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Microwave-induced heavy metal removal from dewatered biosolids for cost-effective composting

Simeng Li [a,*], Runwei Li [b], Youneng Tang [b], Gang Chen [b]

[a] Department of Civil Engineering, California State Polytechnic University Pomona, 3801 West Temple Avenue, Pomona, CA, 91768, USA

[b] Department of Civil and Environmental Engineering, FAMU-FSU College of Engineering, 2525 Pottsdamer Street, Tallahassee, FL, 32310, USA

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A B S T R A C T

Urbanization and population growth have resulted in the accumulation of heavy metals in biosolids, and these metals act as potent environmental contaminants. In this study, a novel microwave-mediated method of extracting heavy metals from dewatered biosolids was developed. With an electromagnetic power of 140 W for a contact time of 10 s, microwave irradiation effectively induced the immobilization of heavy metals. The treated biosolids were subsequently mixed with acetic acid, sulfuric acid, or ethylenediaminetetraacetic acid (EDTA) for heavy metal extraction. The biosolids in this study were contaminated by different heavy metals, including Cd, Cu, Fe, Pb, Ni, and Zn. Among them, the concentrations of Cd (94.3 ± 14.2 mg/kg) and Pb (888.7 ± 79.8 mg/kg) were considerably above the limits allowed for land application. Conventional extraction methods were found to be insufficient in lowering heavy metal contents below regulation limits, while the microwave-mediated method efficiently increased heavy metal removal by as much as ~3x. After the biosolids were treated, Cd and Pb concentrations decreased to 80.2 ± 2.7 and 159.8 ± 22.1 mg/kg, respectively. The treated biosolids and their products were eligible for land application as an alternative treatment. The microwave-mediated method also lowered the demand for extractants. Using a reduced concentration of sulfuric acid, acetic acid, or EDTA, at least 90% of Cu, 70% of Zn and Pb, 45% of Fe, and 20% of Ni were simultaneously removed from the contaminated biosolids. Cost analyses revealed that the microwave-mediated method could decrease the net total cost of biosolid handling by as much as 62.7%. Considering its simplicity, cost-effectiveness, and minimal environmental impacts, the proposed method offers a promising solution to the problem of heavy metal accumulation in biosolids.

1. Introduction

Biosolids are the final products of sludge produced during sewage treatment in wastewater treatment plants (WWTPs) (Margui et al., 2016). With the continuing rapid growth of human populations and the global trend towards urbanization, an increasingly large amount of sewage and its resulting biosolids are being produced worldwide (Mateo-Sagasta et al., 2015). For example, the WWTPs in China yielded more than 6 million metric tons of dry biosolids in 2013, with a mean annual growth of 13% from 2007 to 2013 (Yang et al., 2015). It has been a serious challenge for many countries to properly dispose the increasing amount of biosolids (Londono et al., 2017), especially considering that conventional disposal methods, such as sanitary landfilling and incineration, are neither environmentally friendly nor sustainable (Abusoglu et al., 2017).

An alternative to unsustainable disposal practices is the post-composting of biosolids for potential use as organic fertilizers in agricultural soils, as they are usually rich in diverse plant nutrients (e.g., N and P) and organic matter (Sharma et al., 2017). With the land application of biosolids, the physical, chemical, and biological properties of the amended soils (e.g., porosity, moisture, pH, cation exchange capacity, humus content, microbial biomass, and enzymatic activities) can be greatly improved (Koutroubas et al., 2014; Lloret et al., 2016). As a result, agricultural production can be enhanced with minimal need for energy-intensive synthetic fertilizers, while the nutrient losses via leaching and surface runoff can be effectively mitigated (Alcantara et al., 2015; Arduini et al., 2018).

Due to its waste-to-resource nature, land application of

* Corresponding author.
E-mail address: sli@cpp.edu (S. Li).

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biosolids has earned popularity in recent years (Sharma et al., 2017). For instance, at least 55% of the biosolids (i.e., 17.8 million metric tons of dry biosolids) generated in the United States are utilized to regain soil fertility and improve soil properties in agricultural lands and forests (Miller-Robbie et al., 2015). However, the commercialization of biosolid-derived fertilizers for agriculture must be performed cautiously, as it is widely known that biosolids often contain toxic metal residues (i.e., heavy metals) (Ashekuzzaman et al., 2019). The species and concentrations of heavy metals in biosolids produced in different WWTPs may differ due to dissimilar chemical compositions of wastewater and different treatment technologies (Westerhoff et al., 2015). Commonly found heavy metals in biosolids include, but are not limited to As, Cd, Cu, Pb, Hg, Mo, Ni, Se, and Zn (Lara-Villa et al., 2011; Muchuweti et al., 2005). Heavy metals in sewage can be from the merging of industrial wastewater, commercial products, and storm water runoff, among other sources (Westerhoff et al., 2015). Besides their uptake by plants, the increased leaching of heavy metal ions into groundwater has also been observed in field studies (Koutroubas et al., 2014; Latare et al., 2014). Depending on the levels of toxicity and dosages of the heavy metals, as well as the land application rate, untreated or inappropriately treated biosolids may be detrimental to plant growth and harm the food web (Koupaie and Eskicioglu, 2015).

