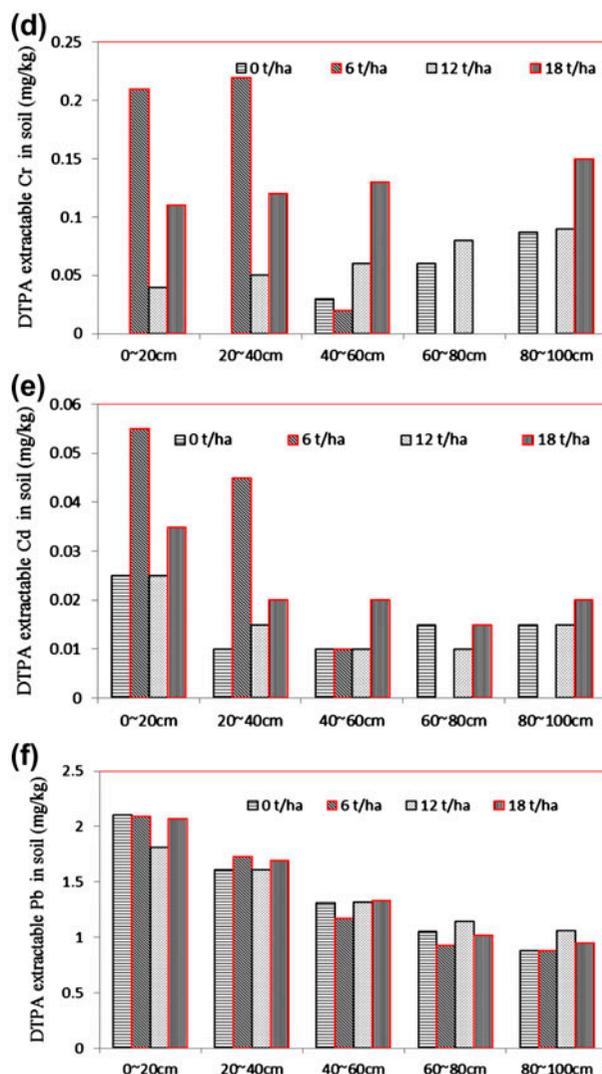


Fig. 2. (Continued).

heavy metals for plants (Fig. 2(c)–(f)). The content of DTPA-extractable Cr in the 0–40 cm control soil layer was nearly zero, while the content increased with the depth in 40–100 cm soil layer. The strong bioactivity and weak mobility of Cr caused Cr to be absorbed by the grass, which restrained its migration from the surface to the deep soil. Wong et al. reported that the level of DTPA-extractable Cr was below 2 mg/kg, indicating the low extractability of Cr in all of the sludge samples considered [27]. For the 6 t/ha compost sludge application, Cr mainly accumulated in the 0–40 cm surface layer. As the compost sludge application amount increases, the accumulation of DTPA-extractable Cr gradually increased in the deep soil. For Cu and Pb, whose transfer ability and plant absorbability are lower than that of Ni, the

DTPA-extractable concentrations were found to gradually decrease with the increase of soil depth. The DTPA-extractable Cd content was greater than 0.01 mg/kg, which was below the national standard value for forestry but higher than the national standard value for vegetables. The Pb and Ni results did not agree with the transfer ability or the bioavailability of the results of Fuentes et al. [28].

From the experimental results, we can see that the long-term application of compost sludge will present some pollution risk to both soil and groundwater.

