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Modified bentonite as a conditioning agent for stabilising heavy metals and retaining nutrients in sewage sludge for agricultural uses

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ABSTRACT

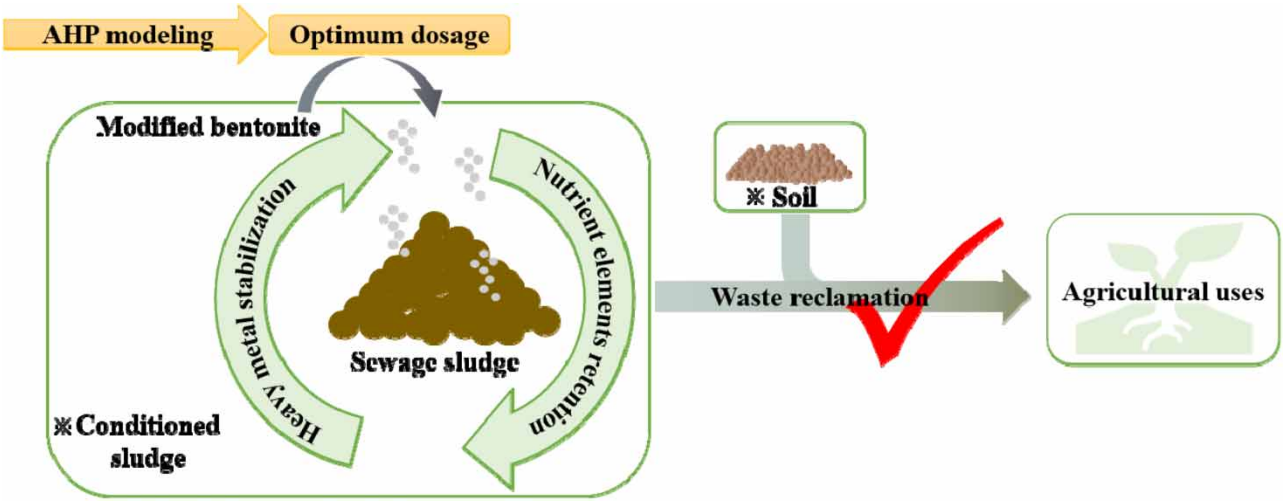
The management and disposal of excess sludge are emerging issues owing to the high costs associated with treatment. In this study, the viability of a modified bentonite was investigated as a conditioning agent for the stabilisation of heavy metals (i.e., Cu, Zn, Cr, Pb, and Cd) and the retention of nutrient species (i.e., total nitrogen (TN), total phosphorus (TP), available nitrogen (available N), and Olsen-phosphorus (Olsen-P)) in sewage sludge for agricultural use. Five grams of modified bentonite resulted in the highest stabilisation rate of heavy metals and strongly contributed to the stabilisation of heavy metals. However, increased amounts of modified bentonite might increase the TN, available N, and TP losses in the conditioned sewage sludge. Through the analytic hierarchy process modelling, optimal concentrations of nutrient species and heavy metals remaining in the conditioned sewage sludge were achieved when the ratio of bentonite to sewage sludge was 1:12.5 (4 g bentonite : 50 g sludge). Moreover, the optimal mixing ratio of the conditioned sewage sludge to the soil (1:2) was suggested for agricultural use. Based on these observations, modified bentonite allowed the sewage sludge to be used as a fertiliser in agriculture by stabilising heavy metals and retaining nutrient species.

Key words: analytic hierarchy process modelling, heavy metals, modified bentonite, nutrients, sewage sludge

HIGHLIGHTS

- The modified bentonite stabilised the heavy metals and retained the nutrient elements in the sewage sludge.
- The AHP modelling revealed the optimal ratio of bentonite to sewage sludge (4 g bentonite : 50 g sludge).
- The optimal mixing ratio of the conditioned sewage sludge to the soil (1:2) was suggested for agricultural use.
- The modified bentonite allowed sewage sludge to be used as a fertiliser in agriculture.

GRAPHICAL ABSTRACT



INTRODUCTION

The excessive production of sewage sludge is the main obstacle to biological wastewater treatment processes worldwide; this is because its treatment, disposal, and management procedures account for approximately 20–50% of the total operating cost of conventional wastewater treatment plants (Karr & Keinath 1978). Adequate sewage sludge treatment, disposal, and management techniques have thus become increasingly important in the fields of environmental science and engineering (Zhu *et al.* 2013). Land application is considered to be one of the most effective techniques for the final disposal of sewage sludge as high concentrations of organic materials and nutrients (i.e., nitrogen and phosphorus) may enable improved agricultural productivity (Insam *et al.* 2015). Although land application combines the disposal of sewage sludge and the reutilization of nutrients, the accumulation of toxic substances (e.g., heavy metals) through adsorption, complexation, precipitation, and solubilisation in soils may pose serious risks to crops and human health (Zhu *et al.* 2013). Therefore, the development of a treatment method that can remove and/or stabilise toxic heavy metals while retaining nutrients for the land application of sewage sludge is urgently needed.

