Long-term Impact of Organic Amendments on the Bioavailability of Heavy Metals in Mudflat Soil and Their Uptake by Maize

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Research Article

Keywords: organic amendments, mudflat soils, bioavailability, heavy metals, bioconcentration, sewage sludge

Posted Date: June 22nd, 2021

DOI: https://doi.org/10.21203/rs.3.rs-549265/v1

Abstract

Organic amendments (OAs) can be a sustainable and effective method for mudflat soil improvement. A long-term field experiment was conducted to investigate the potential of OA application to mudflat soil improvement. We measured the pH, soil organic matter (SOM), salinity, maize growth, and heavy metal (HM) accumulation in OA-amended soils and maize tissues 23 months after three OAs, sewage sludge (SS), Chinese medical residue (CMR), and cattle manure (CM), were applied at the application rates of 0, 30, 75, 150, and 300 t ha^{-1}. OA application significantly increased the SOM and decreased the salinity of mudflat soils. Additionally, CMR and CM application decreased soil pH. The maize biomass and HM contents in soil and maize increased after OA application. The bioavailability and bioconcentration of HMs were generally in the sequence of SS>CMR>CM. The average bioavailability ratios of HMs were in the order of Cd>Zn>Cu>Mn>Ni. The bioconcentration of Zn and Cd by maize was highest, followed by Mn, Cu, and Ni. SOM, pH, and salinity were the important factors regulating soil available HMs and, subsequently, HM accumulation in maize. Among the three OAs, SS is most effective in decreasing soil salinity and increasing the SOM, bioavailability and bioconcentration of HMs. On the other hand, CM was the best OA because it promoted significant maize growth yet maintained low HM contamination risk.

Introduction

Coastal mudflats, occurring in places with low gradients, abundant fine sediments, as well as active tidal forces and weak wave action, are the transition area between land and sea. The core of mudflats includes not only the intertidal zone but also the supra-tidal and subtidal areas (Long et al., 2016). As an evolving special ecosystem in the sea and land transition zone, the coastal mudflats are expanding at a rate of 3000 ha every year (Han et al., 2017). Under the shortage of arable land, coastal mudflats are considered a promising agricultural soil resource. However, the properties of mudflat soils, such as high salinity and alkalinity and low nutrient levels, are barriers to agricultural development. Large-scale soil reclamation using chemical fertilizer is too expensive. Furthermore, chemical fertilizer not only hardly maintains soil fertility at a high level but also causes soil acidification (Miao et al., 2011). Alternatively, organic wastes can be applied to improve soil physicochemical properties and crop yield (Yuan et al., 2014), which provides a sustainable way to improve soil fertility. The rapid industrialization and urbanization have increased the demand for disposal of solid wastes. An alternative to traditional disposal of organic wastes, such as landfills and incineration, is land application of organic wastes in which resources are recycled.

Sewage sludge (SS) and cattle manure (CM) are common organic amendments (OAs) that are rich in nutrients and organic matter (Eid et al., 2020). The production of SS and CM is 2.47×10^{7} Mg/yr and 8.10×10^{6} Mg/yr, respectively, in China (Thangarajan et al., 2013). SS and CM have been widely used in soil reclamation to enhance crop yield in agricultural practice (Mokgolo et al., 2019). However, the heavy metals (HMs) carried by those OAs also bring potential risks to food safety (Protano et al., 2020). The risk of HM contamination is particularly high in China because HM contents in SS in China are higher than those in the EU and the US (Dai et al., 2007). Cattle manure has been identified as the main source of Cu, Pb, and Zn in agricultural soils (Qiutong & Mingkui, 2017). In addition to increasing the HM contents in arable land, OA application also enhances HM bioavailability (Qiutong & Mingkui, 2017). OA application can promote HM transfer in soil-plant systems, exposing humans to the risk of soil HM contamination (Jolly et al., 2013; Wan et al., 2020).

Chinese medicine residue (CMR), the remnant after pharmaceutical compounds are extracted from medicinal herbs (Zhou et al., 2018), is an OA that has received less attention than SS and CM. Due to low extraction rate of the active ingredients of medicinal herbs, the production of CMR is up to 1.5 million tons a year in China (Wang et al., 2010). The improper disposal of CMR not only causes a waste of resources but also pollutes the environment. Similar to crop residues, CMR is rich in organic matters, sugars, cellulose, crude proteins, alkaloids and a few inorganic elements such as nitrogen (N), phosphorus (P), manganese (Mn), and zinc (Zn) and possesses favorable air permeability and light quality, which can improve the permeability of soil.

Previous studies on HM risk from OAs mainly focused on agricultural soils and conventional OAs. To our knowledge, there are few studies that have investigated how OAs, particularly CMR, affect HM contamination in saline-alkaline soil, such as mudflat soil. There are even fewer studies on the long-term assessment of HM contamination in soils and maize following green manuring in an OA-amended mudflat soil. Thus, the objectives of the present study were to 1) explore the long-term impact of application of SS, CMR, and CM on mudflat soil properties and maize growth, 2) assess the HM accumulation in the mudflat soil and maize, 3) identify the main environmental factors that control the fate of HMs, and 4) evaluate the ecological risk of HMs in the amended soils.

Material And Methods

Field description

The experiment was conducted in the Zhaoying reclamation area (121° 23′23"E, 32° 20′03"N) in Rudong county of Jiangsu Province, which is one of the four mudflat concentrated areas (Sheyang, Dafeng, Dongtai, and Rudong counties) (Wang & Wall, 2010). The coastal mudflats of Jiangsu Province, covering an area of 6.53×10^5 ha, account for a quarter of the total mudflats area of the whole country (Long et al., 2016). Approximately 37% (558 km^2) of all reclaimed areas (2250 km^2) in Jiangsu was used for agriculture in 2004 (Wang & Wall, 2010). This area belongs to the subtropical humid monsoon climate, with obvious transitional maritime and monsoon. Precipitation is mainly concentrated in June to August. The SS, CM, and CMR were collected in the municipal wastewater treatment plant of Rudong, a dairy farm of Rudong, and a Chinese medical factory, respectively. The physicochemical properties of OAs and the selected mudflat soils are shown in Table 1. The contents of HMs in sewage sludge were lower than the permissible thresholds regulated by Disposal of sludge from municipal wastewater treatment plant-Quality of sludge used in land improvement (GB/T 24600—2009).

Experimental design

Forty-five random experimental blocks that were 4.0 m x 4.0 m were established. There OAs, SS, CMR, and CM, with five application rates (0, 30, 75, 150, 300 t ha^{-1}) were studied in triplicate. Adjacent plots were 50 cm apart and separated by a 20 cm wide and 30 cm deep ditch. All OAs were added into soil through one-off application and mixed with the plow layer (0-20 cm deep) by rototiller on Oct 20, 2011. A green manure plant, ryegrass, was planted afterward and mowed on June 22, 2013. The maize seed was sowed on July 12, 2013. The soil samples and plant samples were collected on September 22, 2013 (70 days after seed sowing).

Soil analysis

Soil pretreatments including air-drying, removal of visible organic residues, grinding and sifting (1 mm and 0.150 mm) were conducted. Soil organic matter (SOM) was measured by the externally heated potassium dichromate ($K_2Cr_2O_7$) oxidation method (Yeomans & Bremner, 1988). Soil pH was measured by a pH meter (Model IQ150, Spectrum, USA) in soil slurries with a soil-to-water ratio of 1:5. Soil salinity was measured by the gravimetric method. For analysis of total heavy metals in soil samples, 0.5 g air-dried sample was put through the 0.150-mm mesh and digested in 20 mL of triacidmixture (HNO_3:H_2SO_4:$HClO_4$ 5:1:1) for 8 h at 80°C, and each sample had duplicates. After complete digestion solution was filtered and the filtrate was analyzed separately for manganese (Mn), copper (Cu), zinc (Zn), cadmium (Cd), and nickel (Ni) using an Atomic Absorption Spectrometer (AAS) (Model SOLAAR M6, Thermo Elemental, Thermo Fisher Scientific Inc., USA). Available metals were analyzed by diethylene triamine pentaacetic acid (DTPA) extraction method. 25.0 g air-dried sample through 1-mm mesh size was extracted in 50 ml DTPA solution for 2 h. After the complete extraction solution was filtered and the filtrate was analyzed using FAAS. For extracting total heavy metals in OA samples, 0.35 g air-dried soil sample through the 0.150 mm mesh sieve was digested with 6 mL nitric acid (HNO_3), 3 mL hydrochloric acid (HCl), and 0.25 mL hydrogen peroxide (H_2O_2) by microwave digestion system (Model MARS 6, CEM Corporation, USA). The digestion solution was filtered and detected for Cd, Cu, Ni, Mn, and Zn using AAS(Zeng et al., 2011).