The presence of toxic heavy metals in biosolids is the major obstacle to their land application (Margui et al., 2016). It is necessary to extract heavy metals from biosolids and lower their contents below the regulation limits prior to any agricultural usage (Sharma et al., 2017). Most heavy metals can be extracted chemically, physically, and/or biologically (Camargo et al., 2016). Physical methods, such as heat treatment (Shi et al., 2013) and electroremediation (Elicker et al., 2014), are plagued by high costs. Chemical methods employ a variety of metal extractants, including inorganic acids (Gheju et al., 2011), organic acids (Dacera and Babel, 2006), salts (Navarro-Gonzalez et al., 2017), chelating agents (Manouchehri et al., 2006), and mixed reagents (Du et al., 2015). Although the costs of chemical methods are lower than those of physical methods, the risk of secondary pollution due to chemical residues is discouraging (Camargo et al., 2016). Biological methods, such as bioleaching (Wen et al., 2013) and vermicomposting (Azizi et al., 2013), are economically viable and environmentally friendly, but they are demanding in terms of the requisite environmental conditions and may not be suitable for certain high-toxicity metals (Camargo et al., 2016). Although biological methods have gained almost equal prominence in recent years, chemical methods are still the most commonly used and widely studied because of their efficiency and simplicity (Camargo et al., 2016).

In an early study, the performance of heavy metal extraction was explored using two inorganic acids (nitric and hydrochloric), two organic acids (citric and oxalic), and one strong chelator (ethylenediaminetetraacetic acid, i.e., EDTA) (Gheju et al., 2011). It was found that organic reagents had the highest removal efficiencies, followed by EDTA. However, none of the extractants alone was able to effectively remove all species of heavy metals. Hence, the combined use of two or more reagents was suggested to enhance removal efficiencies (Gheju et al., 2011). In a later study, nitrite was added during sludge acidification to enhance metal removal (Du et al., 2015), and a significant improvement in Cu removal from 3–7% to 45–64% and in Zn removal from ~70% to >81% was observed; this was attributed to the disruption of extracellular polymeric substances (EPS) assisted by free nitrous acid (Du et al., 2015). In some recent studies, chemical extraction has also been integrated with physical pretreatment/co-treatment. For example, ultrasound was used to assist heavy metal extraction via citric acid (Wang et al., 2015); although the rate of extraction in this study was accelerated, the efficiency of extraction was minimally affected.

Electromagnetic waves (EWs) are electric and magnetic fields that propagate through space and carry electromagnetic energy (Ishimaru, 2017). As one of the most common types of EWs, microwaves have a frequency in the range of 300 MHz to 300 GHz (Ishimaru, 2017). Significant alteration of the physical and chemical properties of sewage sludge have been observed after its exposure in microwave radiation (Yu et al., 2010). It was found that microwave irradiation was able to disintegrate sludge, destroy microbial cells, and release EPS (Yu et al., 2010). Most recent endeavors have utilized microwave irradiation as a pretreatment technique to remove EPS and heavy metals from sludge before anaerobic digestion (Danesh et al., 2008; Margui et al., 2016; Tyagi and Lo, 2013). When using microwave to enhance contaminant desorption, the primary factor affecting contaminant removal is the affinity of the contaminant for the medium matrix (Gheju et al., 2011; Yuan et al., 2017). However, effective metal removal remains highly dependent on the use of chemical reagents, intensive electromagnetic power, and long contact times (Margui et al., 2016; Tyagi and Lo, 2013). It was noted in a recent study that electromagnetic power attenuation should increase with increased moisture content (Krapivin et al., 2018). From this perspective, if dewatered biosolids are treated instead of high-moisture sludge, electromagnetic energy may be better preserved and more efficiently transmitted to stimulate the release of heavy metals. However, the impact of moisture on the efficacy of microwave-assisted metal extraction has not been investigated previously.

After heavy metal removal, biosolids can be composted with sawdust, wood chips, yard clippings, food wastes, and/or crop residues to make excellent mulch and top soil for horticultural and landscaping purposes (Basta et al., 2015; Lima et al., 2018). Composting can be achieved under either anaerobic or aerobic conditions (Fang et al., 2016). Aerobic composting is considered more efficient and useful than anaerobic composting, although more nutrients are lost from biomass during aerobic composting (Fang et al., 2016). Factors affecting aerobic composting include aeration, moisture, C/N ratios, and temperature (Mejias et al., 2017). High-quality compost with minimal heavy metals can be commercialized to compensate for the costs of treatment and operation. Nevertheless, the overall cost-effectiveness of co-composting biosolids that are treated by microwave-induced heavy metal extraction has not been explored to-date.

The main objectives of this study were to: (1) examine the viability of using minimal microwaves and contact times to efficiently induce the extraction of diverse heavy metals from dewatered biosolids and (2) discuss the economic and environmental feasibility of the proposed microwave-mediated heavy metal extraction. To achieve these goals, we compared the heavy metal removal efficiencies of conventional chemical methods with those using the novel microwave treatment. The treated biosolids were thereafter co-composted with wood chips. A cost analysis of seven different biosolid-handling scenarios was conducted to explore the economic and environmental feasibility of our proposed treatments. Our study addresses a widely applicable but often costly problem that lacks an optimal solution; however, additional pilot studies should be performed in the near future to investigate the practicality of microwave treatments at larger scales.

2. Materials and methods

2.1. Biosolids

The biosolids in this study were collected from the Thomas P. Smith Water Reclamation (TPS) Facility in Tallahassee, FL and the Wastewater Treatment Plant of the City of Graceville (Graceville, FL). The TPS Facility, with a sufficient capacity to treat 100,300 m^3 of wastewater daily, accepts only domestic wastewater from the City of Tallahassee, whereas the WWTP of the City of Graceville also receives industrial wastewater from its surroundings. Biosolids at the TPS Facility were treated by thickening, anaerobic digestion, dewatering, and thermal heat-drying, while biosolids at the WWTP of the City of Graceville were naturally air-dried. Upon sampling, the biosolids were directly sent to the laboratory and stored in a refrigerator at 4 °C before use.