Over the past decades, several methods, including biological treatments (Pathak *et al.* 2009), chemical treatments (Stylianou *et al.* 2007), electrochemical treatments (Hanay *et al.* 2009), ion-exchange treatments (Dąbrowski *et al.* 2004), membrane treatment (Chaudry *et al.* 1998), and thermal treatments (Zorpas *et al.* 2001), have been widely used for the treatment and disposal of sewage sludge. However, biological treatments have been regarded as the preferable approach for selectively removing heavy metals from sewage sludge due to their effectiveness and economic feasibility. Many studies have reported that bioleaching processes and aerobic and anaerobic composting processes can significantly change the chemical forms, concentrations, and bioavailability of heavy metals in sewage sludge (Amir *et al.* 2005; Liu *et al.* 2007; Zhu *et al.* 2013). However, biological treatments (i.e., bioleaching, aerobic, and anaerobic composting processes) require a longer period of operation and inevitable energy consumption, compared to physical and/or chemical treatments, including membrane separation, chlorination, and thermal treatments, which limit their practical applications for the treatment and disposal of sewage sludge (Chaudry *et al.* 1998; Zorpas *et al.* 2001; Fraissler *et al.* 2009; Zhu *et al.* 2013). Chemical treatments (i.e., chemical extraction and chemical precipitation) have several advantages, including a shorter reaction time, easy operating procedures, and higher removal efficiencies for heavy metals, compared to biological treatments (i.e., bioleaching, aerobic and anaerobic composting processes) (Babel & del Mundo Dacera 2006; Ren *et al.* 2015). Various types of chelating reagents (e.g., ethylenediaminetetraacetic acid [EDTA]), organic acids (e.g., citric acid), and inorganic acids (e.g., hydrofluoric acid) are available for the removal and stabilisation of heavy metals from sewage sludge (Amir *et al.* 2005; Babel & del Mundo Dacera 2006; Ren *et al.* 2015). Sequential chemical extraction methods, which utilise a series of chemical agents in the sequence of extractants with increasing strength, were found to be effective at removing a wide range of heavy metals compared to the single use of each chemical extractant (Amir *et al.* 2005; Babel & del Mundo Dacera 2006). Some researchers have attempted to combine the Fenton-like reaction with a bioleaching process to shorten the bioleaching period (Zhu *et al.* 2013; Fontmorin & Sillanpää 2015). Despite the high removal efficiencies of heavy metals from sewage sludge, chemical treatments require a large amount of chemicals to control the pH of the sewage sludge, acid-corrosion-resistant apparatus, and additional treatments for hazardous wastes, which could lead to increases in operating costs (Babel & del Mundo Dacera 2006).

In recent years, natural minerals have received remarkable attention as eco-friendly adsorbents for the effective removal and stabilisation of heavy metals from sewage sludge and wastewater owing to their high ion-exchangeable capability and environmental safety (Sprynskyy *et al.* 2007). Natural zeolites might account for a considerable fraction of heavy metals from sewage sludge during the composting process (Cd = 100%, Cu > 28%, Cr > 10%; Fe > 41%, Mn > 9%, Ni > 50%, Pb > 50%, Zn > 46%) (Zorpas *et al.* 2000). In addition, bentonite clays can remove approximately 93% of Cr (III) from tannery wastewater (Tahir & Naseem 2007). Meanwhile, bentonite has a positive effect on the biochemical properties of soil (e.g., concentration of TN and TP) during long-term soil amendment (Mi *et al.* 2021). Although previous studies have demonstrated that natural materials (e.g., bentonite and zeolite) can play a critical role in the stabilisation and removal of heavy metals from sewage sludge and wastewater (Tahir & Naseem 2007), no comprehensive research has been conducted on both the retention of nutrients (i.e., nitrogen and phosphorus) and stabilisation of heavy metals through sewage sludge treatment using bentonite for agricultural reuse.

The main objective of this study was to evaluate the feasibility of using modified bentonite as a conditioning agent to treat sewage sludge for agricultural reutilization. Therefore, we investigated the effects of modified bentonite concentrations on the stabilisation of heavy metals and retention of nutrients to assess the criteria of the modified bentonite treatment. Moreover, the optimal conditions of the modified bentonite treatment to maximise the stabilisation of heavy metals and minimise the loss of nutrients were determined using an analytic hierarchy process (AHP) model to promote the agricultural use of sewage sludge.

MATERIALS AND METHODS

Preparation of modified bentonite

The organic bentonites purchased from a relevant company (Beijing, China) were activated with an organic modifier (hexadecyl trimethylammonium bromide; HDTMA) from the Na-bentonite sample, according to the organic activation method (wet process) proposed by Li *et al.* (2016). Briefly, 2% (w/v) Na-bentonite sample and 0.5% (w/v) HDTMA sample were mixed for 3 h at 60 °C to improve the physicochemical properties of bentonites (i.e., interlayer spaces, specific surface area, and hydrophobicity) that are closely related to the adsorption capacity of heavy metals and nutrients. The maximum heavy metal content is less than 0.001% (Li *et al.* 2021).

Collection of sewage sludge and soil samples

Sewage sludge was obtained after two-stage anaerobic sludge digestion at a wastewater treatment plant in Beijing, China. The sewage sludge was air-dried at room temperature (~20 °C), ground with a grinding machine, and then screened through a 100-mesh stainless steel sieve. To collect unpolluted soils, adequate amounts of soil were sampled using an auger from the topsoil layer (depth = 0–20 cm) in Yangling district (Xianyang, Shaanxi, China), air-dried at room temperature (~20 °C), and then filtered with a 100-mesh nylon filter (NLSJ-100, Shanghai Bio Biotechnology, Shanghai, China) to remove stones, animals, and plant residues. The sewage sludge and soils used in this study were the same as those used in previous studies in our laboratory, and the basic characteristics of sewage sludge and soil are presented in the Supplementary Information, SI.1 (Ren *et al.* 2015).

After the pre-treatment procedures, the physicochemical properties of the sewage sludge and soil samples, such as pH, relative humidity, total alkalinity, organic matter (OM), total nitrogen (TN), total phosphorus (TP), available nitrogen (available N), Olsen-P, total suspended solids (TSS), volatile suspended solids (VSS), and soluble chemical oxygen demand (SCOD), were immediately measured using relevant methods to identify their effects on the stabilisation of heavy metals, and the retention of nutrients (Ministry of Housing and Urban-Rural Development of PRC 2005; Ren *et al.* 2015; Kou *et al.* 2020). All samples of sewage sludge, soil, and their mixtures were digested with a mixture of HCl/HNO_3 (Ren *et al.* 2015). The concentrations of heavy metals (i.e., Cr, Cu, Cd, Zn, and Pb) in the mixture of the sewage sludge and soil samples were measured using atomic absorption spectroscopy (AAS, Perkin Elmer Analyst 300). Prior to further analyses and experiments, pre-treated sewage sludge and soil samples were stored in 50 mL polyethylene containers at 4 °C in a refrigerator.