Plant analysis

The biomass of maize was obtained by weighing the aboveground parts from each plot. For heavy metal analysis, oven-dried samples were homogenized by grinding in a stainless steel blender then passed through a sieve of 2-mm mesh size. For extraction of metals (Ni, Cu, Cd, Zn, Mn) from plant samples, 0.5 g oven-dried sample was digested in 10 L HNO_3:H_2SO_4:$HClO_4$ (5:1:1) until transparent color appeared. Metal concentrations were determined after filtering the digested samples using ICP-MS (Model iCAP 6300, Thermo Fisher Scientific Inc., USA) (Zuo et al., 2019a). For accuracy, soil and maize samples used for analysis had duplicates. Furthermore, certified reference materials (CRM) obtained from the China Institute of Geophysical and Geochemical Exploration for soil and plant were used to ensure the reliability and accuracy of test results.

Data analysis

One-way analysis of variance was performed using SPSS (version:20.0) to determine the effect of the OA application rate on the soil properties and HM contents in soils and maize. The mean values and standard deviation values were obtained via LSD. The significance of relative parameters was determined through Duncan's test at the level of $P < 0.05$. The correlation analysis was performed according to the method of Pearson or Spearman, depending on whether the statistics satisfy the normal distribution. PCA was performed by OriginPro (2020).

Results

Soil properties

SS application significantly increased SOM contents, which ranged from 7.13 to 32.29 g/kg (Table 2). When the SS application rate was higher than 150 t ha^{-1}, the further increase in SOM in amended soils was negligible. Soil salinity (7.67-2.06%) and pH (8.80-7.15%) significantly decreased with the increasing SS application rates. Soil pH, SOM, and salinity were impacted by CMR application significantly. The ranges of pH, SOM, and salinity were 9.19 to 8.73, 5.90 to 17.20 g/kg, and 7.15 to 2.62%, respectively. The SOM in CM-amended soils ranged from 13.00 to 19.60 g/kg. The SOM did not increase significantly with CM application until the CM application rate ≥150 t ha^{-1}. The pH and salinity significantly decreased with CM application rate, which ranged from 9.0 to 7.48 and 8.12 to 4.70%, respectively. The SOM values in amended soils were in the order of SS>CM>CMR. The lowest pH occurred in SS-amended soils, followed by CM- and CMR-amended soils. The order of salinity in amended soils was CM>CMR>SS.

Maize growth

The maize biomass increased significantly in CMR-amended soils, which ranged from 42.2 to 93.0 kg (Fig. 1). The biomass of maize was the highest at the CMR application rate of 150 t ha^{-1}, and the increase in the biomass at the 150 t ha^{-1} rate was 2.20-fold in compared to that in the control soils. CM application at 150 t ha^{-1} and SS application at 75, 150, and 300 t ha^{-1} significantly increased the maize biomass compared to the control.

Heavy metals in the mudflat soils

The relationship between the OA application rates and the total amount of soil HMs is shown in Fig. 2. CMR application led to the highest accumulation of the soil total Mn ($P<0.01$), which increased by 38.4% at the 300 t ha^{-1} CMR application rate, compared to the control. SS application had no impact on total Mn contents. The soil total Zn contents at the 300 t ha^{-1} application rate increased by 184.7%, 61.2% and 60.3% in SS-, CMR-, and CM-amended soils, respectively, compared to those in the control soils. All OA treatments showed that soil total Zn content increased significantly with the application rates. Compared with the control soils, the soil total Cu in SS-amended soils increased by 167.8% at the 300 t ha^{-1} rate. However, the soil total Cu contents in CMR- and CM-amended soils were not impacted by the OA application rate significantly. OA application had a negligible impact on the soil total Ni contents except at the 300 t ha^{-1} CMR application rate. Soil total Cd concentrations after OA treatments were significantly higher than in the control soil. The soil total Cd contents increased significantly with increasing OA application rates ($P<0.01$). Compared with those in the control soils, the soil total Cd contents at the 300 t ha^{-1} rate increased by 61.0%, 72.0%, and 89.0% in SS-, CMR-, and CM-amended soils, respectively. The total HM concentrations were in the order of Mn>Zn>Ni>Cu>Cd in CMR- and CM-amended soils and Mn> Zn > Cu > Ni >Cd in SS-amended soils.

The bioavailability ratio (BR) of HMs, the ratio of bioavailable HM contents to soil total HM contents, was used to represent the bioavailability and mobility of HMs in a soil-to-crop system after OA application. The BR of Mn increased with the increasing SS application rates ($p<0.01$), which ranged from 8.4% to 10.2%, and increased by 21.4% at the 300 t ha^{-1} SS application rate compared to that in the control soils (Fig. 3). However, the BR of Mn in CMR-amended soils was significantly lower than that in control soils, and CM application had no impact on the BR of Mn. SS application changed the BR of Zn significantly ($p<0.01$), and there was a peak (45.80%) at the 75 t ha^{-1} application rate. The BR of Zn increased by 243.9% at the 300 t ha^{-1} CM application rate compared to that in the control soils ($p<0.05$). The BR of Zn of CMR-amended soils did not change with increasing application rates. The BR of Zn in SS-amended soils was significantly higher than that in CM- and CMR-amended soils. The BR of Cu in SS-amended soils increased by 96.3%, reached a peak at the 150 t ha^{-1} application rate compared to that in the control soils ($P<0.001$) and was significantly higher than that in CM- and CMR-amended soils. In contrast, the BR of Cu did not change significantly in CMR- and CM-amended soils. The BR of Ni increased by 216.5% and 14.8% at the 300 t ha^{-1} rate, in SS- and CMR-amended soils, respectively, compared to that in the control soils ($p<0.05$). The BR of Ni was not affected by the CM application rate. The BR of Ni in SS-amended soils was significantly higher than that in CM- and CMR-amended soils. The BR of Cd increased by 65.8% at the 300 t ha^{-1} SS application rate compared to that in the control soils ($p<0.05$). The BR of Cd significantly decreased with the CMR and CM application rates. The BR of Cd in SS-amended soils was significantly higher than that in CM- and CMR-amended soils.

Heavy metals in maize parts

OA application significantly increased the Cu contents in the maize (Table 3). The Cu contents in the straw were higher than those in the grain and leaf. The Cu contents in the straw, grain, and leaf at the 300 t ha^{-1} rate increased by 41.8%, 2.0%, and 12.4% in SS-amended soils and by 48.9%, 143.2%, and 57.1% in CMR-amended soils, respectively, compared to those in the control soils. The Cu contents of maize in CM-amended soils showed no change with CM application.

The Mn contents in the maize were significantly affected by the application rate in SS- and CMR-amended soils. The leaf was the primary organ storing Mn (Table 3). The Mn contents in the straw, grain, and leaf increased by 132.0%, 77.69% and 231.26% and by 74.14%, 5.30% and 86.43% at the 300 t ha^{-1} SS and CMR application rates, respectively, compared to those in the control soils. CM application had no influence on the Mn contents in the maize. There were significant increases in Mn in leaves when the SS and CMR application rates were equal to or higher than 150 and 30 t ha^{-1}, respectively.

Similarly, the Zn contents in the straw were higher than those in the grain and leaf (Table 3). At the 300 t ha^{-1} rate, the Zn contents in the straw, grain, and leaf in SS-amended soils increased by 280.6%, 34.8%, and 839.5%, respectively; the Zn contents in the straw and leaf in CMR-amended soils increased by 22.6% and 59.3%, respectively; the Zn contents in the straw in CM-amended soils increased by 133.3% compared to those in the control soils. In addition, SS amendment led to the highest Zn contents in the straw and leaf of maize among the three OAs.