2.2. Biosolid characterization

Sampled biosolids were characterized for their moisture, and total solid and heavy metal contents before further treatment. The moisture content (MC) was determined using the gravimetric method by weighing the sample before and after drying at 105 °C until there was no observable weight change (Li et al., 2019b). The total solid content (TS) was then calculated via mass balance (i.e., TS = 100% − MC). The total volatile solid (TVS) content was estimated based on the weight difference before and after complete combustion at 750 °C (Li et al., 2018). The aqua regia (i.e., the mixture of HNO_3 and HCl in a molar ratio of 1:3) digestion method was used to thoroughly digest the biosolids at 110 °C for 45 min to determine the heavy metal contents (Santoro et al., 2017). After cooling the sample to room temperature (25 °C), the heavy metal contents were quantified using atomic emission spectrometry (4100 MP-AES, Agilent Technologies, Santa Clara, CA) (Nguyen et al., 2015). Triplicate tests were conducted for each sample.

2.3. Heavy metal extraction by acids and EDTA

Heavy metals in the biosolids (5 g) were extracted using 10 mL of 20% (v/v) acetic acid, 20% (v/v) sulfuric acid, or 50 mM EDTA, along with deionized (DI) water extraction, which served as the control. The slurry was contained in 15-mL centrifuge tubes and was well-mixed using a rugged rotator (Glas-Col LLC, Terre Haute, IN) for 24 h at room temperature. These mixtures were subsequently centrifuged at approximately 1600 g for 20 min to separate the liquid and solid phases. The supernatant liquid was collected for the determination of extracted heavy metals using atomic emission spectrometry (4100 MP-AES, Agilent Technologies, Santa Clara, CA) (Nguyen et al., 2015). Triplicate experiments were conducted for each treatment and control.

2.4. Microwave-induced heavy metal extraction

For the pretreatment of biosolids by microwave irradiation, a shallow and wide translucent plastic container containing 5 g of biosolids was placed in the center of a 1-kW 2450-MHz Emerson® household microwave oven (Model MW8107WA, Parsippany, NJ). The thickness of the biosolid layer was approximately 1.5 cm. Microwave irradiation was applied for 10 s to each sample, with a power level of 140 W according to the manufacturer's handbook.

We first explored the impacts of moisture content on the effectiveness of microwave irradiation in inducing heavy metal extraction with 10% (v/v) acetic acid. Two biosolid-to-liquid (i.e., acetic acid) weight ratios, 1:1 and 1:5, were used for comparison. Immediately after microwave treatment, the samples of both biosolid-to-liquid ratios, as well as the controls (no microwave treatment) with a biosolid-to-liquid ratio of 1:5, were centrifuged at approximately 1600 g for 20 min; this was followed by analyses of the heavy metal contents in the supernatant liquids.

After identifying the preferable biosolid-to-liquid ratio for the microwave treatment (= 1:5), this ratio was used for comparing the acid- or EDTA-based heavy metal extraction from biosolids with and without microwave treatment. The flowchart of the microwave-mediated method is shown in Fig. S1 in the Supplementary Materials. In order to balance/reduce the overall costs, when the biosolids were pretreated with microwave irradiation, the concentrations of acids or EDTA used for heavy metal extraction were reduced from 20% (v/v) acids or 50 mM EDTA to 10% (v/v) for acids or 25 mM for EDTA. Given the same volume of liquid extractant, the overall chemical dosages were reduced by half for microwave-induced extraction. Triplicate experiments were conducted for each treatment and control.

2.5. Co-composting of biosolids after microwave-mediated heavy metal extraction

After microwave-mediated heavy metal extraction, biosolids were composted together with shredded wood chips (in 1-cm chunks) under aerobic conditions following the Berkeley method for fast composting (Gautam et al., 2010). The wood-to-biosolid volumetric ratio was between 0.1:1 and 1:1. The moisture content was adjusted to and maintained at 50–60% by supplying distilled water via a sprinkler when the moisture content fell below this range. The mixture was turned for the first time after two days of composting. Afterwards, turnings were repeated every other day. The O_2 consumption and CO_2 emission were continuously monitored using a Micro-Oxymax respirometer (Columbus Instruments, Columbus, OH).

2.6. Cost analysis

The functional unit is fundamental for the comparison of costs among different extraction methods (Li and Chen, 2018). For cost analyses, the management of one metric ton of dewatered biosolids (from the WWTP of the City of Graceville) was assumed. All materials, energy consumption, costs, emissions, and compost recovery were determined in reference to this functional unit. Seven scenarios were considered within the system boundary (Fig. S2). For the scenarios with heavy metal extraction, it was expected that all of the detected heavy metals in the biosolids would meet the pollutant concentration limits of high-quality biosolids for land applications (USEPA, 1994) after the extraction. Under each extraction scenario, the required dosages of acetic acid, sulfuric acid, or EDTA to meet the treatment goals were estimated from the experimental results derived from this study. For the road transport of pretreated biosolids, a distance of 50 km was assumed for both landfilling and composting, and the compost recovered from the treated biosolids would be commercialized for agricultural use, while the composting residues would be disposed in landfills. The prices of raw materials, energy consumption, transportation, and post-treatment, as listed in Table S1, were based on the 2017–2018 U.S. market rates.

2.7. Statistical analysis

Using SPSS v. 14.0 (IBM, Armonk, NY), statistical analyses were conducted to analyze the means and errors of the experimental data. According to the results from the analysis of variance (ANOVA), a significant level of 5% ($p < 0.05$) was used to evaluate the differences between the tested values (Li et al., 2019a).