Sewage sludge treatments using modified bentonite

The modified bentonite was applied as a conditioning agent to determine its effect on the stabilisation of heavy metals (i.e., Cu, Zn, Cr, Pb, and Cd) and the retention of nutrients (i.e., nitrogen and phosphorus) in the sewage sludge, which are known to be most frequently found in raw sewage sludge samples (Ren *et al.* 2015). To determine the optimal dose, the sewage sludge (weight of each sample = 50 g) was treated using five different doses of modified bentonite (1, 2, 3, 4, and 5 g). Each mixed sample was screened through a 100-mesh stainless-steel sieve. After the addition of 30 mL of deionised water, each mixture was fully mixed for one week and then maintained for three weeks.

Mixing of the sewage sludge and soil samples

The conditioned sewage sludge with the modified bentonites was mixed with the soils at ratios of 1:1, 1:2, and 1:4 (g/g) at ambient temperature (25 ± 2 °C) for one week at a field moisture capacity of 70%, and then dried at 103 °C for 24 h. After the extraction procedures, the concentrations of heavy metals (extracted by leaching liquid) and nutrients (i.e., nitrogen and phosphorus) were identified using various analytical methods to verify the viability of the conditioned sludge as fertilisers for agricultural uses.

Stabilisation of heavy metals

According to the method proposed by the China EPA (2007), the leaching solution was acetic acid solution (1.72% (v/v) acetic acid). The conditional sludge (5 g) mixed with 100 mL leaching solution was added to a 500 mL polyethylene bottle at a solid-liquid ratio of 1:20. Thereafter, the samples were agitated for 18 h (rpm = 30 ± 2, temperature = 23 ± 2 °C). The supernatants of each sample were obtained after centrifugation, and the concentrations of heavy metals (i.e., Cu, Cr, Pb, Zn, and Cd) were measured by AAS. The stabilisation rate of the heavy metals was calculated as follows:

$$\text{Stabilisation rate (\%)} = \left(1 - \frac{C_L \cdot V_L}{C_S \cdot M_S}\right) \times 100\% \quad (1)$$

where C_L is the concentration of heavy metals in the leachate (mg/L), V_L is the volume of added leaching liquor (L), C_S is the concentration of heavy metals in the sewage sludge (mg/kg), and M_S is the mass of the sewage sludge sample (kg) (Yu *et al.* 2017).

Chemical speciation of heavy metals

The chemical speciation of heavy metals in the raw and conditioned sewage sludge samples was identified using the sequential extraction method (i.e., the modified BCR-sequential extraction procedure) proposed by Pan *et al.* (2009). The chemical forms of Cr, Cu, Cd, Zn, and Pb extracted from the raw and conditioned sewage sludge samples were divided into four fractions: (a) acid-exchangeable, (b) reducible, (c) oxidised, and (d) residual fractions (Kou *et al.* 2020). The concentrations of heavy metals in the sequentially extracted samples from the raw and conditioned sewage sludge were also determined using AAS.

Nutrient determination

The concentrations of TN, TP, available N, and Olsen P were monitored in five sludge samples with different amounts of modified bentonite (from 1 to 5 g). The concentration of TN was detected using an ultraviolet-visible spectrophotometer (220/275 nm) from each 0.01 g sample by treatment with 50 mL of alkaline potassium persulfate solution and 10 mL (1 + 9) HCl, and the concentration of TP was determined using an ultraviolet-visible spectrophotometer (700 nm) from each 0.1 g sample by pretreatment with 50 mL of alkaline potassium persulfate solution. The available-N concentration was determined by the method of alkali N-proliferation, and Olsen-P was extracted using phosphorus-free activated carbon and 0.5 mol/L $NaHCO_3$ with mixing at 25 °C for 30 min at 160 rpm. The measurement method for each parameter, including raw sewage sludge and soil, was based on the processes of Kou *et al.* (2020).

AHP modelling

In this study, the best bentonite amount was selected based on the stabilisation rates of heavy metals and the retention of nutrient elements in the sludge. AHP modelling was used to evaluate the efficiency of the sludge treated with different amounts of bentonite. The following four steps were applied to establish the AHP model: (i) establishment of the evaluation index system, (ii) determination of the weights by AHP, (iii) selection of the membership function with fuzzification, and

Table 1 | Criteria of hierarchical division for evaluating capacity of sludge treatment

Goal layer	Factor layer	Sub-factor layer	Unit
Sludge treatment ability evaluation	Heavy metals	Cu stabilization rate	%
		Cd stabilization rate	%
		Pb stabilization rate	%
		Zn stabilization rate	%
		Cr stabilization rate	%
	Nutrient elements	TN retention	g/kg
		TP retention	g/kg
		Available-N retention	mg/kg
		Olsen-P retention	mg/kg

(iv) comprehensive evaluation. Two main factors and nine sub-factors were selected to comprehensively elucidate the optimal dose of the bentonites to maximise the stabilisation of heavy metals and minimise the loss of nutrients (Table 1).

The subordinate function was used to determine the single-factor evaluation matrix with fuzzification, which attempted to define the probability of basic events and the uncertainty of calculations (Li *et al.* 2020b). Corresponding to different practical problems, there are different methods for determining subordinate functions. The stabilisation rates of heavy metals (Cu, Cd, Pb, Zn, and Cr) in the sludge treated with bentonite were assigned five grades: 'very good' (more than 80%), 'good' (60–80%), 'general' (40–60%), 'poor' (20–40%), and 'very poor' (less than 20%), respectively (SI. 2).

