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Ecotoxicological Assessment of Potentially Toxic Elements in Waterworks Sludge Amended Soils Using Bermudagrass Bioassay

Sai Leung Ng

Graduate Institute of Earth Science, Chinese Culture University, Taipei 11114, Taiwan; wsl7@ulive.pccu.edu.tw; Tel.: +886-2-2861-0511 (ext. 25722)

Abstract: Waterworks sludge has the potential to be used as a soil amendment, but the ecotoxicological risk of potentially toxic elements should not be underestimated. In this regard, this study determined the contents of nine potentially toxic elements (Cr, Ni, Cd, Cu, Pb, Zn, As, Mn, and Al) of bermudagrass [*Cynodon dactylon* (L.) Pers.] grown in waterworks sludge amended soils. Treatments involved different loading rates of waterworks sludge, soil types, and fertilization options that represented different scenarios of greening applications. The recommended metal levels in plant tissues and maximum tolerable levels for feeding cattle are adopted as benchmarks for gauging the ecotoxicological risk to the first and second trophic levels of the ecosystem, respectively. No recommended levels for potentially toxic elements are exceeded when sludge loading rate is not higher than 50% (*wt*/*wt*). When various fertilization treatments are applied to 25% (*wt*/*wt*) sludge amended soils, the accumulation of aluminum and zinc deserves our attention because a few samples exceed the recommended levels. They are mainly samples of below-ground biomass. Overall, using waterworks sludge as a soil amendment does not cause an obvious ecotoxicological risk. The findings can provide a valuable reference to other cities for the sustainable management of waterworks sludge.

Keywords: *Cynodon dactylon* (L.) Pers.; environmental risk; greening; pot experiment; soil amendment

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1. Introduction

The accessibility of clean water for drinking and domestic use is a human right [1,2], but clean water does not exist naturally. Transforming raw water into clean water requires a series of purification techniques that may involve coagulation and flocculation, sedimentation, filtration, and disinfection [3]. As one integral procedure in the process of water treatment, coagulants, such as aluminum sulphate and/or iron-based salts, are added to the raw water to enable the flocculation of suspended particles [4,5]. The flocs collected by means of sedimentation and filtration are known as waterworks sludge [6], water treatment residuals [7], drinking water sludge [8], or simply called alum sludge [9].

The growth of the global population and the change in lifestyle triggers the increasing demand for clean water that undoubtedly translates into the increase in the production of waterworks sludge. It is difficult to obtain accurate data on the global production of waterworks sludge [6]. Dependent on the treatment techniques used and the quality of raw water, waterworks production comprises 1–3% of the total volume of raw water [6]. It is believed the amount of water treatment sludge generation would be highly likely to rise further in many parts of the world [5,6,10].

In Hong Kong, 56 tonnes of waterworks sludge are produced every day [11]. Waterworks sludge initially is dewatered and compressed to become sludge cakes. The cakes are treated as a special type of waste and the landfill is the only channel for sludge disposal [11,12]. Because a large amount of energy is required for the production of dry sludge cakes and Hong Kong's landfill capacity is anticipated to be exhausted in the coming decade [12], disposal of waterworks sludge at landfills has been proved to be an unsustainable practice [13]. Therefore, the development of alternate outlets for the disposal of waterworks sludge is urgently needed [14,15].

Waterworks sludge has been considered a useless material for a long time [6]. In recent years, scientists have attempted to use waterworks sludge in different fields, such as wastewater treatment [8,16,17], urban stormwater treatment [18], building and construction [9,19], industrial production [20], and horticulture and arboriculture [21–23]. Good reviews on the uses of waterworks sludge have been provided by [5–7,24].

Among these applications, the horticultural and arboricultural uses for waterworks sludge merit increasing studies because vast amounts of waterworks sludge can be used, and, at the same time, economic rewards can be generated [11]. Existing literature has shown that waterworks sludge has the potential to be a soil substitute or amendment in greening [21,23]. On one hand, waterworks sludge could improve the physical properties of the soil, such as water retaining capacity and hydraulic conductivity [25]. On the other hand, it is "clean" as it does not contain pathogens and poisonous compounds, as opposed to sewage sludge, in most cases [6]. Nevertheless, questions regarding the ecotoxicity of potentially toxic elements have been raised [26,27]. Some specific questions are asked, e.g., will potentially toxic elements accumulate in the plants grown on the soils mixed with waterworks sludge? Will the ecotoxicological risk increase if fertilization treatments are applied to sludge amended soils? Are the plants grown in sludge amended soils safe for consumption by animals or wildlife?