3. Results and discussion

3.1. Properties of untreated biosolids

The biosolids from the two different WWTPs had similar properties. The values of TS were 25% and 21% for biosolids from the

TPS Facility and the WWTP of the City of Graceville, while those of TVS were 86% and 78%. Both biosolids had a relatively high organic matter content, as reflected by the high ratio of TVS to TS (≥78%). From the gravimetric analyses, it was found that the moisture contents of these biosolids were 75% and 79%, and thus their solid-to-liquid ratios were 1:3 and 1:3.76, respectively.

Different heavy metals were detected in the sampled biosolids (Fig. 1). The species and amounts of heavy metals were dependent on the characteristics of the wastewater received by the two different WWTPs. Because only domestic wastewater was treated in the TPS Facility, relatively fewer species and smaller amounts of heavy metals were found in its biosolids (i.e., Cu (195.2 ± 3.7 mg/kg dry solids), Fe (1586.2 ± 20.5 mg/kg dry solids) and Zn (146.0 ± 1.3 mg/kg dry solids)). Storm water runoff could be the main contributor of these heavy metals to the TPS biosolids, which were likely picked up during a storm event before the storm water joined the sewage (Westerhoff et al., 2015).

More species of heavy metals occurred in larger quantities in the biosolids from the WWTP of the City of Graceville, including Cd (94.3 ± 14.2 mg/kg dry solids), Cu (887.9 ± 9.4 mg/kg dry solids), Fe (12779.3 ± 1452.2 mg/kg dry solids), Ni (139.3 ± 0.4 mg/kg dry solids), Pb (888.7 ± 79.8 mg/kg dry solids), and Zn (698.4 ± 0.3 mg/kg dry solids). Compared to the heavy metal contents in the biosolids from other WWTPs reported in the literature (Mattsson et al., 2017; Yang et al., 2014), the mean contents of Cd (94.3 mg/kg dry solids), Cu (887.9 mg/kg dry solids) and Pb (888.7 mg/kg dry solids) in the biosolids from the WWTP of the City of Graceville were particularly high. For example, the mean Cd content in the biosolids from WWTPs in East China was 7.32 mg/kg dry solids, while those of Cu and Pb from the WWTP of the City of Graceville were 671.87 and 140.47 mg/kg dry solids (Yang et al., 2014). Through this comparison, the urgent need to treat the grievously contaminated biosolids from the WWTP of the City of Graceville was revealed.

The heavy metal contents in the biosolids from both WWTPs were also compared with the “Ceiling Concentrations” and “Pollutant Concentrations” regulated by the United States Environmental Protection Agency (USEPA) (Fig. 1). To qualify for land application, biosolids must at least meet the “Ceiling Concentrations”, while those that meet the “Pollutant Concentrations” can be deemed as “high quality”, and may be applied to lawns and home gardens. As shown in Fig. 1, both the Cd and Pb contents in the biosolids from the WWTP of the City of Graceville (94.3 and 888.7 mg/kg dry solids, respectively) exceeded the ceiling concentration limits (85 and 840 mg/kg dry solids) and were far beyond the pollutant concentration limits (39 and 300 mg/kg dry solids) set for high-quality biosolids (USEPA, 1994). Some of the detected heavy metals are important pollutants. For example, Cd is non-essential and highly toxic as a powerful enzyme inhibitor (Cabral et al., 2015). Even at low concentrations of 1.5–10 mg/L, Cd can penetrate into plant cells and induce oxidative stress due to the overproduction of reactive oxygen species, thus causing severe physiological and genetic damages and ultimately impairing growth and productivity (Marques et al., 2019). Lead also has a wide range of adverse health effects on plants (Kushwaha et al., 2018).

The Cd and Pb that is accumulated in plants can enter human bodies through food intake and dermal contact (Kushwaha et al., 2018; Xue et al., 2017). Cadmium, Pb, and other heavy metals are considered the most hazardous substances in the environment (Cabral et al., 2015). Chronic exposure to these heavy metals may considerably increase the risk of cancer and liver damage, as well as disruption of the endocrine and reproduction systems in humans (Xue et al., 2017). Without these heavy metals being effectively extracted, the direct land application of biosolids is not possible, as high heavy metal contents in untreated biosolids can significantly increase the heavy metal contents in the environment once applied to soils. Considering the exigency of treatment, only the biosolids from the WWTP of the City of Graceville were used for the heavy metal extraction experiments in this study.

3.2. Effectiveness of heavy metal extraction with acid or EDTA

After the extraction of heavy metals using acetic acid, sulfuric acid, or EDTA, the six different species of heavy metals were removed at various levels (Fig. 2). Heavy metal removal was metal-specific; for example, with the use of 20% (v/v) sulfuric acid, significant removal efficiencies of 84.3 ± 3.7% were observed for Cu, 66.7 ± 5.3% for Fe, and 78.2 ± 4.8% for Zn, while the removal efficiencies for Cd (6.7 ± 0.5%), Ni (7.7 ± 0.4%), and Pb (9.4 ± 0.4%) were much less pronounced. The different efficiencies among various heavy metal species were attributed to the dissimilar bonding strengths between the diverse forms of these metals and the matrices of the biosolids (Wang et al., 2015). Additionally, different extractants demonstrated contrasting capacities for extracting heavy metals from biosolids. For instance, when 20% (v/v) of acetic acid was used instead of 20% (v/v) of sulfuric acid, the removal efficiencies of Cu, Fe, and Zn declined drastically to 18.9 ± 0.6%, 15.7 ± 1.1%, and 46.8 ± 3.1%, respectively. These results were consistent with prior studies in which inorganic acids (e.g., sulfuric