RESULTS AND DISCUSSION

Properties of heavy metals and nutrients in sewage sludge and soil

Combined with the results of previous studies, the physicochemical properties of sewage sludge and soil samples are summarised in Table 2. The pH of the sewage sludge (pH = 6.2–8.2) did not significantly differ from that of the soils (pH = 5.1–7.7) whereas a markedly greater proportion of nutrients was found in the sewage sludge (TN = 24.0–34.4 g/kg; TP = 19.5–48.0 g/kg; available-N = 394.9–2,390.0 mg/kg; Olsen-P = 260.2–453.5 mg/kg) than the soil (TN = 0.4–1.7 g/kg; TP = 0.4–6.0 g/kg; available-N = 35.0–590.0 mg/kg; Olsen-P = 6.2–70.1 mg/kg). These results imply that sewage sludge has great potential for improving soil fertility. Although nutrients (i.e., TN and TP) exist in sewage sludge, they cannot be used directly as the concentration of heavy metals in sewage sludge is markedly higher than that in the soil. The concentrations of Cd, Pb, Cu, Zn, and Cr in the sewage sludge satisfied the limits for heavy metals in the sludge established by the European Union (EU) Directive 86/278/European Communities Commission (ECC). However, the levels of some heavy metals (e.g., Cu and Zn) significantly exceeded the limits for soils suggested by EU Directive 86/278/ECC (ECC 1986), enabling the utilisation of sewage sludge to enhance agricultural productivity. Therefore, an adequate pre-treatment step for extracting or stabilising heavy metals from sewage sludge is necessary to prevent the accumulation of heavy metals in agricultural soils.

Effect of modified bentonite on the stabilisation of heavy metals

The stabilisation rates of heavy metals from sewage sludge (sample weight = 50 g) as a function of the modified bentonite amount (1–5 g) are shown in Figure 1. The stabilisation rates of the heavy metals (i.e., Cu, Cd, Pb, Zn, and Cr) increased gradually with increasing amounts of modified bentonite. However, the degree of increase varied considerably depending on the range of the applied amounts of modified bentonite. The stabilisation rates of heavy metals (i.e., Cu, Cd, Pb, Zn, and Cr) increased rapidly in the range of 1–4 g of modified bentonite, and their stabilisation rates were slightly increased when the modified bentonite amount exceeded 4 g. Among the selected heavy metals, Cu and Pb were the most effectively stabilised from the sewage sludge using 5 g of modified bentonite (the highest stabilisation rate of Cu = 90.1%; the highest stabilisation rate of Pb = 91.2%). The highest stabilisation rates of Cd and Cr were achieved with 5 g of modified bentonite (Cd = 73.2%; Cr = 80.3%). In addition, approximately 62.8% of Zn could be stabilised with 5 g of modified bentonite. These observations indicate that the modified bentonite as a conditioning agent is effective for promoting the stabilisation of heavy metals from sewage sludge. The stabilisation of heavy metals after conditioning of sewage sludge can be explained by the dominant component (montmorillonite) in modified bentonite. Montmorillonite can adsorb heavy metals through cation exchange reactions in the interlayers caused by the electrostatic interaction between the positively charged metals

Table 2 | Physicochemical properties of the sewage sludge and soil samples

Parameters	Sewage sludge	Soil	Limits for heavy metals[a]		References
			In sludge	In soil	
pH	6.2[5] – 8.2[10]	5.1[9] – 7.7[10]	–	–	[1,2,4–10]
TN (g/kg)	24.0[6] – 34.4[5]	0.4[10] – 1.7[2]	–	–	[1,2,5,6,8–10]
TP (g/kg)	19.5[5] – 48.0[6]	0.4[9] – 6.0[10]	–	–	[1,2,4–6,9,10]
Available-N (mg/kg)	394.9[1] – 2,390.0[10]	35.0[7] – 590.0[10]	–	–	[1,6,7,9,10]
Olsen-P (mg/kg)	260.2[6] – 453.5[1]	6.2[10] – 70.1[7]	–	–	[1,6,7,9,10]
Cu (mg/kg)	65.9[4] – 1,735.4[1]	0.01[9] – 34.8[2]	1,000–1,750	50–140	[1–4,6,8–10]
Cd (mg/kg)	3.3[1] – 28.0[3]	0.05[9] – 2.3[10]	20–40	1–3	[1–4,6,8–10]
Pb (mg/kg)	48.5[4] – 469.2[10]	13.5[8] – 42.4[10]	750–1,200	50–300	[1,3–6,8–10]
Zn (mg/kg)	413.3[3] – 2,885.0[10]	23.5[10] – 62.1[8]	2,500–4,000	150–300	[1,3–6,8–10]
Cr (mg/kg)	69.3[4] – 676.3[3]	6.2[10] – 9.3[1]	–	–	[1,3,4,8–10]

[a]Council Directive 86/278/EEC (ECC 1986).

[1] Ren *et al.* (2015); [2] Li *et al.* (2015); [3] Chen *et al.* (2015); [4] Wang *et al.* (2018); [5] Ye *et al.* (2017); [6] Kou *et al.* (2019); [7] Pan *et al.* (2019); [8] Li *et al.* (2020a); [9] Dong *et al.* (2020); [10] Kou *et al.* (2020).

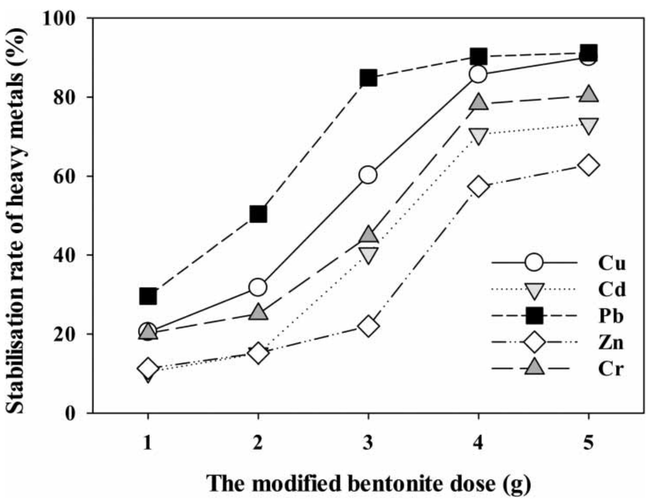


Figure 1 | Stabilization rate of the heavy metals from the sewage sludge (weight of each sample = 50 g) as a function of the modified bentonite dose (1–5 g).

and the negatively charged mineral surfaces and the formation of inner-sphere complexes induced by Si–O− and Al–O− functional groups at the edges of minerals (Kraepiel *et al.* 1999).