### 3.3. Effect of compost sludge on the nitrogen and phosphorus at different depths of the soil

Because compost sludge is rich in nitrogen, with a level of 36.7 g/kg, its application would increase the  $\text{NH}_4^+$ -N content in the 0–20 cm top soil layer. For deeper soil layers, the  $\text{NH}_4^+$ -N content decreased, but the magnitude of the decrease was not large. In the different soil layers, the  $\text{NH}_4^+$ -N content changed with the different sludge addition treatments, but the difference was not statistically significant (Fig. 3(a)). In each soil profile, the  $\text{NO}_3^-$ -N concentration decreased with increasing depths for all of the sludge additions considered (Fig. 3(b)). The  $\text{NO}_3^-$ -N content levels were lower than that of the control soil, which indicated that compost sludge application rate differences did not change the levels of  $\text{NO}_3^-$ -N significantly. Because nitrate and ammonium nitrogen can be directly absorbed by plants, the downward migration of these two forms of nitrogen was observed to be insignificant [29]. Mature compost sludge contains a higher content of nitrate than ammonium; therefore, the nitrate concentration was significantly higher than that of ammonium in the surface soil. Because turf grass (such as bluegrass and tall fescue) roots have a strong  $\text{NO}_3^-$ -N affinity and absorption capacity, the nitrate content in the surface soil was higher than the  $\text{NH}_4^+$ -N content [30]. For the 18 t/ha sludge treatment, the  $\text{NO}_3^-$ -N content in the 80–100 cm soil layer was significantly higher than that for the other treatments, indicating that a large amount of application of compost sludge poses a potential risk of  $\text{NO}_3^-$ -N contamination to groundwater. The result herein could be applied to determine the appropriate amount of compost sludge to reduce the impact of  $\text{NO}_3^-$ -N on the water environment.

After two years of compost sludge application, the content of the available phosphorus (AP) in the surface 0–20 cm soil layer was nearly two times higher than that in the control treatment (Fig. 3(c)). For deeper soil layers, the available soil phosphorus content decreased. However, the contents of AP in the

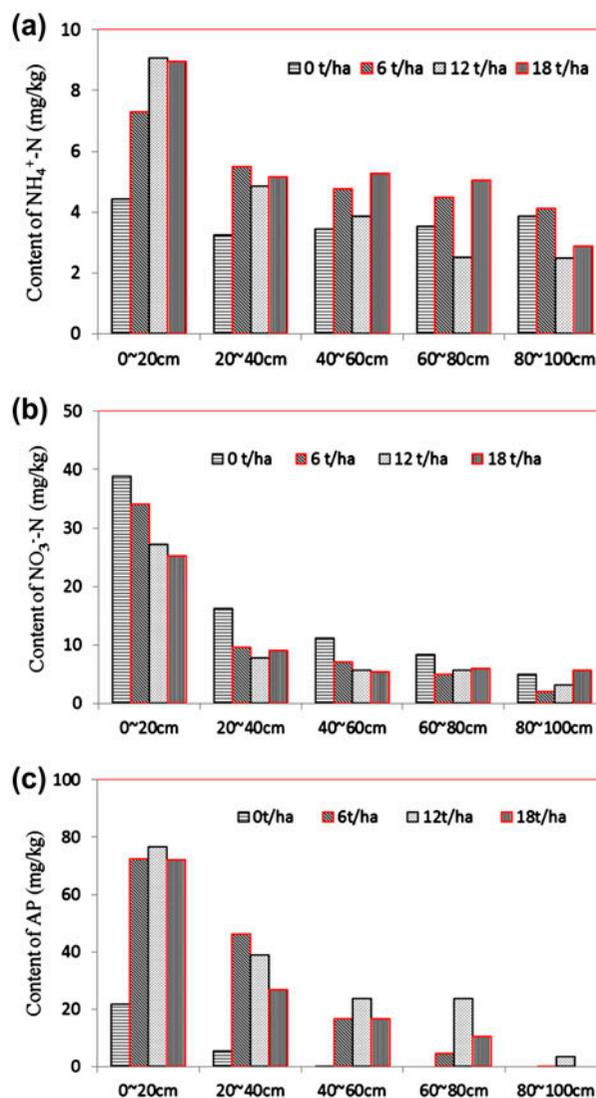


Fig. 3. Content of nitrogen and phosphorus in the 0–100 cm grassland soil profile at different dosages of compost sewage sludge.

different soil layers of the different sludge treatments were higher than those of the control treatment. For three types of compost sludge addition dosages, the contents of the AP were all greater than 60 mg/kg, suggesting a leaching risk. When the AP content was less than 60 mg/kg in arable soil, lower concentrations of phosphorus in the drainage were detected. Notably, when the AP content was greater than 60 mg/kg, the phosphorus content in the drainage increased rapidly. This result is consistent with the research results reported by Heckrath et al. [31], who considered 60 mg/kg had exceeded the absorption level for the crops grown and the soil phosphorus holding capacity; therefore, 60 mg/kg of AP can be used as a

migration-potential critical indicator to assess the phosphorus content in the drainage.