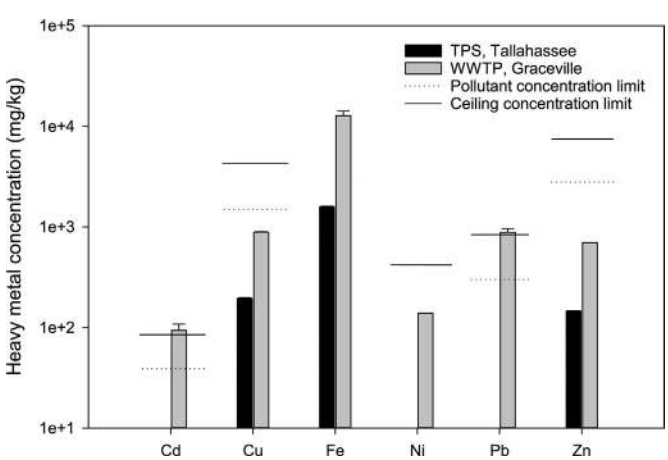

Fig. 1. Concentrations of heavy metals extracted from biosolids collected at the Thomas P. Smith Water Reclamation Facility (TPS, Tallahassee) and Wastewater Treatment Plant of the City of Graceville (WWTP, Graceville).

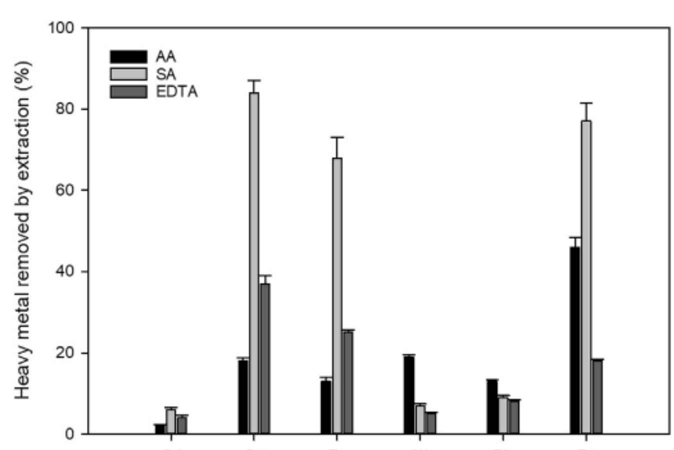

Fig. 2. Percentages of heavy metals removed from biosolids via extraction with acetic acid (AA), sulfuric acid (SA), or ethylenediaminetetraacetic acid (EDTA).

acid) have generally proven more efficient than organic acids (e.g., acetic acid) for heavy metal extraction (Akcil et al., 2015; Suanon et al., 2016). However, the exceptions observed in this study were Ni and Pb, which had better removal efficiencies when acetic acid was used for extraction. This might be due to the different binding interactions between the two metals and the carboxylic acid functional group (Trakal et al., 2016; Yang et al., 2017).

Based on the results from our experiments, the effectiveness of EDTA in extracting heavy metals from biosolids was between that of acetic acid and sulfuric acid for Cd, Cu, and Fe, and was the lowest among the three extractants for Ni, Pb, and Zn. The differences in extraction efficacy with the use of different extractants were likely caused by their dissimilar preferences for mobilizing the metals and forming chelates and complex ions (Cameselle and Pena, 2016). Compared to Cu, Fe, and Zn, which showed better removal efficiencies, only limited amounts of Cd, Ni, and Pb were removed (<20% with acetic acid and <10% with sulfuric acid and EDTA) because of the strong immobilization of these metals in the biosolids (Mahar et al., 2015; Song et al., 2009).

Before treatment of the biosolids by acetic acid, sulfuric acid, or EDTA, the two species of heavy metals that were above the EPA limits for land application were Cd (94.3 ± 14.2 mg/kg dry solids) and Pb (888.7 ± 79.8 mg/kg dry solids). After acetic acid treatment, Cd and Pb contents declined to 91.2 ± 3.5 and 737.9 ± 26.2 mg/kg dry solids; after the sulfuric acid treatment, they were 87.3 ± 4.6 and 804.5 ± 32.1 mg/kg dry solids; and after EDTA treatment, Cd and Pb contents decreased to 89.7 ± 2.9 and 817.9 ± 41.6 mg/kg dry solids. All of the three extractants were able to lower the Pb content slightly below the ceiling concentration limit (840 mg/kg dry solids) but not below the pollutant concentration limit (300 mg/kg dry solids). None of these conventional methods could effectively reduce the Cd content to meet even the ceiling concentration limit (85 mg/kg dry solids), indicating that more potent approaches are required for Cd removal before any land application of the biosolids or their products can be allowed.

3.3. *Influence of moisture content on microwave-mediated heavy metal extraction*

Power loss (or electromagnetic attenuation) often occurs when the EWs travel through media during heavy metal extraction from biosolids (Danesh et al., 2008). The common factors impacting the power loss include medium type, moisture, and frequency of the electromagnetic wave (Krapivin et al., 2018). The power loss during heavy metal extraction in response to the moisture of the biosolids was characterized in this study (Fig. 3). Considering the microwave as a frequency-dependent complex dielectric permittivity (Krapivin et al., 2018), the power loss was calculated with Eq. (1):

$$Power\ Loss = -8.6859\frac{\omega}{c}\left[\frac{\varepsilon'}{2}\left(\sqrt{1+\left(\frac{\sigma}{\varepsilon'\varepsilon_0\omega}\right)^2}-1\right)\right]^{\frac{1}{2}}, \quad (1)$$

where, ω is the angular frequency of the wave ($= 2\pi f$; f is the frequency in cycles per second, or Hz), c is the speed of light (3×10^8 m/s), σ is the electrical conductivity (mho/m or siemens/m), ε' is the real relative complex dielectric permittivity, and ε_0 is the electrical permittivity of free space (8.85×10^{-12} F/m). Power loss gradually increased with the increase in the moisture content of biosolids following a sigmoidal pattern ($R^2 = 0.999$, $p < 0.003$) (Fig. 3), indicating that the actual power loss during heavy metal extraction could be identified as a function of the moisture content of the biosolids using the empirical model based on the experimental results. The simulation also showed that when the moisture