The Ni contents in the grain and leaf at the 300 t ha^{-1} rate in SS-amended soils increased by 91.1% and 51.8%, respectively, compared with those in the control soils (Table 3). The Ni contents of the straw, grain, and leaf at the 300 t ha^{-1} rate in CMR-amended soils increased by 149.1%, 53.7% and 150.0%, respectively, compared to those in the control soils. The Ni contents in straw were significantly affected by CM application and increased by 79.0% at the 300 t ha^{-1} rate compared to those in the control soils.

OA significantly increased the Cd contents in the maize (Table 3). The Cd contents in the straw, grain, and leaf increased by 124.2%, 885.1% and 308.8% in SS-amended soils and 53.6%, 136.2%, and 14.9% in CMR-amended soils at the 300 t ha^{-1} rate, respectively, compared to the controls. In addition, SS amendment led to the highest Cd contents in the grain and leaf of maize among the three OAs.

Bioconcentration factors

We adopted the bioconcentration factors (BCFs), the ratio of HM contents in plant parts to HM contents in soils, to characterize the efficiency of HM accumulation in maize aboveground parts. For Mn, the leaf of maize was the main aboveground part with accumulation (Fig.4). The BCFs of Mn in aboveground tissues of maize were significantly impacted by OA application, except the BCF of Mn in the grain and leaf in CM-amended soils. The sequence of BCFs of Mn in the straw, grain and leaf followed SS>CM>CMR. For Zn, the highest BCFs of aboveground parts occurred in the straw of maize. The BCFs of Zn in the straw followed this order: SS>CMR >CM. The BCFs of Zn in the aboveground parts increased with OA application in SS-amended soils. For Cu, the order of accumulation in aboveground parts was straw>leaf>grain. The average BCFs of Cu were in the order of SS>CMR>CM. The orders of BCF of Ni of aboveground parts were grain>leaf>straw and SS>CMR>CM. For Cd, the BCFs of aboveground parts were impacted significantly by SS and CMR application. The BCFs of Cd were in the order of SS>CMR>CM. The leaf had the highest BCFs of Cd, followed by the grain and straw.

In SS-amended soils, the BCFs generally followed the order: Zn>Cd>Mn>Cu>Ni>. In CMR-and CM-amended soils, the BCFs generally followed the order Cd>Zn>Cu>Mn>Ni>.

Principal component analysis (PCA)

PCA for bioavailable HM contents in soil and soil properties is shown in Fig. 5 a. The first principal component, accounting for 85.69% of variation, could represent the contents of bioavailable HMs (and SOM. Salinity and SOM showed a negative correlation and reversed control of the BRs of HMs. The data were mainly clustered by OA type rather than by OA application rate. The comprehensive score sites of SS-amended soils were mainly distributed along the first and second quadrants compared to those of CMR- and CM-amended soils, which mainly amassed in the second and third quadrants. SS-amended soils were mainly associated with significant variations in SOM and the BRs of HMs.

PCA of HMs in plant tissues and soil properties extracted two principle components, which explained a cumulative variance of 74.3% (Fig. 5 b). PC1 explained 62.8% of the cumulative proportion and contained most HM concentrations in aboveground parts and SOM, pH, and salinity. PC2 (11.5%) represented the Ni contents in grain and the Cu contents in leaf.

Correlation analysis

In SS-amended soils, the BRs of all treatments did not correlate with salinity (Table 4). The BRs of HMs showed a positive correlation with SOM and a negative correlation with pH.

In CMR-amended soils, pH had a positive correlation with the BRs of Mn and Cd and a negative correlation with the BRs of Zn, Cu, and Ni. Only the BRs of Cd were correlated with SOM. The impact of salinity on the BRs of HMs was minimum except the BRs of Cd showed a negative correlation with salinity.

The BR of Cd in CM-amended soils showed a correlation with SOM ($r = 0.976^{**}$), pH ($r = -0.967^{**}$), and salinity ($r = -0.897^{**}$). There was no correlation between the BR of Cu and Ni with SOM, pH, and salinity. The BRs of Zn positively correlated with pH, and the BR of Mn and Zn negatively correlated with salinity.

For the mixed data from three OAs, the BRs of HMs were generally positively correlated with SOM and negatively correlated with pH and salinity, except the BR of Ni was not correlated with any soil parameters.

Discussion

Soil properties

Previous studies confirmed that the application of CMR (Ma et al., 2019), SS (Eid et al., 2019b; Lin et al., 2018), and CM (Watanabe et al., 2019) significantly increased SOM, which was consistent with the current study. Increased SOM is an important index of soil fertility. SS was the most effective in increasing SOM, followed by CM, and then CMR, although the organic matter carried by CMR was highest. This might be attributed to that SS might have led to the formation of the most stable soil aggregate that prevents SOM from decomposition.

Soil pH values showed a slight reduction after OA application in the present experiment. The decreased pH values also occurred in the soils improved by biosolids (Zuo et al., 2019a), traditional Chinese medicine residues (Ma et al., 2019), and municipal solid waste compost with 50% recommended doses of fertilizer (Meena et al., 2016). The decreased pH after OA application might be attributed to the humic acid resulting from the degradation of OAs (Moreno, Garcia et al. 1997), the oxidation of N and S compounds (Logan, Lindsay et al. 1997) and the neutralization of relatively low pH OAs, particularly at high application rates (Bai, Zuo et al. 2017).

Soil salinity decreased after OA application, which was also reported by Zuo et al. (2019a). OA application can improve the soil structure by binding mineral particles to organic polymers (Leogrande and Vitti 2019), which contributes to reduced bulk density and increased porosity, water holding capacity (Shi, Zhao et al. 2016) and hydraulic conductivity. The improvement of soil structure makes the water infiltration faster, bolstering salt leaching (Grigg, Sheridan et al. 2006). As a result, soil salinity decreases. SS was most effective in lowering soil salinity probably because SS-amended soils had the highest SOM, which promotes soil aggregate formation. There was a negative correlation between organic matter and salinity (SS: r=-0,922, MR: r=-.873, CM: r=-0.614, P<0.01). Unlike saline mudflat soil, OA application generally increased soil salinity in nonsaline soils due to the high salinity of OAs. For example, Eid et al. (2019b) reported that the salinity of original soils (EC: 0.06 mS cm^{-1}) significantly increased after applying SS (EC: 2.07 mS cm^{-1}) at rates of 0 to 50 g kg^{-1}.

Maize growth

OA application significantly increased the maize yield. OAs have been found to increase the yield of maize (Zhang et al., 2012) and rice (Chen et al., 2017) . The improved soil quality, such as increased SOM and other nutrients (data not shown), and decreased salinity and pH may contribute to the increased crop yield. In addition, OAs had a positive impact on soil microbial activities and nutrient supply, which was beneficial for crop growth (Yadav et al., 2000). In the current experiment, the biomass of maize increased with the increasing CMR application. Though CMR application introduced massive HMs into mudflats, it did not decrease the growth of maize, which is consistent with the findings of previous studies (Ma et al., 2019). Jin et al. (2018) found that the application of CMR significantly increased the yield of *Pleurotus ostreatus* and nutrients in growing substrates. As SS application increased SOM the most, it might have led to the greatest increase in the maize yield. The absence of further increases in maize biomass at higher application rates suggested that the massive HMs caused by SS application may have inhibited the maize growth, which was previously reported by Watanabe et al. (2019).

Heavy metals in the soil

OA application was considered an additional source for soil HMs (Chaney, 2012). In SS-amended soils, soil total HM contents (Zn, Cu, and Cd) increased with the increasing SS application rates, but soil Mn and Ni contents were unchanged because their concentration differences between SS and mudflat soil were small (Table 1), which was also demonstrated by Bai et al. (2017b). Liang et al. (2010) found that

SS application significantly increased Mn and Cu contents in soils and Zn contents in crops. In CMR-amended soils, soil total metal contents (Mn, Zn, and Cd) increased with increasing CMR application, except Cu and Ni. Previous studies have found that CMR application significantly increased soil total heavy metals (Cr, Cd, Pb, As, and Hg) (Ma et al., 2019). The Zn and Cd contents increased significantly after CM application. Soil total HM contents in amended soils complied with the limitations of the Chinese soil risk control standard (GB15618-2018), except soil total Zn above 150 t ha^{-1} SS application rates. The Zn contents in SS-amended soils were much higher than those in CMR- and CM-amended soils at the same application rates, which may have resulted from the excessive Zn in SS (Table 1). The HM contents of CMR and CM-amended soils were similar in the current study due to the similar HM contents in CMR and CM. In general, the highest level of HMs in SS led to the highest HM contents in the amended soil.