With these questions in mind, this study conducted a series of pot experiments that used bermudagrass [*Cynodon dactylon* (L.) Pers.] as a bioassay for the evaluation of sludge amended soils. Specifically, the treatments involved different loading rates of waterworks sludge, soil types, and fertilization options that represented different scenarios of greening applications. The metal contents in the plant tissues of bermudagrass were monitored to evaluate the ecotoxicological risk of the use of waterworks sludge in greening.

This study can supplement the existing literature on the sustainable management of waterwork sludges. On the practical side, a huge burden on existing landfills can be alleviated if the use of sludge amended soils is proven to be environmentally safe. The experience can provide a valuable reference to other cities around the world.

2. Materials and Methods

2.1. Sludge and Plant Species

The waterworks sludge used in this study was provided by the Tai Po Water Treatment Works of Hong Kong. Wet waterworks sludge was oven-dried at 105 °C for 24 h and cooled down to room temperature, then the dried sludge was ground into small pellets of a diameter less than 2 mm. Two samples of waterworks sludge were digested with nitric acid and hydrochloric acid according to ASTM D3974-09 (Standard Practices for Extraction of Trace Elements from Sediments) [28], then the composition of potentially toxic elements and key chemical constituents (Table 1) were determined using flame atomic absorption spectrometry (FAAS) [29].

Different amounts of sludge pellets were mixed with two types of soils (i.e., decomposed granite (DG) and volcanic soil (VS)) to prepare the growth substrate for different treatments in the subsequent experiments. DG is the weathered materials collected from granite regolith [30]. In Hong Kong, DG is commonly used in gardening and greening because it is reasonably affordable and abundant [31]. The DG used in this study was provided by a local dealer of landscaping supplies. Another soil, VS, which is similarly widespread in Hong Kong [32], was gathered from the Tai Mo Shan Country Park. Although VS is not frequently used for greening, it is included in this study to illustrate situations, such as afforestation or reforestation, that in-situ materials are used.

Table 1. Composition of potentially toxic elements and key chemical constituents of waterworks sludge used in this study.

Parameter	(mg/kg)
Cr	0.6
Ni	160
Cd	0.6
Cu	159
Pb	51
As	202
Zn	199
Al	112,000
Mg	1720
Ca	3,410
Total nitrogen	10,200
Total phosphorus	8750
Chemical oxygen demand	209,000

Four soil samples were analyzed to determine the key soil physical and chemical properties of DG and VS (Table 2), including soil pH, exchangeable acidity, electrical conductivity, total organic carbon, total nitrogen, total phosphorus, and soil texture) [33–35]. While DG and VS are quite similar in soil chemistry, the major difference is the texture, and as a result, the latter has a higher hydraulic conductivity than the former [36].

Table 2. Chemical composition of waterworks sludge used in this study.

Parameter	DG	VS
Soil pH	5.34	4.88
Exchangeable acidity (cmol/kg)	1.44	1.15
Electrical conductivity (mS/cm)	5.66	1.08
Total organic carbon (%)	0.16	1.09
Total nitrogen (%)	0.11	0.09
Total phosphorus (mg/kg)	0.16	0.01
Soil texture (sand%:silt%:clay%)	72.3:16.6:11.1	33.1:25.8:41.1

Bermudagrass [*Cynodon dactylon* (L.) Pers.] was selected as the bioassay to assess the ecotoxicological risk of waterworks sludge amended soils because of a few reasons. First, bermudagrass is the commonest grass species for greening globally [37]. Thus, it can serve as a baseline for other plant species chosen for greening. Second, bermudagrass seeds are commercially available, hence the standardization of the plants can be easily achieved. Third, the physiology of bermudagrass has been extensively studied [38]. Therefore, it is easy to determine the connection between sludge amended soils and plant development [39].

2.2. Pot Experiment

Experiment 1 intended to determine the amounts of potentially toxic elements accumulated in plants under different sludge loadings. There were five loading rates of waterworks sludge, in terms of sludge weight to the total weight (*wt*/*wt*): 0% (i.e., the control, no waterworks sludge added), 25%, 50%, 75%, and 100% (i.e., only waterwork sludge). Sludge amended DG was first packed into 15 (diameter) × 21 (height) cm plastic pots at a bulk density of roughly 1.6 g/cm^3, then 1.59 g grass seeds, equivalent to 90 g/m^2, were seeded onto the pot (equal to 90 g/m^2). Each treatment had four replicates.