To further analyse the degree of stabilisation of heavy metals in sewage sludge treated with modified bentonite, the speciation of heavy metals in the sewage sludge (sample weight = 50 g) before and after modified bentonite treatment (dose = 3 g) is illustrated in Figure 2. Prior to treatment with modified bentonite (Figure 2(a)), some of the heavy metals in the sewage sludge (i.e., Cu, Pb, and Cr) predominantly consisted of oxidisable (Cu = 21.6%; Pb = 3.6%; Cr = 38.2%), reducible (Cu = 17.6%; Pb = 37.6%; Cr = 18.4%), and exchangeable fractions (Cu = 37.5%; Pb = 51.7%; Cr = 16.8%), which are relatively unstable compared to the residual fractions (Cu = 23.3%, Pb = 7.1%, Cr = 26.6%). Although the residual fractions were found to be major components of total Cd (58.6%) and total Zn (61.2%), they contained considerable amounts of oxidisable (Cd = 34.7%; *Zn* = 16.5%), reducible (Cd = 4.6%; Zn = 9.6%), and exchangeable fractions (Cd = 2.1%; Zn = 12.7%). The oxidisable fractions are indicative of the capacity to bind to organic materials, while the exchangeable and reducible fractions are closely associated with the mobility, absorptivity, and bioavailability of heavy metals to crops (Wang *et al.* 2018). Therefore, a conditioning step is required to stabilise the heavy metals in sewage sludge.

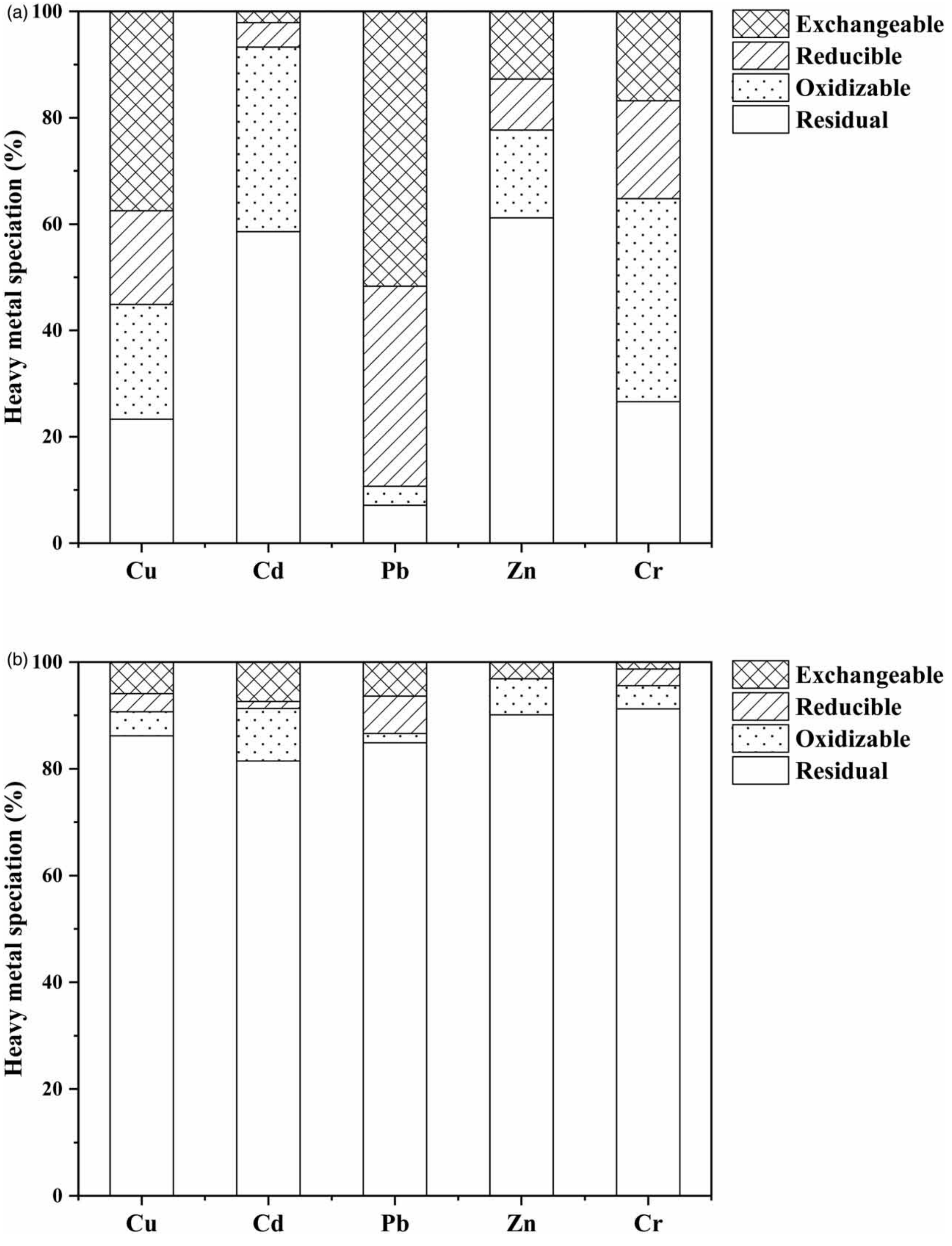


Figure 2 | Comparison of the heavy metal speciation in the sewage sludge (weight of each sample = 50 g) before and after conditioning with 3 g of the modified bentonite: (a) the raw sewage sludge and (b) the conditioned sewage sludge.