The risk of HM contamination depends on not only the total soil HM concentration but also the bioavailable HM concentrations (Liu & Sun, 2013). The bioavailability of HMs was in the order of SS> CMR>CM, except for Mn (SS>CM>CMR), indicating that SS is the most effective in increasing HM bioavailability. The order of the average BRs of HMs was Cd>Mn>Cu>Zn>Ni in CMR- and CM-amended soils, while in SS-amended soils, the average BRs of HMs followed the order of Cd> Zn > Cu > Mn>Ni , suggesting that bioavailable fractions of Cd, Mn, Cu, Zn are high and therefore might lead to more plant bioconcentration. Mobile Cd was found to account for approximately 40~50% of Cd forms, contributing to the highest average BR values among all HMs (Ramos et al., 1994). For example, 37% and 45% of the total Cd was leached with 0.1 M of NaCl from two soil columns (Norrström, 2005). The responses of the BRs of HMs to OA application rates varied with the kind of OAs. The bioavailability of HMs (Zn, Cu, Cd, Mn, and Ni) increased significantly with the increasing SS application in the present study, consistent with the results of a previous study (Kidd et al., 2007). CMR application increased the bioavailability of Ni and decreased the bioavailability of Mn and Cd, and only the bioavailability of Zn increased with the CM application rates.

The bioavailability of HMs is associated with some chemical processes, such as sorption/desorption and precipitation/dissolution (Skrbic & Durisic-Mladenovic, 2010), which are regulated by environmental factors such as pH, SOM, and salinity (Du Laing et al., 2002).

The bioavailability of HMs was positively related to SOM, except for Ni and Mn in this study (Table 4 Figure 5 A). Bai et al. (2017a) reported that the bioavailability ratio of HMs showed a positive correlation between SOM after SS application in mudflats, except for Zn. Increased organic acid resulting from the decomposition of SOM can enhance the bioavailability of HMs (Ma et al., 2020; Zuo et al., 2019b). SOM, acting as a chelate, can form soluble complexation with HMs, thus enhancing the bioavailability of HMs (Vega et al., 2004). For example, OA application increased dissolved soil organic matter, which complexed with soil Cd, promoting the mobilization of Cd at alkaline pH by forming water-soluble Cd contents (Fang et al., 2018). The degree of correlation between the bioavailability of HMs and SOM is higher for SS than for CMR and CM, which may be attributed to the fact that the binding capacity for heavy metals varies with organic acid molar weight and functional group (Ma et al., 2020).

The chemical forms of heavy metal were strongly impacted by salinity. Soil salinity increased the bioavailability of soil HM (Wang & Song, 2019). The effects of salinity on HM bioavailability are regulated by two main opposite processes: (1) the competition between salt-derived cations and positively charged heavy metals over sorption sites on the solid phase and (2) the complexation capacity of salt-derived anions with heavy metals (Zhou et al., 2019). Mujeeb et al. (2020) reported that soil salinity increased the bioavailability of Cu, Mn, Zn, and Pb. However, the bioavailability of HMs mostly decreased with the increasing soil salinity. The negative correlation between soil salinity and HM availability may be attributed to the fact that salinity was negatively related to SOM and that the effect of salinity was masked by that of SOM (Fig. 5). In addition, previous studies mainly focused on the influence of single salts, such as NaCl (Wang & Song, 2019), ignoring the effects of other salts or the specific mixing ratio of salts. Nevertheless, salt types and concentrations in real soils are extremely complex, and the effect of salinity varied with the kind of salt. For example, due to the low stability of Cu-chloride complexes, NaCl had no significant impact on Cu mobility (Du Laing et al., 2008),

Correlations between pH and the bioavailability of HMs vary with OAs and HM species. The effect of pH on the bioavailability of HMs was most significant in CMR, followed by SS and then CM treatments. The BRs of HMs had positive correlation with pH when all OA treatments were combined, except Ni and Mn. In general, the bioavailability of HMs increased with the decreasing pH because lower pH values enhance the transformation from the immobile form to the bioavailable form (Yu et al., 2016), for example, through decreased adsorption of dissolved ions of HMs by a negative surface with decreasing pH (Sungur et al., 2014).

The accumulation and bioconcentration of heavy metals in maize

The HM contents in maize straw, grain, and leaf were mostly amplified with increasing OA application, which was consistent with the results of previous studies (Bose & Bhattacharyya, 2008; Eid et al., 2019b; Taghipour & Jalali, 2019) that had demonstrated that OA application increased the uptake of HMs in wheat (Eid et al., 2019a), spinach (Eid et al., 2017), and sugarcane (Nogueira et al., 2013). The HM concentrations in aboveground parts of maize were lower than the threshold of the national standards for food safety, although Cd contents in the grain and leaf of maize surpassed the threshold (GB2762-1017) at the 150 and 300 t ha^{-1} SS application rate. Thus, except for the SS application rate of 150 and 300 t ha^{-1}, mudflat soil amendment by SS, CMR, and CM may be practical and safe. In addition, the HM contents in maize grown in SS-amended soil is the highest due to the highest HM contents is SS.

The responses of BCFs of HMs to OA application varied with the OA types (Fig.4). The BCFs of Zn in SS-amended soils were also much higher than those of other OA treatments, which may be attributed to the high bioavailability of Zn in SS-amended soil. The bioconcentration of HM in maize generally followed the order SS>CMR>CM, which is consistent with the order of bioavailability of HM. The impact of OA on BCFs of HMs varied with HM and tissue types. The BCFs of Cu and Ni in aboveground tissues were not affected by SS amendment, while the BCFs of Cd, Mn, and Zn increased with the increasing SS application rates. Eid et al. (2019b) reported that the BCFs of Cu and Mn in root decreased with the increasing SS application, while the BCFs of Ni and Zn in root increased after SS application. Kidd et al. (2007) reported that the bioaccumulation factors for Mn and Zn decreased after SS application. In general, the bioconcentration of Cd and Zn by maize was highest, followed by Cu, Mn, and Ni,, which was consistent with the results of many studies of wheat straw (Ul Hassan et al., 2017), and wheat grain (Chen et al., 2018), indicating that Zn and Cd are preferably absorbed by plants, so the potential risk of Zn and Cd accumulation in plants is high.

The main aboveground reservoirs of HM varied with the type of HMs (Bai et al., 2017b). For example, the BCFs of Cu and Zn in the straw were higher than those of other HMs, suggesting that Cu and Zn accumulate more easily in the straw. The bioconcentration and transfer of HM in plants vary with plant tissues (Taghipour & Jalali, 2019; Zhang et al., 2018). The BCFs of HMs (Mn, Cu, and Ni,) in the straw, grain and leaf were lower than 1 in all treatments, which implied the limited accumulation ability of these HMs in aboveground parts of maize (Yoon et al., 2006). Orman et al. (2014) showed that the BCFs of HMs (Mn, Cu, Ni, and Zn) in shoot and root of Alfalfa were lower than 1 after SS application (0-80 t ha^{-1}). However, the BCFs of Cd in the grain and leaf in SS-amended soils at the 300 t ha^{-1} application rate were higher than 1, suggesting that the grain and leaf absorb Cd more easily, which poses a great risk for grain consumption or straw return to field. Compared to other HMs, Cd easily transfers from soils to grain, and Cd, Cu, and Mn easily accumulate in leaf. However, Li et al. (2012) demonstrated that the BCF of straw (49.5%) for Cd was higher than the BCF of grain (3.6%) in maize, which implied that there was more Cd accumulation in straw than in grain. Carbonell et al. (2011) reported that the grain of maize accumulated the least Ni and Cd. This discrepancy may be attributed to the different physicochemical properties of soil and OAs from the current study.