Because natural soils and waterworks sludge chemically are low in fertility, fertilization is probably needed when sludge amended soils are used in greening. Therefore, Experiment 2 was conducted to examine the accumulation of potentially toxic elements in plants under different fertilization treatments. The loading rate of waterworks sludge was controlled at 25% *wt*/*wt*, which was determined by Experiment 1 as the optimum loading rate for plant growth. Two types of soils (i.e., DG and VS) were used to prepare sludge amended substrates for Experiment 2. They were packed into plastic pots in a bulk density of 1.6 g/cm^3, identical to Experiment 1. The sowing rate of the 1.59 g/pot was also identical to Experiment 1.

There were seven treatments in total: nitrogen (N), phosphorus (P), peat moss (M), nitrogen and phosphorus (NP), nitrogen and peat moss (NM), phosphorus and peat moss (PM), and nitrogen, phosphorus, and peat moss (NPM). The control treatment was 25% (*wt*/*wt*) sludge amended soil without fertilization. In Experiment 2, the DG group had four replicates, but the VS group had three replicates, respectively.

Both Experiments 1 and 2 were conducted in a greenhouse. The temperature was maintained at 25 ± 3 °C. Pots were watered daily at a rate of 5 mL/cm^2, which was equivalent to 1800 mm of the long-term annual mean precipitation of the Pearl River Delta Area [40]. Pots were rotated and moved to different locations every week to eliminate the environmental effect of their placement [41].

The duration of both Experiments 1 and 2 was 4 months. The above-ground biomass was harvested at a height of 1 cm above the soil's surface every 30 ± 2 days. Therefore, there were a total of four harvests of above-ground biomass in the study period. After the experiments had been completed, the pot was disassembled to harvest the below-ground biomass. Thus, there was only one harvest of below-ground biomass.

Because a few pots were not able to produce biomass, there were some cases of missing data. Eventually, Experiment 1 yielded 48 samples, and Experiment 2 yielded 139 samples. All biomass samples were oven-dried at a temperature of 105 °C for 8 h.

2.3. Chemical Analysis

The digestion method was adopted by Sasmaz et al. [42] and Yabanli et al. [43]. The oven-dried grass samples were turned into ash by heating at 480 °C for 2 h. Approximately 0.5 g of ash was reacted with HCl (2.0 mL, 30%), HNO_3 (2.0 mL, 65%), and H_2O_2 (2.0 mL, 30%). The mixture was heated at a 95 °C hot plate for approximately 1 h until the sample was completely dissolved. Then, distilled water was added to make the volume 25 mL. Concentrations of nine potentially toxic elements (Cr, Ni, Cd, Cu, Pb, As, Zn, Mn, and Al) were determined using flame atomic absorption spectrometry (FAAS) [29].

2.4. Assessment Benchmark

The metal levels in plant tissues recommended by Kabata-Pendias [44] were used as benchmarks to assess the ecotoxicological risk of potentially toxic elements to plants. If the level of a specific metal in plant tissues was higher than the recommended level, it was an indication of excessive accumulation of that metal. The recommended levels suggested by Kabata-Pendias [44] were used in gauging the general situation of the first trophic level (i.e., producers) in the food chain. To assess the ecotoxicological risk of potentially toxic elements to the second trophic level (i.e., herbivores), the maximum tolerable levels (MTL) in the feed for cattle recommended by the National Research Council [45] were adopted as benchmarks. Table 3 lists the recommended levels of nine potentially toxic elements.

2.5. Statistical Test

Data were organized and statistical analysis was performed using IBM SPSS Statistics 25. Data normality was checked by Shapiro–Wilk's test and sample homoscedasticity was tested by Levene's test. One-way analysis of variance (ANOVA) was performed to evaluate the difference in metal levels of above-ground biomass between different treatments. No statistical analysis was conducted to evaluate the data of below-ground samples because only one harvest was performed after the completion of the experiments.

Table 3. Recommended levels of nine potentially toxic elements.

Parameter	Kabata-Pendias [44] (mg/kg)	Maximum Tolerable Levels [45] (mg/kg)
Cr	5	100
Ni	10	100
Cd	5	10
Cu	20	40
Pb	30	100
As	5	30
Zn	100	500
Mn	400	2000
Al	–	1000

3. Results and Discussion

3.1. Experiment 1

Experiment 1 represents the situations under different loading rates of waterworks sludge. Figure 1 shows nine metal levels of the above-ground biomass of bermudagrass in Experiment 1. The levels of all metals in plant tissues under the control treatment (i.e., 0% loading rate) were lower than the recommended levels.

