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Incorporation of Mg-modified zeolite in municipal solid waste compost reduces heavy metal concentration in soil and corn plant

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Abstract

Heavy metals in municipal solid waste (MSW) composts mostly result in soil contamination if no appropriate management is conducted before the land application. In order to decrease the concentration of Pb, Cu, Mn, Zn, Ni and Cd in compost and as a result reduce the heavy metals uptake by corn plant, three rates including 5%, 10% and 15% (weight basis) of natural and Mg-modified zeolites were applied to the MSW composting process. A greenhouse experiment was done in order to investigate potential effects of composts containing natural zeolite- and Mg-modified zeolites- (CNZ and CMZ, respectively) on the distribution of metals in corn plant. Furthermore, accumulation of available metals in soil and plant effect on biomass production was studied. Pb, Ni, Cu, Zn, Cd and Mn in CNZ-amended soil decreased by 11%, 16%, 8%, 14%, 11% and 14% as comparison to zeolite-free compost, respectively, while in CMZ treatments, the decreases were 16%, 21%, 16%, 26%, 22% and 17%, respectively. Amended soils with compost containing Mg-modified zeolite decreased the heavy metal concentrations, up to 39%, 61% and 62%, in the roots, stalks, and leaves of corn, respectively, compared to zeolite-free compost. Compost containing zeolite (particularly it Mg-modified forms) resulted a decrease of 30 to 60 % in the Pb, Cu, Mn and Ni bioaccumulation factor in all parts of corn. The amount of dry shoot weight in CNZ- and CMZ-amended soils were 40% and 56% higher than those of soil amended with zeolite-free compost.

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Introduction

Composting is an impressive way to change municipal solid waste (MSW) into fertilizer. Numerous studies have shown the application of stabilized MSW compost in agriculture is beneficial for environment, soil, and the crops (Achiba *et al.*, 2009; Baldwin & Shelton, 1999). The application of organic wastes as soil amendments is vital in view of the fundamental declining in the organic matter content of many soils, especially in dry areas (Hernandez *et al.*, 2015). Nonetheless, toxic matters such as heavy metals in the compost mostly result in soil contamination if no appropriate management is conducted before land application (Achiba *et al.*, 2009; Chen *et al.*, 2010; Hargreaves *et al.*, 2008; Yuksel, 2015). Heavy metals can be absorbed by plants and pollute the food chain and menace human health (Hargreaves *et al.*, 2008; Jardao *et al.*, 2006).

In order to decrease the solvability and bioavailability of heavy metals in compost, chemical stabilization can be carried out by adding some non- noxious compounds to the composting materials (Wuana *et al.* 2010). Nowadays, zeolite is attracted extremely attention in the scientific society and is being vastly used in industry, contamination control and agriculture (Wen *et al.*, 2016). In many applications, different modification processes were carried out to improve the sorption capacity of natural zeolite. The ordinary amendment techniques include treatments with acid, and salts of alkaline metals (Huang *et al.*, 2014). Modified zeolite with $MgCl_2$. $6H_2O$ salt has been displayed to adsorb more heavy metals than natural zeolite. Multivalent cations, which it can be in the form of a chloride salt and/or a sulfate salt such as $MgCl_2$ and $MgSO_4$, improve the adsorption rates of the zeolites, because heavy metals stick to the mg-modified zeolite. Thus, a large energy might be needed to break them (Choi *et al.*, 2016).

As previously mentioned, numerous studies have reported natural zeolite improves compost quality (Singh & Kalamdhad, 2014; Stylianou *et al.*, 2008; Villasenor *et al.*, 2011; Zorpas *et al.*, 2000). Nevertheless, limited information is accessible on effect of Mg-modified zeolites on the composting materials for stabilization of heavy metals. In addition, data on the effects of MSW compost containing natural and Mg-modified zeolite on metals distribution in the root, stalk and leaf of corn plant are rare. Therefore, this study focused on determining the effects of using MSW compost containing natural and Mg-modified zeolite on: (i) concentration of heavy metals in compost and soil, (ii) accumulation and capability of translocation of metals from roots to other parts of plant, (iii) plant biomass.

Materials and methods

Materials preparation

Natural zeolite

The natural zeolite applied in this study was obtained from a mine in Semnan province, Iran. The natural zeolite has a chemical content of: SiO_2=65.90, Al_2O_3=11.20, K_2O=2.31, Na_2O=2.10, CaO=3.20, Fe_2O_3=1.25, MgO=0.52, LOI=11.89 in weighted percentage, and SiO_2/Al_2O_3=5.9 (Malekian *et al.*, 2011). X-ray diffraction technique (XRD) was used for characterization of the zeolite sample. X-ray pattern exhibited that the sample consisted mostly of clinoptilolite with an ideal formula of KNa_2Ca_2 $(Si_{29}$ $Al_7)$ $O_{72}.24$ H_2O and quartz (SiO_2) (Fig.1). Zeolite with particle size smaller than 250 μm was collected using ASTM standard sieves for further tests.

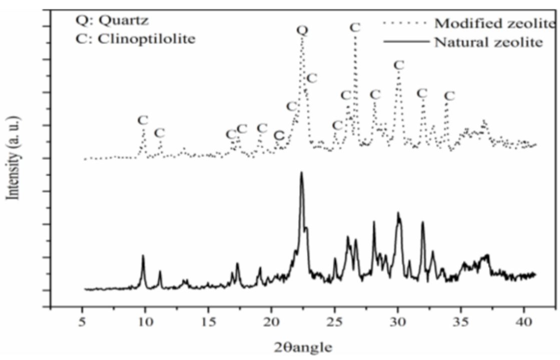


Fig. 1. XRD patterns of the Mg-modified and natural zeolites.