The HM contents in the maize tissues showed a significantly positive correlation with soil bioavailable HM contents, which is consistent with the results of previous studies (Bai et al., 2013; Bai et al., 2017b) (Table 5). The soil bioavailable HM contents could accurately predict HM uptake by plants (Wan et al., 2020)Generally, SS is associated with stronger correlation than CMR and CM, and Zn and Ni showed better correlation than other HMs. The differences may be attributed to the various accumulation potentials of tissues for different HM species. In addition to soil properties, such as pH, SOM, and salinity, the interactions of multiple HMs in soils, such as coexistence and antagonistic or synergistic actions, may multiply, suppressing the effect of single heavy metals on soils or crops (Puschenreiter et al., 2005). For example, the increase in bioavailable Cd may lead to a decrease in the bioconcentration of Zn under saline conditions (Taghipour & Jalali, 2019).

Conclusion

We conducted a long-term assessment (i.e. 23 months after OA application) of HM contamination in soils and maize following green manuring in an OA-amended mudflat soil. We found that OA application significantly increased the SOM and decreased the salinity of mudflat soils. Additionally, OA application decreased soil pH. The maize biomass and HM contents in soil and maize increased after OA application. The bioavailability and bioconcentration of HMs were generally in the sequence of SS>CMR>CM. The average bioavailability ratios of HMs were in the order of Cd>Zn>Cu>Mn>Ni. The bioconcentration of Zn and Cd by maize was highest, followed by Mn, Cu, and Ni. SOM, pH, and salinity were the important factors regulating soil available HMs and, subsequently, HM accumulation in maize. Among the three OAs, SS is most effective in decreasing soil salinity and increasing the SOM, bioavailability and bioconcentration of HMs. On the other hand, CM showed overall best performance in increasing maize yield while maintaining low risk of HM contamination. Our data also indicate that the optimal application rate for SS and CM is 30 t ha^{-1}, at which the yield of maize reaches a plateau, while the risk of HM contamination stays low.

Abbreviations

Organic Amendment (OA), sewage sludge (SS), Chinese medical residue (CMR), and cattle manure (CM), Heavy metal (HM), Principle Component Analysis (PCA)

Declarations

Acknowledgements

This study was supported by the Fundamental Research Funds for the Central Universities of China (310432103), the Interdisciplinary Research Funds of Beijing Normal University, National Natural Science Foundation of China (41828701), and the Blue-Blue Project and High-Rank Talent of Yangzhou University and Jiangsu Province.

Ethics approval and consent to participate: Not applicable

Consent for publication: Not applicable

Availability of data and materials: The datasets used and analysed during the current study are available from the corresponding author on reasonable request.

Competing interests: The authors declare that they have no competing interests

Funding: This study was funded by the Fundamental Research Funds for the Central Universities of China (310432103), the Interdisciplinary Research Funds of Beijing Normal University, National Natural Science Foundation of China (41828701), and the Blue-Blue Project and High-Rank Talent of Yangzhou University and Jiangsu Province.

Author's contributions: All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Ying Yu, Chuanhui Gu, and Wengang Zuo. The first draft of the manuscript was written by Ying Yu and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript. The detailed contributions were broken down as follows: Conceptualization: Chuanhui Gu and Yanchao Bai; Methodology: Ying Yu and Wengang Zuo; Formal analysis and investigation: Ying Yu and Wengang Zuo; Writing - original draft preparation: Ying Yu; Writing - review and editing: Ying Yu, Chuanhui Gu, Yanchao Bai, and Wengang Zuo; Resources: Yanchao Bai and Chuanhui Gu; Supervision: Chuanhui Gu and Yanchao Bai.

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Tables

Table 1 The physicochemical properties in the mudflat soil and OAs.

	pH	OM (g/kg)	Total Mn (mg/kg)	Total Zn (mg/kg)	Total Cu (mg/kg)	Total Cd (mg/kg)	Total Ni (mg/kg)
The mudflat soils	9.02±0.2	3.34±0.33	153.1±3.7	56.2±2.7	15.9±0.3	1.60±0.1	30.9±2.4
SS	6.32	377.00	129.5	2127.3	1121.9	3.30	52.8
CMR	7.74	415.70	133.9	146.7	769.4	2.15	18.0
CM	7.68	242.00	147.9	116.7	713.7	1.96	26.7

Table 2 The effect of OA application rate on the SOM contents, pH, and salinity (mean±SD) in three treatments.

OA	OA application rates (t ha^{-1})	SOM (g/kg)	pH	Total salinity (%)
Sewage sludge	0	7.13 ±1.51^{c}	8.80±0.17^{a}	7.67±0.11^{a}
	30	14.08±4.92^{b}	8.33±0.20^{b}	4.76±0.15^{b}
	75	19.10 ±0.2^{b}	8.01±0.12^{b}	3.08±0.05^{c}
	150	30.00 ±4.5^{a}	7.51±0.18^{c}	2.56±0.25^{d}
	300	32.30 ±3.3^{a}	7.15±0.26^{d}	2.06±0.24^{e}
Chinese medical residue	0	5.90 ±0.6^{c}	9.19±0.03^{a}	7.15±0.23^{a}
	30	7.50 ±1.2^{c}	9.01±0.09^{ab}	6.34±0.14^{b}
	75	8.10 ±1.1^{c}	9.05±0.01^{ab}	5.10±0.46^{c}
	150	12.20 ±2.8^{b}	8.95±0.19^{b}	4.18±0.28^{d}
	300	17.20 ±3.6^{a}	8.73±0.02^{c}	2.62±0.39^{e}
Cattle manure	0	13.00±0.3^{b}	8.80±0.17^{a}	8.12±0.20^{a}
	30	13.50±0.1^{b}	8.33±0.20^{b}	6.90±0.10^{b}
	75	13.70±0.7^{b}	8.01±0.12^{b}	5.36±0.31^{c}
	150	18.20±4.3^{a}	7.51±0.18^{c}	5.38±0.30^{c}
	300	19.60±0.9^{a}	7.15±0.26^{d}	4.70±0.10^{d}

The lowercase (a, b, c, d, and e) represents the significance of the data. Values that follow the same lowercase within each column are not significantly different by Duncan's multiple range test at the level of $P<0.05$.