As shown in Figure 2(b), the residual fractions of most heavy metals (i.e., Cu, Cd, Pb, Zn, and Cr) in the sewage sludge were substantially increased after conditioning with 3 g of modified bentonite (Cu = 86.2%; Cd = 81.5%; Pb = 84.9%; Zn = 90.1%; Cr = 91.2%), whereas considerable decreases were observed for the oxidisable (Cu = 4.5%, Cd = 9.8%, Pb = 1.7%, Zn = 6.8%, Cr = 4.4%), reducible (Cu = 3.4%; Cd = 1.3%; Pb = 7.0%; Zn = 0%; Cr = 3.1%), and exchangeable fractions (Cu = 5.9%;

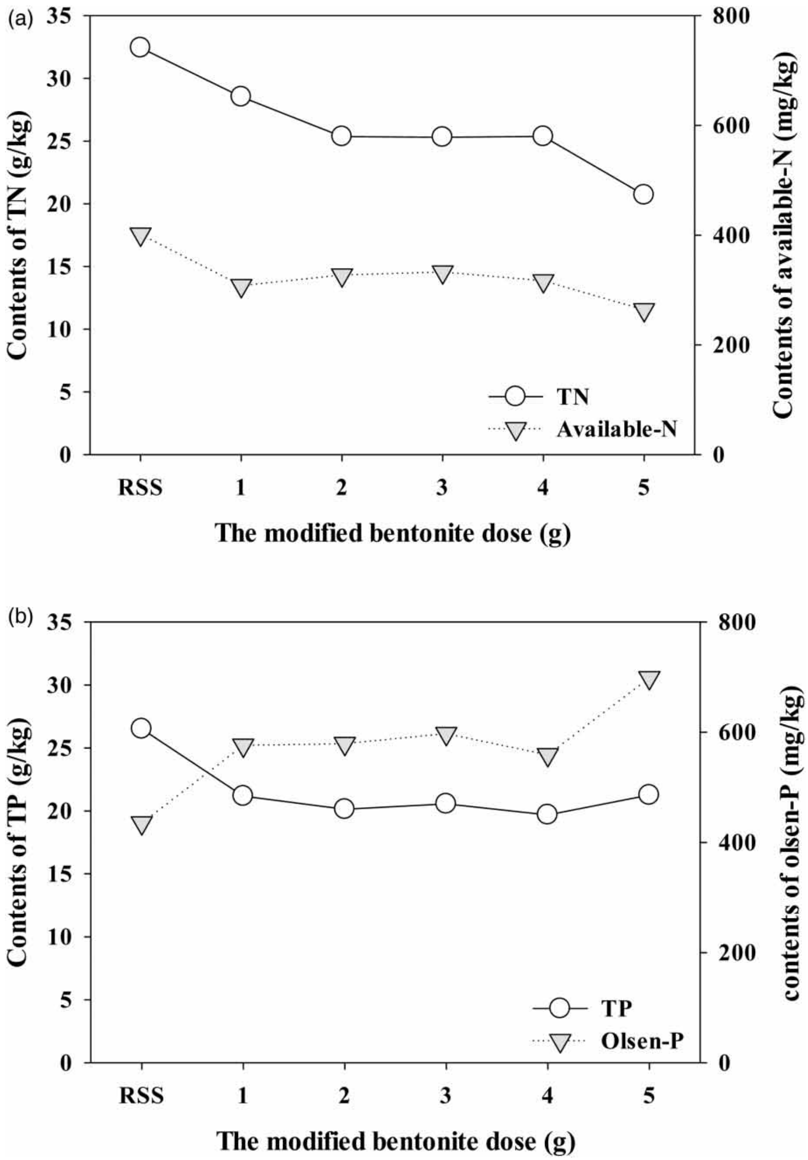


Figure 3 | The retention of nutrients in the sewage sludge (weight of each sample = 50 g) as a function of the modified bentonite dose (1–5 g): (a) nitrogen species and (b) phosphorus species (RSS: raw sewage sludge).

Cd = 7.4%; Pb = 6.4%; Zn = 3.1%; Cr = 1.3%). A possible explanation for the increase in the residual fractions is that the major component of modified bentonite (i.e., montmorillonite) can contribute to the stabilisation of heavy metals in the sewage sludge, which aligns with the results of Usman *et al.* (2005). Altogether, these results imply that conditioning with the modified bentonite may improve the opportunity for sewage sludge to be used as a fertiliser due to the improved stabilisation of heavy metals.

Effect of modified bentonite on nutrients

The contents of nutrients (i.e., TN, available N, TP, and Olsen-P) in the sewage sludge (sample weight = 50 g) before and after conditioning using five different amounts of modified bentonite (1–5 g) are compared in Figure 3. In this study, available N and Olsen P were used as surrogate indicators to evaluate the practical availability of nutrients in the sewage sludge as the high contents of TN and TP did not imply a high supply capacity of nutrients available for crops (Ren *et al.* 2015). As depicted in Figure 3(a), the contents of TN and available N in the sewage sludge were sharply decreased in the range of 1–2 g modified bentonite, and their losses were not significantly changed in the range of 3–4 g modified bentonite. However, remarkable decreases in TN and available N in the sewage sludge were found for the highly modified bentonite range (>4 g). The reduction rates of TN and available N in the conditioned sewage sludge using 5 g of modified bentonite were 36 and 34%, respectively. Similar results were found by Samara *et al.* (2019), who reported that bentonite has a negative impact on the concentration of TN and available N during the treatment of sewage sludge. The losses of TN and available N in the conditioned sewage sludge were attributed to the high cation-exchange capacity of the modified bentonite for capturing inorganic nitrogen species, especially NH_4^+ ions (Redding 2011).