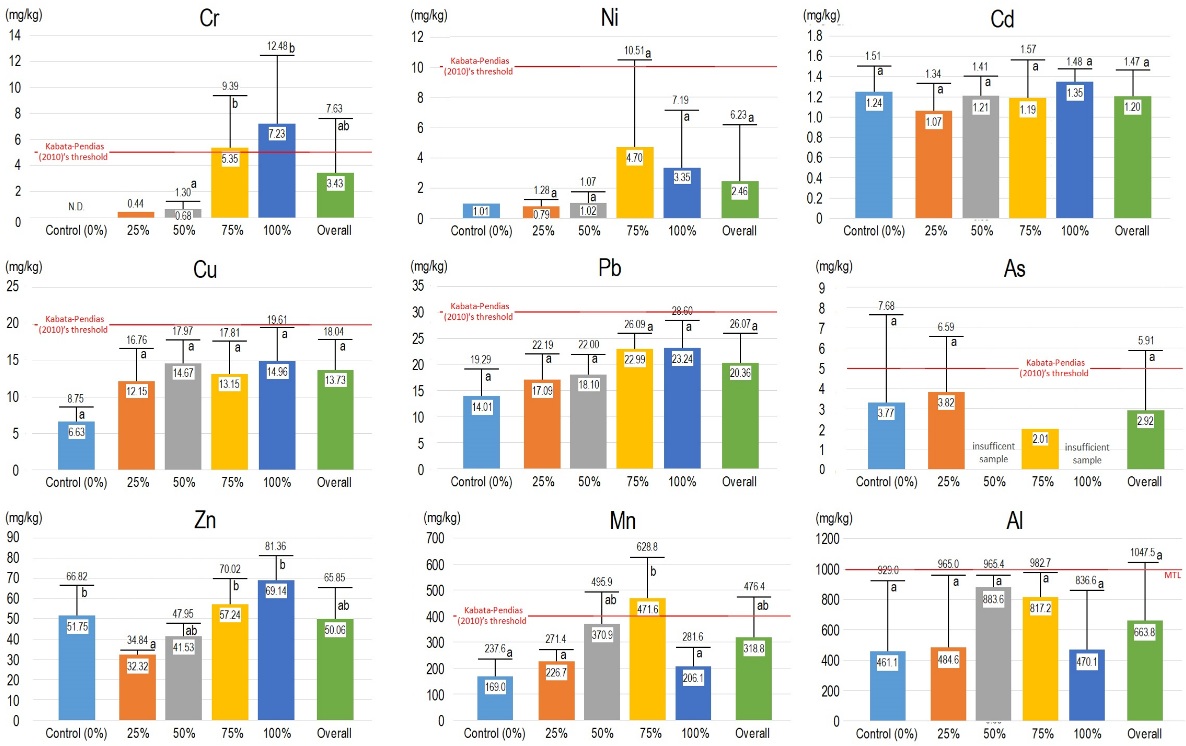

Figure 1. Metal levels of the above-ground biomass of bermudagrass under different sludge loading rates in DG. (Note: N.D. = not determined due to insufficient amount of sample for analysis. Error bars indicate the 95% confidence interval. Different letters denote the statistical difference.)

Compared to the control group, the treatment groups generally have higher levels of higher metals, indicating that the application of waterworks sludge facilitates larger amounts of metals absorbed by plants. The higher the sludge loading rate, the higher the Cr, Ni, Cu, Zn, Mn, and Al contents in plant tissues. The degree and pattern of increase vary in different potentially toxic elements, indicating the different sensitivity of grass in the absorption capacity of potentially toxic elements. On the other hand, the Cd content in plant tissues seems to be indifferent to the change of sludge loading rates. Another interesting finding is that some metal contents (e.g., Ni, Mn, Al) in plant tissues under the treatment of 100% loading rate are lower than that of 75% loading rate. It is probably because the growth of grass is retarded, hence the absorption stops when 100% sludge is used.

When sludge loading rate is low (i.e., 25% or 50%), the levels of all metals or lower than the recommended levels. However, when sludge loading rate is high (i.e., 75% or 100%), the levels of Cr, Zn, and Mn are significantly higher than between the treatments with low loading rates. Furthermore, the levels of Cr and Mn exceed the recommended levels.

The result indicates that, when the soil is loaded with waterworks sludge only, low sludge loading rates do not pose an ecotoxicological risk to the environment. However, high sludge loading rates may cause the excessive accumulation of Cr and Mn, although the exceedances are minor. Because Mn is abundant in the lithosphere, a high level of Mn in grass tissues is not a rare phenomenon [46]. Rocks usually have a relatively high Mn level, ranging between 350 and 2000 mg/kg. Furthermore, Mn is less toxic than most potentially toxic elements [47].