*** represents the significance at $P<0.001$

** represents the significance at $P<0.01$

*represents the significance at $P<0.05$

Table 3 Heavy metal contents in aboveground parts of maize in mg/kg

			OA application rates (t ha^{-1})				
			0	30	75	150	300
SS	Mn	Straw	5. 92±1. 99b	5. 49±1. 20b	7. 51±0. 97b	6. 39±0. 11b	13. 73±2. 97a
		Grain	2. 77±0. 25b	2. 60±0. 16b	2. 67±0. 40b	2. 73±0. 29b	4. 92±0. 26a
		Leaf	33. 82±10. 60c	37. 02±9. 78c	37. 05±7. 52c	71. 65±14. 38b	112. 04±6. 51a
	Cu	Straw	4. 66±0. 50c	5. 42±0. 26b	6. 12±0. 15a	6. 52±0. 35a	6. 61±0. 46a
		Grain	1. 56±0. 11a	1. 74±0. 21a	1. 77±0. 06a	1. 26±0. 13b	1. 59±0. 11a
		Leaf	4. 52±0. 40a	3. 65±0. 37c	3. 41±0. 12c	4. 12±0. 45b	5. 08±0. 14a
	Zn	Straw	124. 69±13. 24c	169. 86±13. 44c	262. 96±20. 18b	312. 46±83. 93b	474. 92±4. 65a
		Grain	23. 11±2. 49b	23. 04±0. 73b	32. 20±2. 75a	30. 94±3. 94a	30. 89±0. 46a
		Leaf	41. 14±6. 67c	78. 26±20. 16c	188. 25±33. 37b	246. 16±51. 22b	385. 70±109. 97a
	Ni	Straw	3. 27±0. 30a	3. 14±0. 26a	3. 26±0. 29a	3. 31±0. 75a	3. 52±1. 09a
		Grain	1. 04±0. 13c	1. 18±0. 14b	1. 52±0. 15b	1. 59±0. 18a	1. 99±0. 40a
		Leaf	1. 92±0. 09b	2. 65±0. 55a	2. 91±0. 19a	2. 86±0. 32a	2. 91±0. 58a
	Cd	Straw	0. 17±0. 03c	0. 34±0. 03b	0. 34±0. 01b	0. 38±0. 01a	0. 39±0. 01a
		Grain	0. 33±0. 05d	0. 68±0. 1c	0. 78±0. 18c	1. 47±0.05b	3. 26±0. 1a
		Leaf	0. 84±0. 2d	1. 02±0. 25d	1. 79±0. 08c	2. 24±0. 06b	3. 44±0. 1a
CMR	Mn	Straw	3. 59±0. 20b	3. 13±0. 38b	3. 93±0. 22b	6. 30±0. 62a	6. 25±1. 35a
		Grain	2. 99±0. 08b	2. 96±0. 07b	3. 04±0. 25b	3. 41±0. 23a	3. 15±0. 14a
		Leaf	21. 77±0. 80c	32. 47±1. 43b	31. 99±0. 84b	32. 13±1. 51b	40. 58±1. 99a
	Cu	Straw	3. 61±0. 23b	3. 86±0. 38b	4. 78±1. 01a	4. 77±0. 56a	5. 370. 58a
		Grain	0. 80±0. 21b	0. 50±0. 14b	0. 81±0. 32b	1. 61±0. 34a	1. 95±0. 41a
		Leaf	3. 13±0. 21c	4. 29±0. 76b	5. 41±1. 01a	5. 86±0. 65a	4. 92±0. 32a
	Zn	Straw	53. 31± 1. 59b	55. 52±4. 42b	58. 77±1. 01b	65. 82±2. 32a	65. 34±3. 05a
		Grain	12. 72±1. 34a	13. 16±0. 19a	14. 60±1. 49a	13. 00±0. 15a	14. 97±0. 98b
		Leaf	23. 21±1. 88b	26. 17±1. 37b	28. 93±3. 61a	36. 51±7. 84a	36. 96±4. 07c
	Ni	Straw	1. 59±0. 14c	1. 96±0. 43b	1. 89±0. 21c	2. 43±0. 29b	3. 97±0. 30a
		Grain	1. 48±0. 15b	1. 58±0. 14b	2. 21±0. 25a	2. 12±0. 13a	2. 28±0. 14a
		Leaf	1. 34±0. 24d	1. 91±0. 15c	2. 36±0. 25b	3. 01±0. 25b	3. 35±0. 05a
	Cd	Straw	0. 16± 0. 01c	0. 15±0. 01c	0. 19±0. 029b	0. 23±0. 02a	0. 24±0. 02a
		Grain	0. 41±0. 08e	0. 57±0. 1d	0. 98±0. 06a	0. 79±0. 06c	0. 96±0. 13b
		Leaf	0. 79±0.06b	0. 87±0. 09b	0. 79±0. 09b	0. 98±0. 08a	0. 91±0. 07a
CM	Mn	Straw	4. 24± 0. 24b	4. 06±0. 46b	4. 08±0. 19b	4. 76±0. 26a	4. 87±0. 49a
		Grain	2. 97±0. 17b	3. 40±0. 30a	3. 43±0. 29a	3. 47±0. 21a	3. 410. 09a
		Leaf	33. 95±3. 67b	35. 60±0. 94a	35. 86±0. 42a	36. 20±0. 21a	37. 23±0. 04a
	Cu	Straw	3. 86±0. 25b	4. 08±0. 01a	4. 46±0. 66a	4. 57±0. 97a	4. 26±0. 29a
		Grain	1. 01±0. 28b	1. 25±0. 25b	1. 66±0. 26b	1. 71±0. 04a	1. 59±0. 15a
		Leaf	2. 54±0. 42b	3. 25±0. 37a	3. 28±0. 16a	3. 26±0. 04a	3. 19±0. 05a
	Zn	Straw	28. 86±2. 53d	56. 45±5. 22b	49. 64±0. 07c	54. 32±1. 43b	67. 33±0. 06a
		Grain	12. 18±1. 34a	11. 75±0. 19a	12. 56±1. 49a	11. 51±0. 15a	9. 85±0. 98a
		Leaf	33. 80±1. 88b	30. 22±1. 37b	39. 71±3. 61a	37. 50±7. 84a	24. 94±4. 07c
	Ni	Straw	1. 43±0. 29b	1. 35±0. 33b	1. 69±0. 54b	2. 50 ±0. 57a	2. 57±0. 03a
		Grain	1. 45±0.03b	1. 42±0.57b	1. 64±0. 13a	1. 59±0. 19a	1. 98±0. 32a
		Leaf	1. 21±0.27c	1. 53±0.03c	1. 31±0.01b	1. 65±0.17a	1. 61±0. 41b
	Cd	Straw	0. 14±0. 03b	0. 14±0. 02b	0. 15±0. 01b	0. 16±0. 02b	0. 18±0. 01a
		Grain	0. 4±0. 1b	0. 43±0. 01b	0. 39±0. 12b	0. 49±0. 05a	0. 51±0. 07a
		Leaf	0. 39±0. 07c	0. 49±0. 05b	0. 53±0/08a	0. 57±0. 05a	0. 6±0. 01a

The lowercase letters (a, b, c, d, and e) represents the significance of data. Values followed by the same lowercase letter within each column are not significantly different by Duncan's multiple range test at the level of P<0.05.

*** represents the significance at P<0.001

** represents the significance at P<0.01

*represents the significance at P<0.05

Table 4 The correlation between the BRs of HMs and SOM, pH, and salinity in three OA treatments and the mixed data of three OAs.

		Mn	Zn	Cu	Ni	Cd
SS	SOM	NS	0.987**	0.974**	0.730	0.985**
	pH	NS	-0.977**	-0.967**	NS	-0.961**
	salinity	NS	NS	NS	NS	NS
CMR	SOM	NS	NS	NS	NS	0.961**
	pH	0.532**	-0.549**	-0.645**	-0.562**	0.562**
	salinity	NS	NS	NS	NS	-0.948**
CM	SOM	NS	NS	NS	NS	0.976**
	pH	NS	0.934**	NS	NS	-0.967**
	salinity	-0.918**	-0.974**	NS	NS	-0.897**
Mixture	SOM	NS	0.972**	0.996**	NS	0.995**
	pH	NS	-0.974**	-0.973**	NS	-0.978**
	salinity	-0.946**	-0.974**	-0.924**	NS	-0.946**

** represents the significance at P<0.01

*represents the significance at P<0.05

NS represents no correlation

Table 5 The correlation between available HMs in amended soils and the HM concentration in aboveground tissues (straw, grain, and leaf).

		Mn	Zn	Cu	Ni	Cd
SS	straw	NS	0.896*	0.949*	NS	NS
	grain	NS	0.924*	NS	0.959*	NS
	leaf	NS	0.922*	NS	NS	0.949*
CMR	straw	NS	0.966*	NS	0.881*	NS
	grain	NS	NS	0.952*	0.927*	NS
	leaf	NS	NS	NS	0.945*	NS
CM	straw	0.929*	NS	NS	NS	NS
	grain	NS	NS	NS	NS	NS
	leaf	NS	NS	NS	NS	0.895*
mixture	straw	0.925*	0.920*	0.936*	0.941*	0.940*
	grain	0.937*	NS	0.960**	0.942*	0.913*
	leaf	0.962**	0.950*	0.980**	0.945*	0.974**