As depicted in Figure 3(b), a substantial reduction in TP was observed in the low modified bentonite range (<1 g), and distinctive differences were not found for the applied modified bentonite amounts >2 g. The highest loss of TP occurred when the sewage sludge was conditioned using 4 g of modified bentonite (TP = 19.69 g/kg). The variations in Olsen-P during conditioning with the modified bentonite were considerably different from those of the TP. The Olsen-P content in the sewage sludge increased substantially with an increase in the amount of modified bentonite. The concentration of Olsen-P in the raw sewage sludge was 435.53 mg/kg, and that of the conditioned sewage sludge using 5 g of the modified bentonite was up to 699.05 mg/kg. These findings indicate that the use of modified bentonite as a conditioning agent may reduce the fixation of phosphorus in sewage sludge. This P reduction, except for the dilution effect, could be attributed to the formation of insoluble P substances that could not be extracted and of cation complexes incorporated or absorbed into minerals (Samara *et al.* 2019). Based on the observed results from the changes in the retention and availability of nutrient species, it can be postulated that optimisation of the modified bentonite amount is essential to minimise the losses of available N and maximise the availability of Olsen-P in the sewage sludge.

Application of the AHP model

To determine the best amount of modified bentonite to be used for sewage sludge, AHP modelling was used to evaluate heavy metal stabilisation and nutrient retention. SI. 3 and SI. 4 show the pairwise comparison of the influencing factors in the aspects of heavy metals and nutrient elements, respectively, consisting of the preference matrix and weights. The hierarchical structure of the evaluation factors was used to establish the preference matrix based on the scoring method evaluated by the experts (SI. 5). The preference matrix was normalised using Saaty's method, and the weight of each factor was calculated based on the results of the normalised data, where w_{mr} was (0.1500, 0.0854, 0.3620, 0.1293, 0.2733) and w_{ne} was (0.4263, 0.2270, 0.2270, 0.1223) (Bologa *et al.* 2018). To confirm whether the weight was reasonable, consistency tests [(consistency ratio (*C.R.*) and maximum eigenvalue (λ_{max})]) were performed, and both consistency tests in SI. 3 ($\lambda_{max} = 5.1174 \geq 5$ and $C.R. = 0.0262 < 0.10$) and SI. 4 ($\lambda_{max} = 4.0104 \geq 4$, $C.R. = 0.0039 < 0.10$) were passed; thus, the allocation of weights in the aspects of heavy metals and nutrient elements was deemed reasonable.

As an example of the addition of 4 g bentonite, the evaluation vector of the factors in the heavy metals, $P_{\text{mr-bo(4 g)}}$, was:

$$P_{\text{mr-bo(4 g)}} = w_{mr} \cdot B_{mr\text{-bo(4 g)}} = (0.1500,\ 0.0854,\ 0.3620,\ 0.1293,\ 0.2733) \cdot \begin{bmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \end{bmatrix}$$

$$= (0.5120, 0.3587,\ 0.1293, 0,\ 0)$$

The evaluation vector of the factors in the nutrient elements, $P_{\text{ne-bo(4 g)}}$, was

$$P_{\text{ne-bo(4 g)}} = w_{ne} \cdot B_{ne\text{-bo(4 g)}} = (0.4236,\ 0.2270, 0.2270,\ 0.1223) \cdot \begin{bmatrix} 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \end{bmatrix} = (0,\ 0.6506,\ 0.3493,\ 0, 0)$$

In this study, the weights between heavy metal elements and nutrient elements were 0.6 and 0.4, respectively. Based on the evaluation vectors of the factors in heavy metals and nutrient elements, the comprehensive evaluation vector ($P_{\text{bo(4 g)}}$) treated

Table 3 | Comprehensive evaluation of different bentonite inputs

Bentonite dose (g/50 g)	$P_{bo(ig)}$	$S_{bo(ig)}$
1	(0.1694, 0.1816, 0.0489, 0.2172, 0.3828)	0.3844
2	(0, 0.3510, 0.2661, 0.2540, 0.1288)	0.4598
3	(0.2172, 0.4000, 0.2540, 0.1288, 0)	0.6764
4	(0.3072, 0.4755, 0.2173, 0, 0)	0.7724
5	(0.3561, 0.3836, 0, 0.0908, 0.1694)	0.6665

with 4 g bentonite was as follows:

$$P_{\mathrm{bo(4\,g)}} = w \cdot B_{bo(4\,\mathrm{g})} = (0.6,\ 0.4) \cdot \begin{bmatrix} 0.5120 & 0.3587 & 0.1293 & 0 & 0 \\ 0 & 0.6506 & 0.3493 & 0 & 0 \end{bmatrix} = (0.3072,\ 0.4755,\ 0.2173,\ 0,\ 0)$$

The weights of ‘very good’, ‘good’, ‘general’, ‘poor’, and ‘very poor’ were 0.3072, 0.4755, 0.2173, 0, and 0, respectively. Thereafter, Bmr-bo(ig) (i = 1–5) was scored either ‘1’ or ‘0’ according to the stabilised performance of heavy metals (SI. 6). Similar to heavy metals, the nutrient elements also obtained the results of Bne-bo(ig) (SI. 7).

Furthermore, ‘very good’, ‘good’, ‘general’, ‘poor’, and ‘very poor’ were assigned values of 1, 0.75, 0.5, 0.25, and 0, respectively. The comprehensive score ($S_{\mathrm{bo(4\,g)}}$) as an evaluation value was

$$S_{bo(4\,\mathrm{g})} = P_{\mathrm{bo(4\,g)}} \cdot sv' = (0.3072,\ 0.4755,\ 0.2173,\ 0,\ 0) \cdot \begin{pmatrix} 1 \\ 0.75 \\ 0.5 \\ 0.25 \\ 0 \end{pmatrix} = 0.7724$$

Based on the results of the evaluation vectors ($P_{\mathrm{mr\text{-}bo}(ig)}$ and $P_{\mathrm{ne\text{-}bo}(ig)}$) shown in SI. 8, the other four comprehensive scores ($S_{\mathrm{bo(ig)}}$) were calculated in the same manner as $S_{\mathrm{bo(4\,g)}}$, and the five comprehensive scores were 0.3844, 0.4598, 0.6764, 0.7724, and 0.6665, respectively (Table 3). Therefore, both the maximum stabilisation of heavy metals and minimal losses of nutrients were obtained in the sludge when the ratio of bentonite to sludge was 1:12.5 (4 g bentonite : 50 g sludge).