While both 25% and 50% loading rates are safe for the environment, the former is preferable when considering the growth performance of the grass. Because 25% loading rate could produce higher productivity than 50% loading rate [23], the former is considered the optimal loading rate as the rate can increase plant productivity and at the same time does not cause an ecotoxicological problem of potentially toxic elements.

3.2. Experiment 2

Experiment 2 represents the situations of 25% loading rate under different fertilization treatments. Figures 2–5 list the levels of potentially toxic elements in the above-ground and below-ground biomass of bermudagrass grown in DG and VS, respectively. The levels of nine potentially toxic elements in the above-ground and below-ground biomass samples of the control group (i.e., no fertilization treatment) are lower than the recommended levels. The results are consistent with Experiment 1 in that 25% sludge loading rate does not cause a harmful accumulation of nine potentially toxic elements in plants.

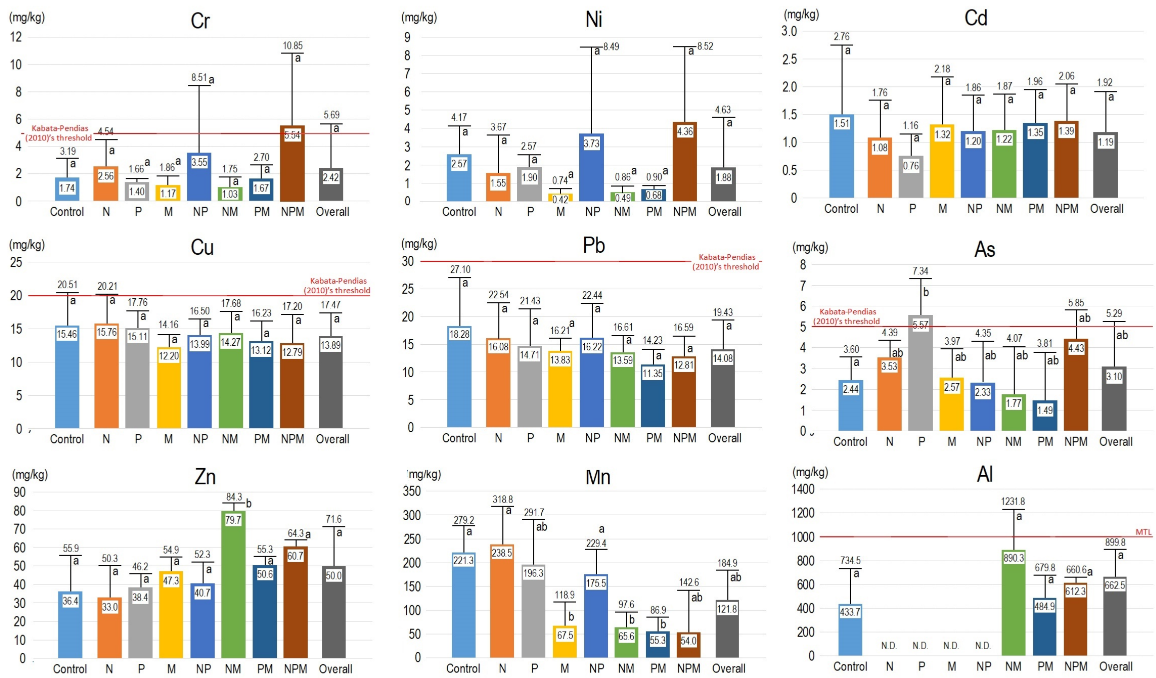

Figure 2. Metal levels of the above-ground biomass of bermudagrass under 25% sludge loading rate and different fertilization treatments in DG. (Note: N.D. = not determined due to insufficient amount of sample for analysis. Error bars indicate the 95% confidence interval. Different letters denote the statistical difference.)