** represents the significance at $P<0.01$

*represents the significance at $P<0.05$

NS represents no correlation

Figures

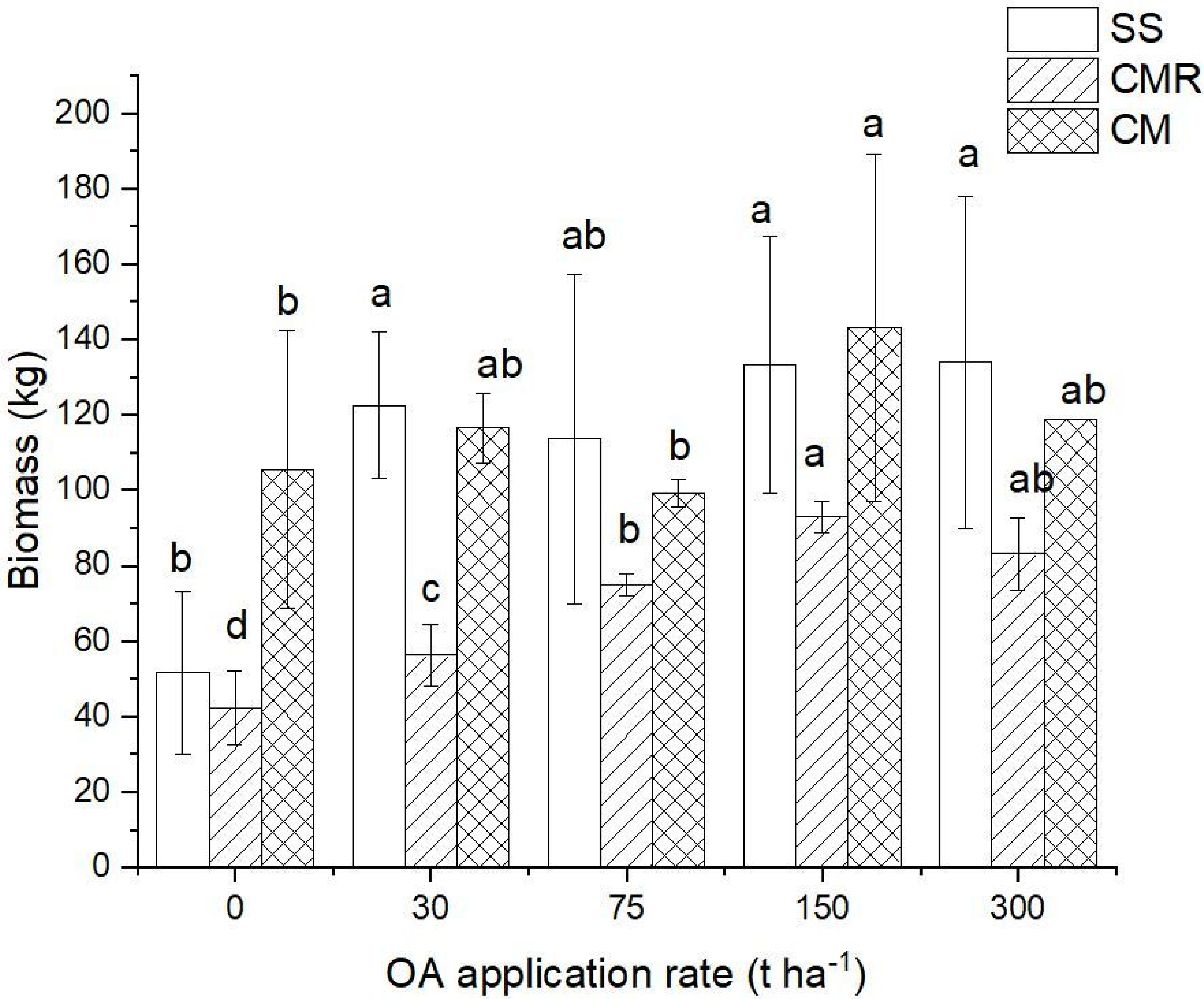

Figure 1

The effect of the OA application rate on the biomass of maize. The values of y are the mean±SD of three replications. Similar letters indicate no significant difference under Duncan’s test at the level of P<0.05.

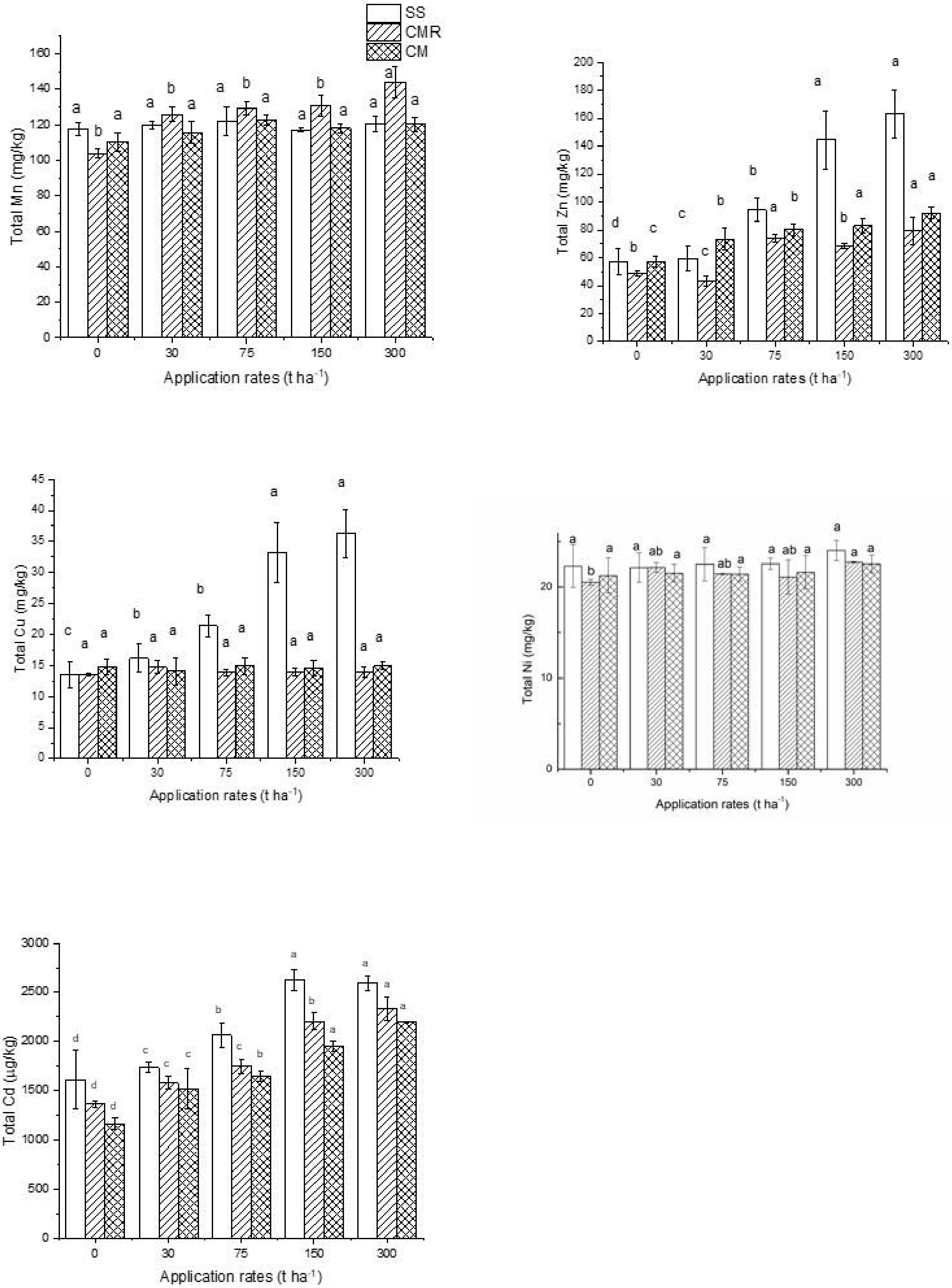

Figure 2

The effect of OA application rate on soil total HMs. The values of y are the mean±SD of three replicates. Similar letters indicate no significant difference under Duncan's test at the level of P<0.05.