### 3.4. Effect of compost sludge on the estradiol toxic equivalency (EEQ) in different treatment soils

The EEQ in Beijing municipal sewage sludge was found to be in the range of 0.9–17.7 ng/g (dry weight), indicating moderate levels of estrogenic effects [32]. The EEQ was below 10 ng/g in the compost sewage sludge, and the average concentration level was 3.8 ng/g (Fig. 4). These results are comparable with previously published values by Song et al. [33]. EEQ was not detected in the control grassland soil and in the compost sludge addition dosage of 6 t/ha. The EEQs were 0.02 and 0.10 ng/g for the compost sludge addition dosage of 12 and 18 t/ha, respectively, indicating that the extensive use of composted sludge can cause the accumulation of endocrine disruptors in the soil.

### 3.5. Effect of compost sludge on soil phosphatase and catalase for different soil treatments

Applying different amounts of compost sludge into grassland soil influenced the catalase and phosphatase activities to a certain extent. The enzymatic activities have been considered as indicators of the soil quality because soil enzymes have been reported to be highly sensitive to soil environmental conditions, especially to heavy metals [34]. The addition of 6 t/ha of compost sludge significantly increased the activities of the two enzymes, while the two types of enzyme activities exhibited a downward trend with an increase in compost sludge dosage (Fig. 5). The catalase activity was lower than that in the control test when the amount of compost sludge added reached 6 t/ha because the soil DTPA-extractable concentrations of the trace elements increased. These results are supported by the findings of Dar [35] and Belén Hinojosa [34], who found

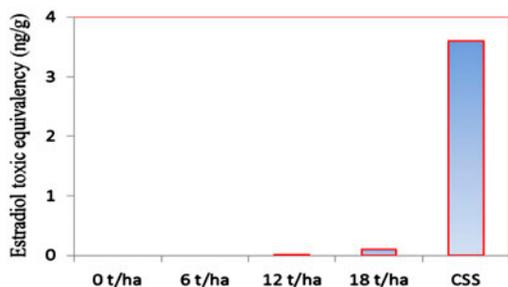


Fig. 4. EEQ in compost sewage sludge and grassland soil.

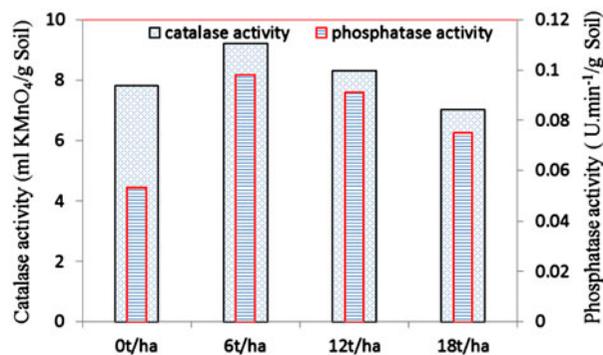


Fig. 5. Variation of the soil phosphatase and catalase at different dosages of compost sewage sludge.

reduced diversity of bacteria and enzymatic activities in the soils. The sensitivity of phosphatase activity was lower than the sensitivity of catalase to compost sludge because the grassland soil was relatively barren, resulting in low phosphatase activity in the control soil. The recovery of soil phosphatase enzyme activities varied widely, ranging from 60 to 90%. In addition, the change of soil enzyme activities varied widely and depended on the soil type and the specific enzyme.