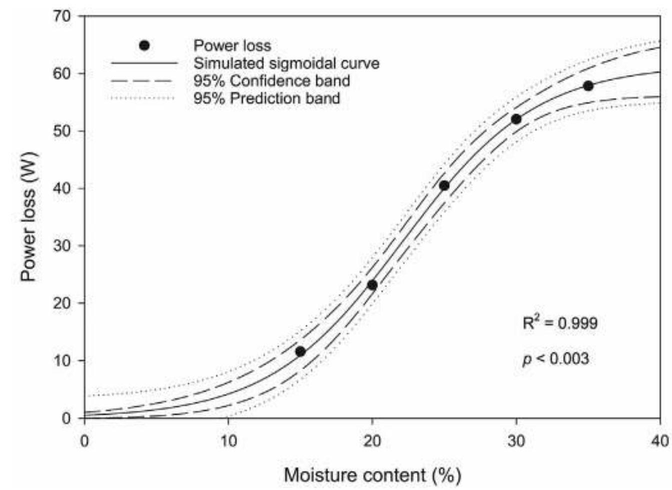

Fig. 3. Nonlinear regression of electromagnetic power loss with biosolid moisture content.

content further increased (>40%), the increase in power loss would be less pronounced and eventually reach a limit near 70 W.

The extraction results after microwave treatments showed that larger amounts of heavy metals were removed from the samples with a biosolid-to-liquid ratio of 1:5 (corresponding to a power loss of ~70 W) than those with a ratio of 1:1 (corresponding to a power loss of ~60 W) (Fig. 4). These results are surprising because they suggest that more heavy metals were removed when more electromagnetic energy was attenuated. We hypothesize that a higher moisture content could facilitate the penetration of liquid extractants through the porous inner structures of biosolids, and thus the released heavy metals due to the microwave mediation could be immediately extracted by the solution. Between the two groups of samples with the same biosolid-to-liquid ratio (i.e., 1:5), it was obvious that microwave mediation significantly improved the removal of all of the investigated heavy metals (Fig. 4). The microwave-induced improvement was particularly pronounced with Cd and Pb, of which the removal efficiencies were increased by more than 5 times. After the microwave-mediated treatment of the biosolids using 10% (v/v) acetic acid (biosolid-to-liquid ratio of 1:5), the resulting Cd content decreased to 80.2 ± 2.7 mg/kg dry solids, while the Pb content was remarkably reduced to 159.8 ± 22.1 mg/kg dry solids. The Cd content was below the ceiling concentration

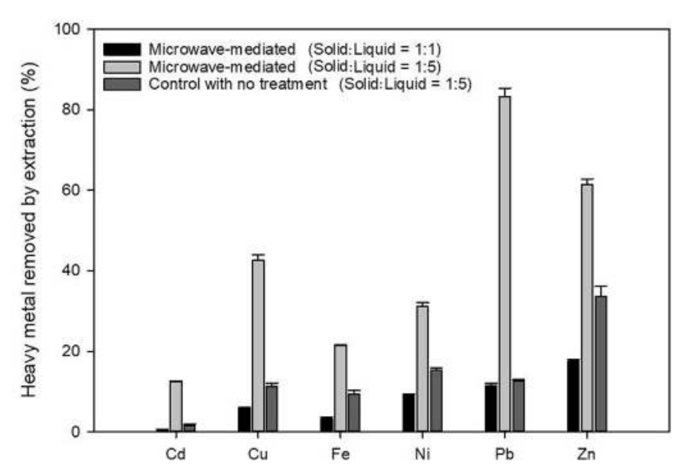

Fig. 4. Percentages of heavy metals removed from biosolids by microwave-mediated heavy metal extraction with different solid-to-liquid weight ratios using 10% (v/v) acetic acid as an extractant.

limit (but above the pollutant concentration limit), and the Pb content met both the ceiling and pollutant concentration limits, implying that it may be possible to use these biosolids for agricultural and other land purposes after microwave-induced heavy metal extraction.

3.4. Improvement of heavy metal extraction with microwave mediation

The pretreatment using microwaves considerably enhanced the removal of heavy metals from the biosolids, in comparison with the removal using the conventional extraction methods (i.e., acids or chelating agents) (Fig. 5). For all of the investigated heavy metals, the improvement with microwave mediation was observed even when a reduced concentration (either 10% (v/v) for acids or 25 mM for EDTA) was used for extraction, indicating that this method could reduce the net cost of treatment. The increase in heavy metal removal efficiency was the most remarkable when microwaves were used to induce extraction by EDTA and the least significant when sulfuric acid was used. For example, the removal efficiency of Zn increased by nearly 300% with EDTA treatment, 40% with acetic acid treatment, and 14% with sulfuric acid treatment. Each EDTA molecule has four carboxylic acid groups and two amine groups with lone pair electrons. Therefore, a larger number of the additional heavy metals released as a result of microwave irradiation were likely able to bind to EDTA more readily than to the acids. However, while greatly improved, the removal efficiencies of Cd (<20%) and Ni (<40%) were still not satisfactory; this may be attributed to the inherent natures of these metals in bonding to the biosolids.