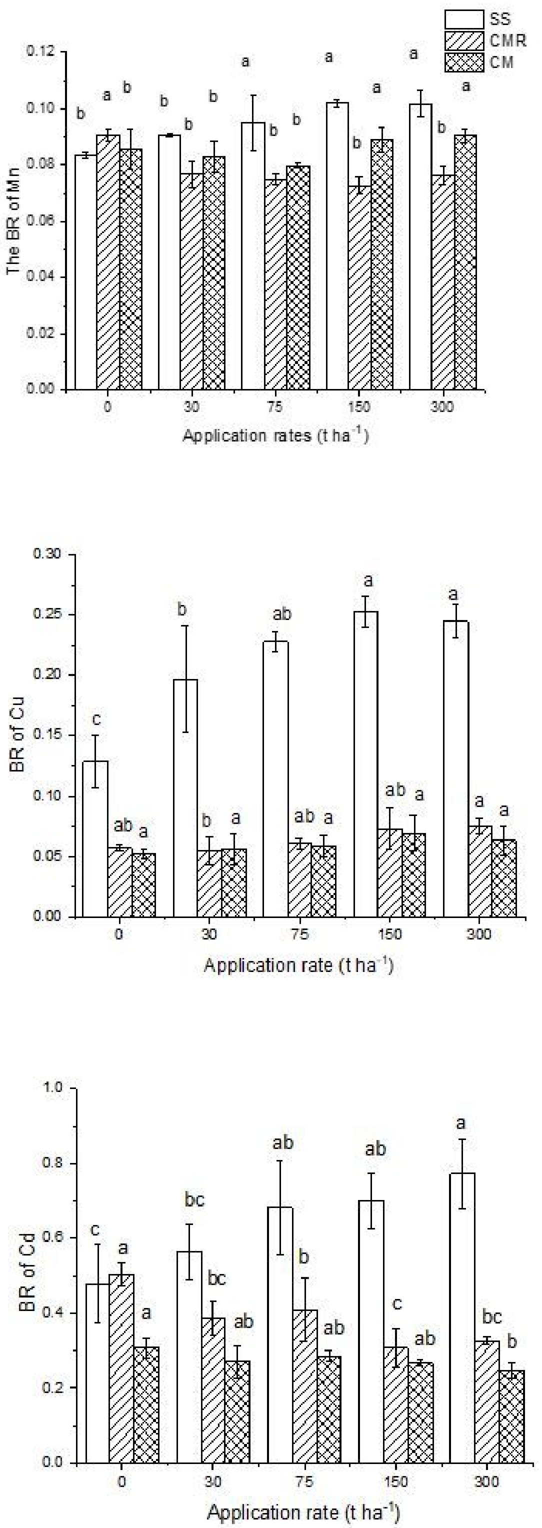

Figure 3

The effect of OA application rate on the BR of HMs. The values of y are the mean±SD of three replicates. Similar letters indicate no significant difference under Duncan's test at the level of P<0.05.

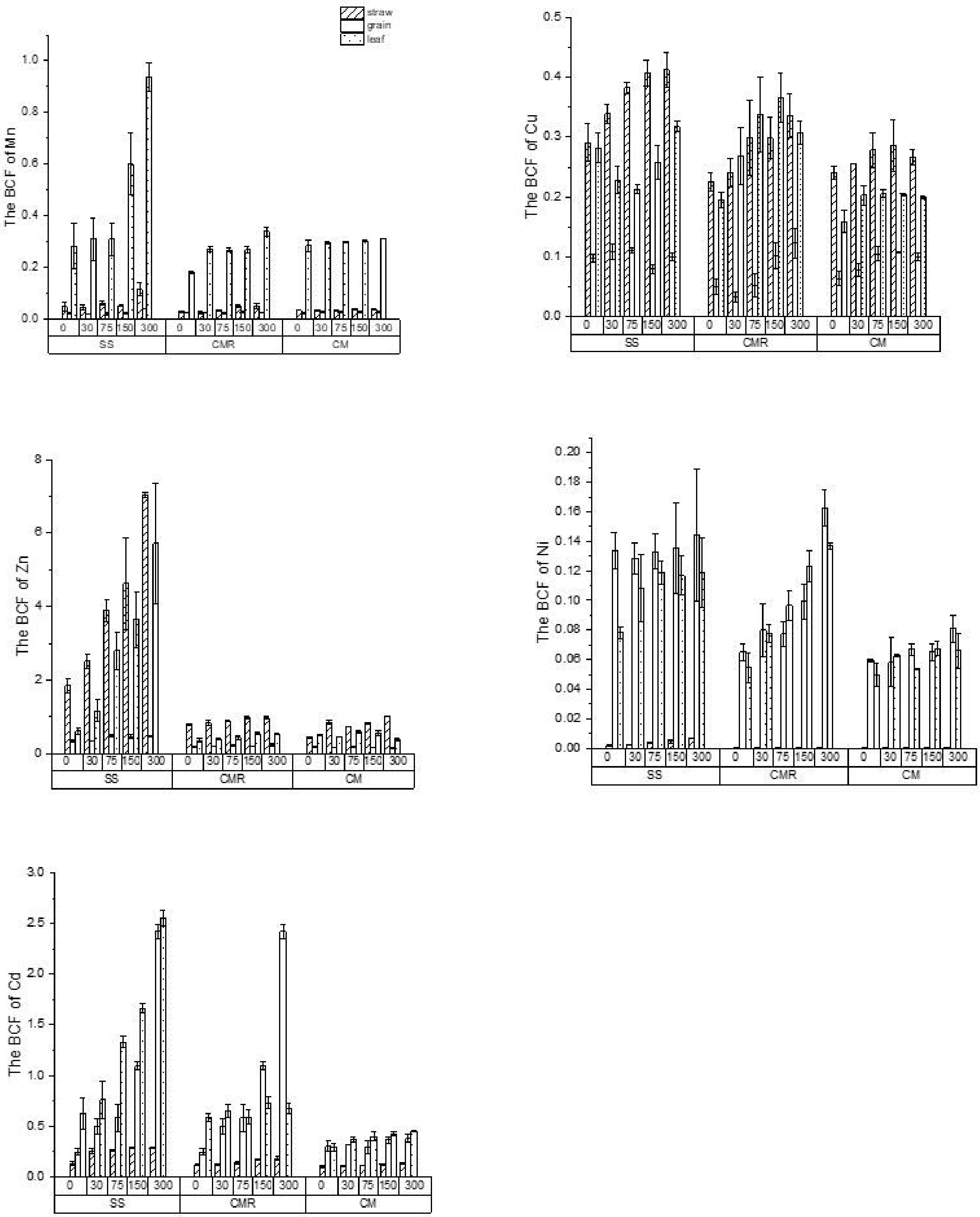

Figure 4

The effect of OA application rate on the BCFs of Cu, Mn, Zn, Ni, and Cd in maize. The values of y are the mean±SD of three replications.

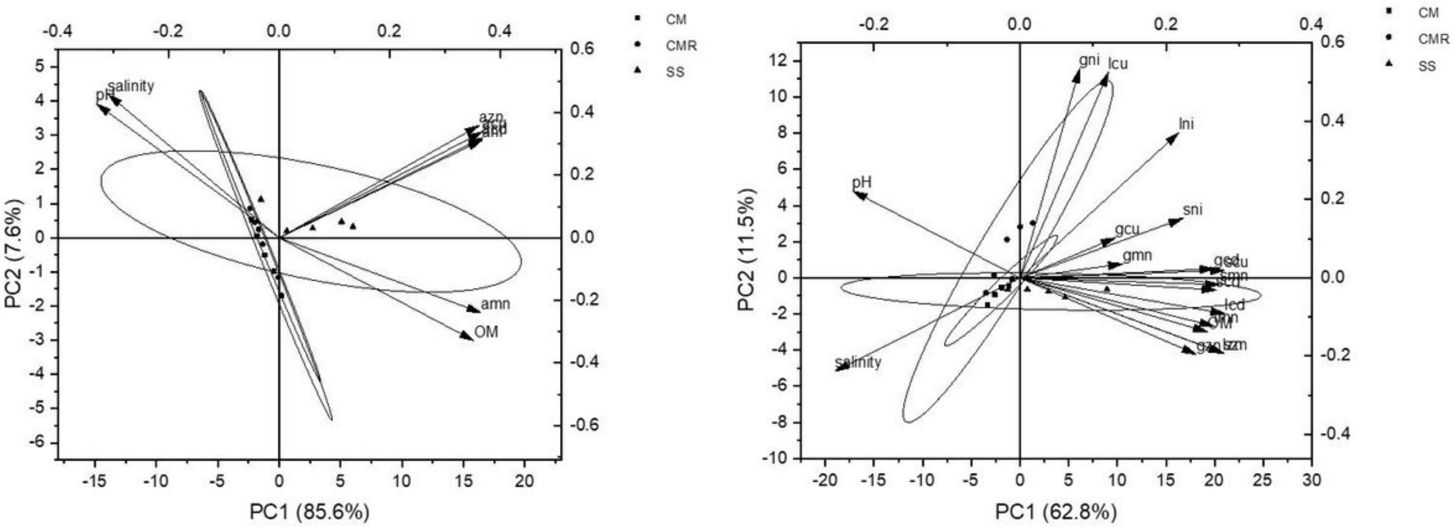

Figure 5

Principal component analysis (PCA) of soil properties (SOM, pH, and salinity), A) Soil bioavailable HMs, and B) HM concentrations in maize tissues. The spots represent five different application rates. The arrows indicate the loadings of explanatory variables.