### 3.6. Effect of compost sludge on surface run-off pollution

During heavy rains or irrigation, lawn drainage run-off might enter the surrounding water environment, thereby leading to contaminants possibly having an impact on the water quality. Under conditions of 3 mm/h irrigation intensity and 6 min of irrigation time, the content of the heavy metals Cd and Cr exceeds the China Surface Water Quality Standards class III limitation value (GB3838-2002), indicating that the heavy metals in compost sludge caused major toxic effects on surface run-off (Fig. 7). In the three treatments, the contents of heavy metals, such as Cu, Pb, Cd, Cr, As, and Ni, as well as nitrogen and phosphorus were higher than those of the control experiment in the lawn-surface drainage water, meeting China Surface Water Quality Standards class III limitation values (Figs. 6 and 7). The result indicated that the lawn drainage water might cause a minimal risk to the surface water environment, with Cd and Cr in run-off water being of greatest concern, which was found in the previous studies of Cornua et al. [36].

For nitrogen, the risk to water transfer and pollution is higher, but our results suggest that by using sludge in comparable doses, the export by run-off is relatively low, at levels far below the limitation values (Beaudoin et al. [37]).

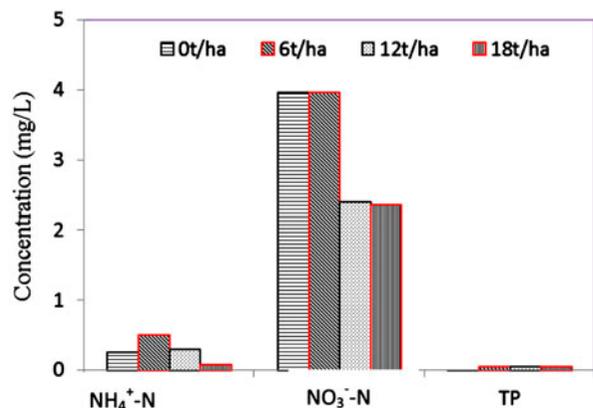


Fig. 6. Content of nitrogen and phosphorus in the surface run-off.

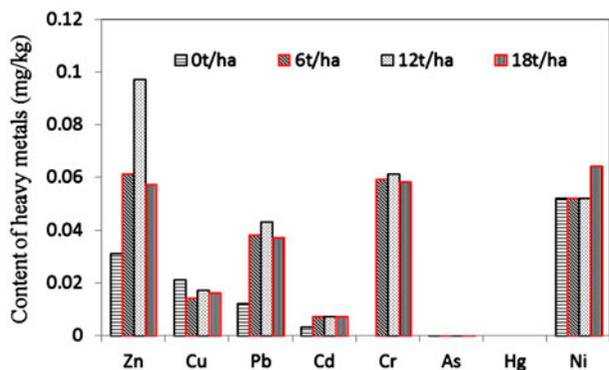


Fig. 7. Content of heavy metals in the surface run-off.

#### 4. Conclusions

This study indicated that in Huabei plain soil environments, the application of compost sludge as soil amendments to landscaping soil, sandy soil, and limestone quarries soil is a possible means of sludge disposal because appropriate amounts of compost sludge could increase the soil fertility and improve soil enzymatic activity. Beijing forestry can potentially consume 4.35 million tons of wet sludge annually, which represents a new approach for sludge disposal that can improve the soil enzymatic activity and soil fertility. Through the results of the two-year study, the appropriate sludge application amount in soil was found to be no greater than 6 t/ha. After approximately 30 years of continuous application, the heavy metals concentration would exceed the soil environmental quality standards. The application of compost sludge could increase the contents of DTPA-extractable heavy metals and improve their bioavailability and mobility; therefore, the addition should be avoided in rainy

seasons to reduce the environmental risks of heavy metals. The application should be re-vegetated and irrigated before seasonal rainfall periods to reduce the risk of N and P losses.

For large-scale application in real field conditions, more in-depth studies are required based on production experiments in a continuous mode to avoid the occurrence of secondary environmental pollution.

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