After microwave-mediated treatment, the heavy metals in the biosolids were all decreased to below their ceiling concentration limits and some were lowered to below their pollutant concentration limits (i.e., they became high quality). For example, there was a remarkable removal efficiency of 82.2% of the Pb content after microwave-mediated acetic acid treatment, and the content was drastically decreased to 159.8 mg/kg dry solids, which was much lower than its regulated pollutant concentration limit of 300 mg/kg dry solids. The significantly reduced heavy metal contents in the biosolids indicated the possibility for high-quality land application.

3.5. Composting of microwave-treated biosolids

The carbon-to-nitrogen (C/N) ratio of composting materials is the key factor for the decomposition of organic matter in compost piles (Molla et al., 2005). Carbon is the basic energy source and building block of life, whereas N is necessary for the syntheses of important molecules, such as proteins, genes, and cellular structures. The decomposition of composting materials can be greatly improved with a C/N ratio ranging from 25 to 30 by balancing the carbonaceous materials (e.g., wood chips) and the nitrogen-rich materials (e.g., biosolids) (Molla et al., 2005). A higher C/N ratio will result in slower decomposition, while a lower C/N ratio will lead to ammonia (NH_3) emissions, which may cause eutrophication in freshwater and marine systems (Agyarko-Mintah et al., 2017). The aerobic composting of biosolids consumes O_2 to oxidize/degrade more recalcitrant organic compounds into low molecular-weight, readily biodegradable organics, while producing CO_2 and water (Mejias et al., 2017). Therefore, the rates and amounts of O_2 consumption and CO_2 emissions during composting can be used to reflect the suitability of the materials for composting.

Under a 30-day composting of the mixtures of wood chips and biosolids, different rates and accumulative amounts of O_2 and CO_2 were observed when different wood-to-biosolid volumetric ratios were prepared (Fig. 6). By simulating the experimental results using a nonlinear least-square regression method (Li et al., 2019a), it was found that the optimal wood-to-biosolid ratio (v/v) should be between 0.6 and 0.8. Therefore, after extracting the heavy metals from the biosolids via the microwave-mediated treatments to meet the regulation limits, for yard wastes, such as leaves, grasses, and branches, a wood-to-biosolid ratio of 0.6–0.8 can be recommended for the preparation of composting materials for land application. The extracted heavy metals in the liquid extractants can be recovered and reused in industrial setting through a tanning process of chemical precipitation and electrocoagulation (Mella et al., 2015). Depending on the diverse approaches, the recovery and reuse of heavy metals from their solutions may further reduce the net cost of biosolid handling (Camargo et al., 2016).

3.6. Cost breakdown of biosolid management

For cost analysis, the management of dewatered biosolids was divided into four processes (i.e., heavy metal extraction, road transport, composting, and landfilling) (Fig. 7). Among these processes, landfilling was found to be the costliest, especially if all of the biosolids were disposed of in landfills without heavy metal extraction. It should be noted that even if the biosolids (without heavy metal extraction) were not used for composting, 111.92 kWh of electricity per functional unit was able to be recovered from

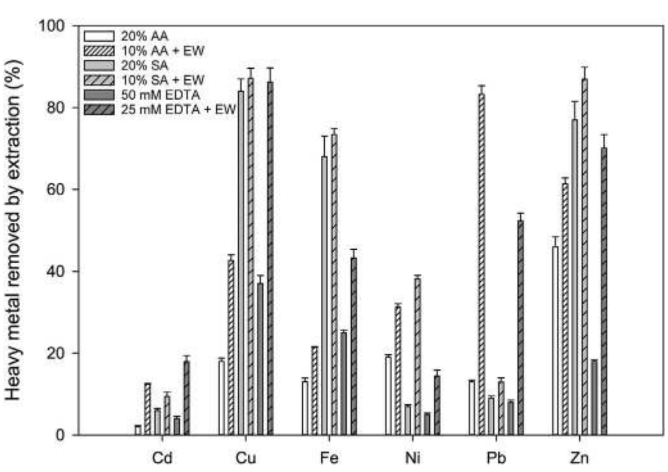

Fig. 5. Comparison of the percentages of heavy metals removed from biosolids using different extraction methods with and without microwave treatment. AA: acetic acid; SA: sulfuric acid; EDTA: ethylenediaminetetraacetic acid; EW: microwave irradiation.

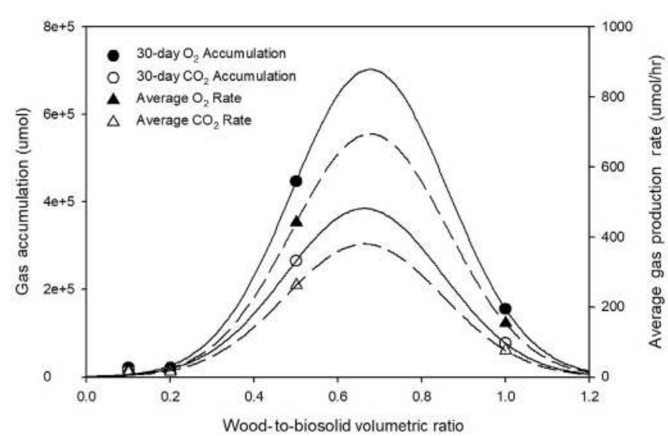

Fig. 6. O_2 consumption and CO_2 emissions during 30 days of composting with different wood-to-biosolid volumetric ratios.