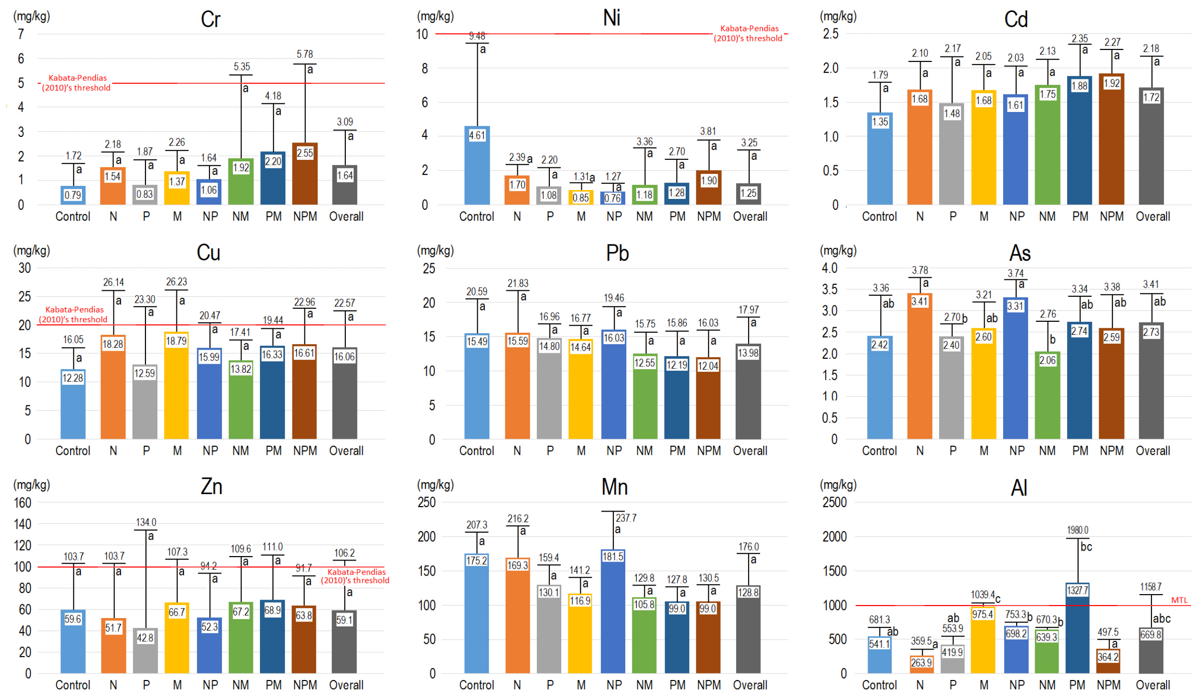

Figure 3. Metal levels of the above-ground biomass of bermudagrass under 25% sludge loading rate and different fertilization treatments in VS. (Note: N.D. = not determined due to insufficient amount of sample for analysis. Error bars indicate the 95% confidence interval. Different letters denote the statistical difference.)