Results for the mixture of sewage sludge and soil

The proportions of heavy metals and nutrients in the mixture of the sewage sludge and soil samples with and without modified bentonite treatment were investigated to determine the optimal mixing ratio of the sewage sludge and the soils (mixing ratios = 1:1–1:4). As shown in Table 4, the raw sewage sludge (RSS) contained a relatively greater amount of bioavailable heavy metals (Cu = 1,301.5 mg/kg; Cd = 1.3 mg/kg; Pb = 49.3 mg/kg; Zn = 780.8 mg/kg; Cr = 268.4 mg/kg) and nutrients (TN = 32.5 g/kg; TP = 26.5 g/kg; available-N = 401.3 mg/kg; Olsen-P = 435.5 mg/kg) than the soils (Cu = 0.9 mg/kg; Cd = 0.04 mg/kg; Pb = 1.2 mg/kg; Zn = 0.9 mg/kg; Cr = 1.3 mg/kg; TN = 1.1 g/kg; TP = 5.1 g/kg; available-N = 116.7 mg/kg; Olsen-P = 26.4 mg/kg). When the RSS and the soils were mixed, the contents of heavy metals and nutrients were considerably reduced by increasing the mixing ratio of the sewage sludge (1:1–1:4). Although the amounts of Cd, Pb, Zn, and Cr in the mixture of the raw sewage sludge and the soils at a mixing ratio of 1:4 satisfied the guidelines proposed by the EU Directive 86/278/ECC (ECC 1986), the Cu content exceeded its maximum allowable value in the soils. These results indicate that the direct use of sewage sludge as a fertiliser may have adverse effects on crops and human health.

Of note, the heavy metal and nutrient contents were higher in the mixture of the conditioned sewage sludge (modified bentonite dose = 4 g) and the soils under different mixing conditions (mixing ratios = 1:1–1:4) (Table 4). Besides the mixture of RSS and soil, after treatment with modified bentonite, the proportions of most bioavailable heavy metals and nutrients (Cu = 156.2 mg/kg; Cd = 0.6 mg/kg; Pb = 6.0 mg/kg; Zn = 147.7 mg/kg; Cr = 22.4 mg/kg; TN = 24.7 g/kg; TP = 19.7 g/kg; available-N = 321.5 mg/kg) were considerably decreased while the contents of Olsen-P were increased to 559.0 mg/kg, which

Table 4 | The contents of heavy metals and nutrients in the mixture of the sewage sludge (untreated and treated by 4 g modified bentonite) and the soils at the different mixing ratios

Sample		Heavy metals (mg/kg)								
		Cu	Cd	Pb	Zn	Cr	TN (g/kg)	TP (g/kg)	Available-N (mg/kg)	Olsen-P (mg/kg)
RSS		1,301.5	1.3	49.3	780.8	268.4	32.5	26.5	401.3	435.5
CSS		156.2	0.6	6.0	147.7	22.4	24.7	19.7	321.5	559.0
Soil		0.9	0.04	1.2	0.9	1.3	1.1	5.1	116.7	26.4
RSS : Soil	1:1	903.3	0.8	30.5	498.2	227.4	24.4	13.4	414.6	301.6
	1:2	285.1	0.3	23.2	369.5	144.1	13.8	8.9	342.9	253.7
	1:4	143.7	0.2	8.0	201.3	66.0	6.6	5.2	282.4	213.4
CSS : Soil	1:1	89.6	0.3	5.0	132.3	20.6	11.0	8.9	177.8	457.5
	1:2	59.6	0.2	4.3	107.8	14.0	9.3	7.7	146.4	389.1
	1:4	46.1	0.1	3.0	78.3	9.6	4.9	6.3	120.2	311.7

RSS: raw sewage sludge. CSS: conditioned sewage sludge.

met the maximum allowable levels of heavy metals in the soils established by EU Directive 86/278/ECC (ECC 1986). Similar to the results from mixing of the RSS and the soils, the amounts of heavy metals and nutrients gradually decreased with increasing mixing ratio of the conditioned sewage sludge (CSS) to the soils from 1:1 to 1:4. The concentration of bioavailable heavy metals was significantly lower, and the loss of N and P decreased slowly with the increase in soil proportion in the mixture of CSS and soils, compared to the mixture of RSS and soils. To avoid the potential toxic effects caused by the accumulation of heavy metals in the soils and to retain nutrient elements to a certain degree, a mixing ratio of 1:2 was recommended for agricultural uses of the conditioned sewage sludge as the contents of heavy metals at this ratio were lower than their minimum allowable values in the soils suggested by EU Directive 86/278/ECC (ECC 1986). Altogether, adequate conditioning using modified bentonite may enable sewage sludge to be directly used as a fertiliser.

CONCLUSIONS

Modified bentonite was applied to condition sewage sludge for agricultural reutilization (i.e., stabilisation of heavy metals and retention of nutrient species). Five grams of modified bentonite exhibited the highest stabilisation rate of heavy metals, thereby effectively stabilising the heavy metals in the conditioned sewage sludge. However, increased amounts of modified bentonite could reduce the retention of TN, available N, and TP in the conditioned sewage sludge, with the exception of Olsen-P. Through AHP modelling, the optimal concentrations of nutrient species and heavy metals were obtained in the conditioned sewage sludge when the ratio of the bentonite dosage to the sewage sludge was 1:12.5 (4 g bentonite:50 g sludge). Thus, a mixing ratio of 1:2 (conditioned sewage sludge : soil) was recommended to avoid the potential toxic risk of heavy metals with the effective retention of nitrogen species in the soils, which satisfied the maximum allowable concentrations of heavy metals in the soils established by EU Directive 86/278/ECC. These findings clearly indicate that adequate conditioning using modified bentonite might enable sewage sludge to be directly applied as a fertiliser in agriculture.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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