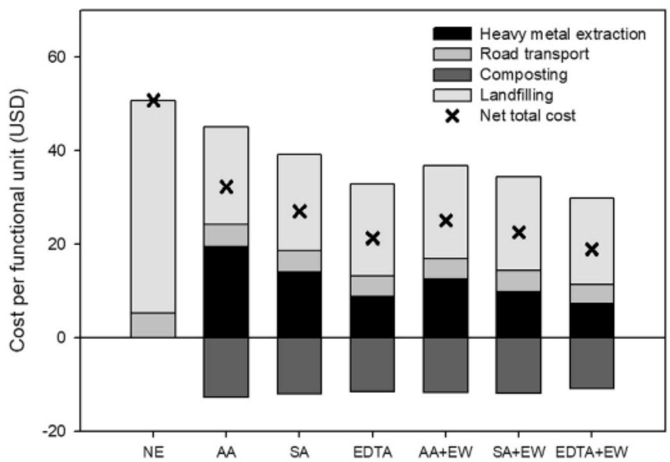

Fig. 7. Cost per functional unit for seven biosolid management scenarios: NE, no extraction of heavy metal; AA, extraction by acetic acid; SA, extraction by sulfuric acid; EDTA, extraction by EDTA; AA + EW, extraction by electromagnetic wave treatment and acetic acid; SA + EW, extraction by electromagnetic wave treatment and sulfuric acid; EDTA + EW, extraction by electromagnetic wave treatment and EDTA.

landfills via the emissions of CH_4, which could also compensate for the other costs, such as composts. Nevertheless, direct disposal of all the untreated biosolids (i.e., NE scenario) would not save on expenses. On the contrary, landfilling the biosolids without heavy metal extraction resulted in the highest net total cost (50.74 USD) among the seven scenarios investigated. In the other scenarios, after heavy metal removal using one of the six different extraction methods, the costs of the composts that could be recovered from the treated biosolids ranged from 10.92 to 12.81 USD per functional unit, depending on the different treatment efficiencies.

Cost analysis indicated that 10 s of microwave irradiation was able to greatly lower the net total cost by as much as 62.7% (comparing to the NE scenario shown in Fig. 7). The most economic approach for managing the biosolids was found to be the combination of microwave irradiation with EDTA treatment (i.e., the EDTA + EW scenario), deriving a considerably reduced net total cost of 18.91 USD per functional unit. These results suggest that microwave-mediated EDTA treatment could be significantly more cost-effective for heavy metal extraction and improve the overall management of dewatered biosolids.

3.7. Implications and future directions

Although many different chemical methods, based on either inorganic/organic acids and/or chelating agents, have been developed to extract heavy metals from biosolids for various land applications, the challenges in terms of sustainability and economic viability remain unresolved (Mattsson et al., 2017). The addition of chemicals (e.g., sulfuric acid, acidic acid, and EDTA) are usually unfavorable to the environment and typically expensive, especially when many tons of biosolids are to be treated. The composting products of biosolids treated by inorganic/organic acid-extraction methods tend to be acidic due to the residual acid adsorbed by the biosolids (Stylianou et al., 2007). These composts are inappropriate for soil liming, which is closely related to nutrient availability and infiltration (Manna et al., 2007). Additionally, the excessive use of EDTA for heavy metal extraction will not yield ideal compost, as EDTA salts are toxic to most plants (Krujatz et al., 2011).

The results of this study demonstrate the potential of using microwave irradiation to reduce the required amount of acid or EDTA, while maintaining high removal efficiencies of heavy metals from biosolids. With less acid or EDTA being used, negative environmental effects can be consequently minimized and more eco-friendly composts can be produced. Our cost analyses show that microwave-induced heavy metal removal should be significantly more cost-effective when compared to conventional methods because of the economic benefits of composting biosolids. A comprehensive life-cycle cost analysis (LCCA) assuming different combinations of scenarios should be conducted in the future to determine the most cost-effective combination of microwave exposure and chemical dosage.

The results of this study also reveal that electromagnetic attenuation gradually increases with the increase of moisture content, indicating that the effectiveness of microwave irradiation can be considerably impacted by the moisture of biosolids. While it is desirable to lower the moisture of biosolids so that less electromagnetic energy is attenuated, the dewatering of biosolids, which can be either time-consuming (if air-drying) or energy-consuming (if drying with machinery), should be kept to a minimum. Meanwhile, an appropriate moisture content could also allow the released heavy metals to be immediately dissolved by the extractant, thus benefiting metal extraction with microwave treatment. Future studies may focus on identifying the optimal moisture content of biosolids to enhance the cost-effectiveness of microwave irradiation for heavy metal immobilization.

4. Conclusions

Microwave irradiation was used in this study to induce the immobilization of heavy metals from dewatered biosolids and to reduce the use of chemicals for metal extraction. Compared to conventional methods, which solely rely on the use of acetic acid, sulfuric acid, or EDTA, microwave irradiation for a short contact time of 10 s was able to stimulate metal extraction and result in a threefold increase in the removal efficiencies for certain heavy metals. All of the heavy metals detected in the biosolids were lowered to below the concentration limits regulated by the US-EPA, which allows for the land application of biosolids to recover their nutrients and energy. Composting was conducted using the treated biosolids, together with wood chips, to produce compost and examine its commercial potential. A wood-to-biosolid ratio of 0.6−0.8 was found to be optimal for the production of high-quality compost. The cost analysis, comparing seven different biosolid handling scenarios, showed that microwave irradiation could drastically decrease the net total cost of biosolid management by as much as 62.7%, implying that it may be a reliable solution to heavy metal contamination in biosolids due to its simplicity and cost-effectiveness. Future studies may focus on improving the microwave treatment with an optimal moisture content and new extractants. Pilot studies should be encouraged to explore the efficacy of this novel approach at larger scales. Process control and plant optimization studies should also be conducted for more feasible full-scale applications.

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Declaration of interest

The authors declare no conflict of interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2019.118342.

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