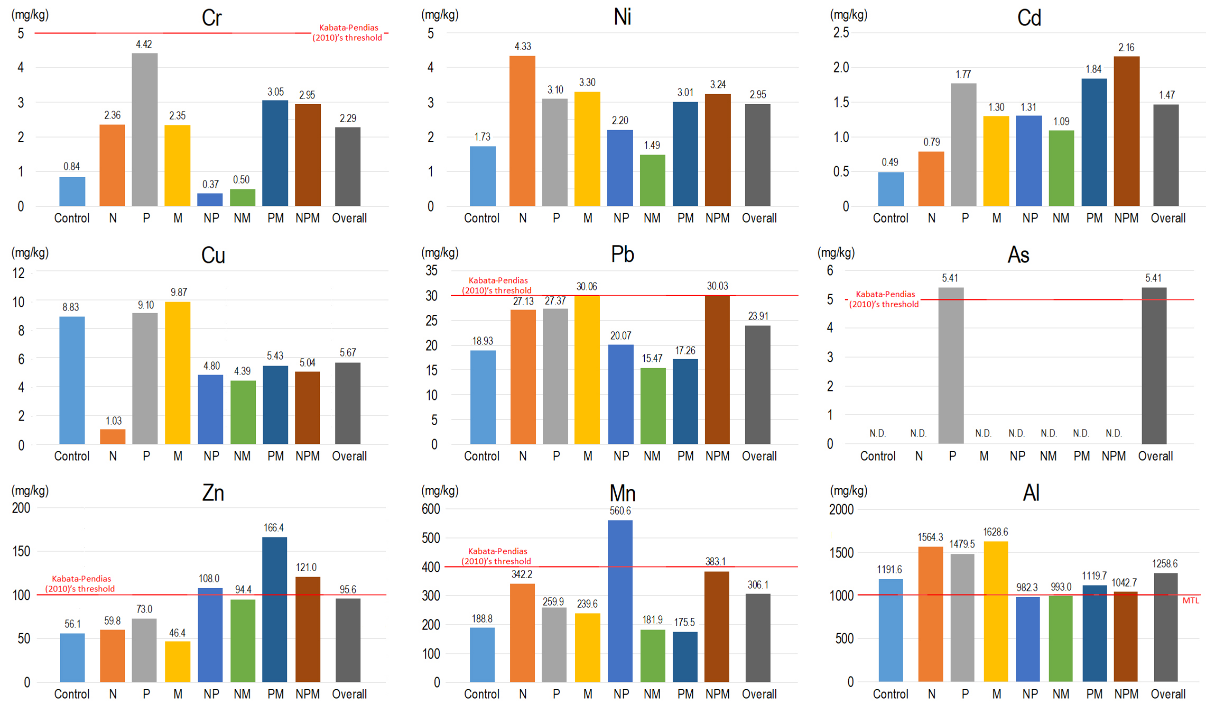

Figure 4. Metal levels of the below-ground biomass of bermudagrass under 25% sludge loading rate and different fertilization treatments in DG. (Note: N.D. = not determined due to insufficient amount of sample for analysis.)

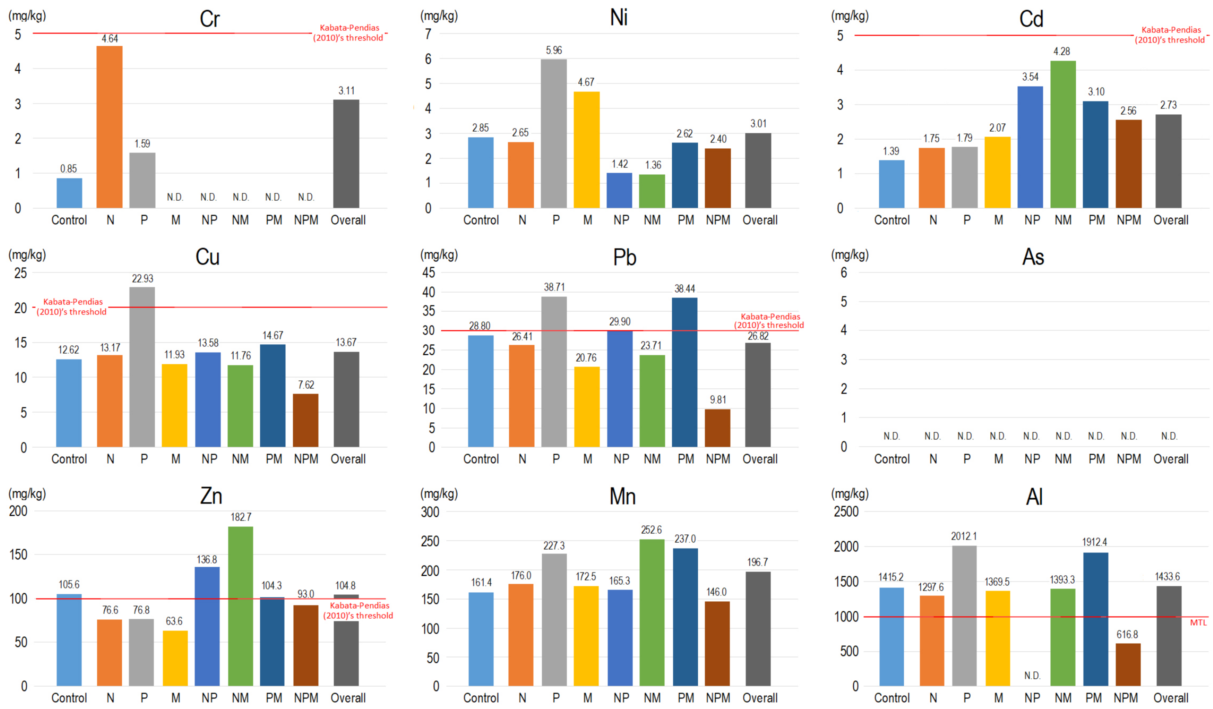

Figure 5. Metal levels of the below-ground biomass of bermudagrass under 25% sludge loading rate and different fertilization treatments in VS. (Note: N.D. = not determined due to insufficient amount of sample for analysis.)

In general, the treatment groups (represented by the "overall" in Figures 2–5 have higher metal contents in plant tissues than the control group, although the differences are not statistically significant. The results may indicate that fertilization enhances the ab-sorption of metals in plants.

Because the fertility of natural soils is generally low, grass grows slowly if there is no fertilization. Therefore, fertilization is a common practice in greening [48]. Although fertilization boosts plant growth, it may increase the uptake of potentially toxic elements at the same time. Therefore, it is important to strike a balance between the benefits (i.e., enhancing plant development) and drawbacks (i.e., ecotoxicological risk) of fertilization when sludge amended soils are used in greening.

Consistent with the results of Experiment 1, the degree and pattern of metal levels vary in different potentially toxic elements because of the different sensitivities of grass in absorbing potentially toxic elements. When one type of fertilizer is used, there are slight increases in the levels of Cr, Cd, Zn, and Al in plant tissues. When fertilizers are combined to use that further boost plant growth, the increases in the levels of these metals in plant tissues become apparent. However, the levels of Cu and As in plant tissues are more or less indifferent to fertilization treatments. Furthermore, the levels of Pb and Mn slightly decrease when fertilizers are combined to use. Nevertheless, the differences in metal levels between single and combined uses of fertilizers are not statistically significant. Future studies are recommended to quantify the relationship between the absorption of potentially toxic elements in plants and soil fertility.

When comparing the metal contents of above-ground and below-ground biomass samples, the latter generally have higher values than the former. In other words, a large part of the absorbed metals is stored in the roots, rather than in the shoots and leaves. Furthermore, below-ground samples have more cases of non-compliance than above-grounded samples. Specifically, Al and Zn contents of below-ground samples commonly exceed the recommended values, prompting the risk of excessive Al and Zn accumulated in the below-ground part of plants when fertilization is applied to sludge amended soils. Al accumulation occurs because of the high Al content in the waterworks sludge. Existing literature has already indicated that the high Al content of waterworks sludge poses the problem of phytotoxicity to the plant communities [19,27]. Because Al is very abundant in the lithosphere, grass usually has a high tolerance to Al. Therefore, no benchmark level of Al is recommended by [45]. Nevertheless, some below-ground samples have an Al level over 1000 mg/kg, implying that it may not be suitable for animal consumption [45]. On the other hand, a high level of Zn in plant tissue probably is because Zn is an essential element for grass [49]. Exceedances of Mn, As, Cu, and Pb are occasionally found in the below-ground samples.

When comparing the metal contents of the biomass samples from DG and VS, there is no obvious difference between them. Nevertheless, the metal contents of the latter seem to be slightly higher values than the former. Both DG and VS have 13 items of exceedance, mainly concentrating on the Al content of below-ground samples. Because the hydraulic conductivity of VS is slightly higher than DG, a longer interaction time between plants and soil water may contribute to a slight increase in the uptake of potentially toxic elements. Future studies are recommended to clarify whether soil type is a significant factor contributing to the differences in the accumulation of potentially toxic elements.

4. Conclusions and Limitations

In conclusion, the ecotoxicological risk of potentially toxic elements is not high when waterworks sludge is used as a soil amendment in greening. There is no exceedance of recommended levels for potentially toxic elements when sludge loading rate is not higher than 50% (*wt*/*wt*), but the optimal loading rate is 25% (*wt*/*wt*). When various fertilization treatments are applied to the soils with a sludge loading rate of 25% (*wt*/*wt*), the ecotoxicological risk of potentially toxic elements is still not high as excessive accumulation of metals in plant tissues is not a common phenomenon. These are minor exceedances, hence their impacts are minimal. There are only very few samples of grass containing excessive amounts of potentially toxic elements. They are mainly below-ground samples. Specifically, Al and Zn deserve more attention because a relatively large amount of these two metals can be found in the below-ground samples. Furthermore, plants grown in sludge amended soils also do not cause an obvious safety problem to herbivores.

This study has a few limitations that future studies should address. First, this study does not represent a thorough assessment of the ecotoxicological risk of waterworks sludge application, as only nine potentially toxic elements are examined. While these potentially toxic elements are either common causes of human poisonings or are closely associated with human ecology, this study represents a preliminary assessment only. For a more comprehensive assessment, other potentially toxic elements, e.g., Hg and Co, should be included in future research. Second, this study uses bermudagrass as a bioassay to tell the situation when waterworks sludge is used in greening. Although bermudagrass is one of the commonest grass species used in greening, other plant species, such as turfgrasses, shrubs, and climbers, would be used as well. It is recommended that a similar assessment of the ecotoxicological risk should be extended to other species. Third, environmental standards for risk management, both regionally and internationally, are not available for the assessment of waterworks sludge. Although the recommended levels proposed by Kabata-Pendias [44] and the National Research Council [45] are used in this study, these recommended levels should be not treated as mandatory standards. Fourth, there is only one harvest of below-ground biomass that does not allow the test of statistical differences between treatments. Furthermore, there are a few missing data because of insufficient amounts of samples for chemical analysis. Consequently, some interpretations are mainly based on the simple comparison of the means. The readers should interpret them with caution.

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