Modified zeolite

Mg-zeolite was produced by modifying natural zeolite with $MgCl_2$. $6H_2O$ salt (Huang *et al.*, 2014). Before the modification process, natural zeolite was washed with distilled water in order to remove soluble impurities and dried at ambient condition for 36 hours. Subsequently, 15kg of air dried zeolite was suspended in a 150-L of 2 M ($MgCl_2$. $6H_2O$) solution. The suspension was mixed for five minutes with 30 min interval for 48 hours, then decanted and washed. Lastly, the modified zeolite was dried at an ambient temperature. Mg-modified zeolite was characterized using XRD technique. A near correlation was apperceived among diffraction patterns of the Mg-modified zeolite and the corresponding natural zeolite, which ascertain the structural integrity of zeolite after modification (Fig. 1).

Composting setup, operation, and analysis

Compost production was performed at the Isfahan Compost Factory. Organic residues of MSW were separated from inorganics to be used for composting process. Particle size of the selected materials was less than 60mm. The organic wastes mixed with the natural and Mg-modified zeolites at different ratios and the composting process was performed for two months. The composts were mixed manually, once every 3 days, throughout the composting period. For this purpose, 21 PVC lysimeters with 60 cm height and 40cm internal diameter were used. At the starting of the composting process, different rates of 5%, 10%, and 15% of the natural or Mg-modified zeolite were added to the MSW and the composting process was carried out for 2 months.

The composts were mixed manually, once every 3 days, throughout the composting period. After maturing, the composts were passed through 6 mm ASTM standard sieves prior to being used in soil amendment studies.

Establishing of the treatments and planting

This part of the study was conducted at the research greenhouse of Isfahan University of Technology, Isfahan, Iran during April to June 2016. The soil texture was a sandy loam containing 80.5% sand, 17.4% silt, and 2.1% clay. Some of the soil characteristics are listed in Table 1. The lysimeters were PVC columns with 25.5cm diameter, 60cm length, and a drain at the bottom. Experiments are conducted in a completely randomized design with three replications. The treatments consisted of 7 types of soil amendments as showed in Table 2.

Table 1. Initial soil characteristics used in the experiment.

Characteristic	EC (1:2) (dS/m)	pH(1:2)	CEC ($cmol_c$/kg)	Oranic Carbon (%)	Total Nitrogen (%)	Nitrate (mg/kg)
Amount	0.38	8	14.95	1.03	0.036	62.5

Table 2. Experimental treatments in the study.

Abbreviations	Treatments	Ratio of zeolite (%) in composting materials
Co	Soil with no amendment	0
C	Compost containing no zeolite+soil	0
CNZ5	Compost containing Natural zeolite+soil	5
CNZ10	Compost containing Natural zeolite+soli	10
CNZ15	Compost containing Natural zeolite+soli	15
CMZ5	Compost containing modified zeolite+soli	5
CMZ10	Compost containing modified zeolite+soli	10
CMZ15	Compost containing modified zeolite+soli	15

The amount of amendment applied to the soil was determined based on 40-Mg h^{-1} in all treatments except Co. The amendments were mixed to the soil surface of the lysimeters (0-15cm) before planting. Corn seeds, single cross 704 cultivar, were planted in 3cm depths in all the lysimeters. Corn was thinned to two plants per lysimeter after 22 days.

Soil moisture in each lysimeter was monitored in three depths (7.5cm, 22.5cm, 37.5cm) using a SM150 Soil Moisture Sensor. The irrigation performed based on 55% depletion of available soil moisture. After harvesting (62 days after germination) soil samples were collected at depths of 0–15 and 15–45cm. The air-dried samples were passed through a 2 mm sieve.

The DTPA extractable Zn, Cd, Cu, Ni, Pb, Mn, and Cr were measured using atomic absorption spectrometry technique (AAS) with air-acetylene, fuel (Lindsay & Norvell 1978). Leaves, stalks, and roots were harvested and dried at 70°C, and then ground by a stainless steel mill to pass through a 1 mm sieve. The samples of plant analyzed for the heavy metals content by AAS. Bioaccumulation factors (BAFs) were calculated as following ratios:

$$BAF_r = \frac{C_r}{C_{soil}} \quad (1)$$

$$BAF_s = \frac{C_s}{C_{soil}} \quad (2)$$

$$BAF_l = \frac{C_l}{C_{soil}} \quad (3)$$

Where C_s, C_r, C_l and C_{soil} are concentration of metals in the stalk, root, leaves and soil, respectively (Carbonell *et al.*, 2011).

Two translocation factors (TF) were introduced as following ratios:

$$TF_s = \frac{C_s}{C_r} \quad (4)$$

$$TF_l = \frac{C_l}{C_s} \quad (5)$$

Where C_s, C_r and C_l are concentration of metals in the stalk, root and leaves, respectively (Bose & Bhattacharyya 2008).

Statistical analyses

The data were analyzed using analysis of variance (ANOVA) procedure and the means comparison test were done by duncan's test means at $p < 0.05$ probability level (SAS Institute 2000). In all tables, mean values denoted by same letters are not significantly different ($p < 0.05$).

Result and discussion

DTPA-extractable heavy metals in composts

The concentration of DTPA-extractable metals in mature composts that is shown in Table 3 follows the trend of Fe >Zn >Cu >Pb > Mn> Ni >Cd. Chromium (Cr) in the all treatments was less than the AAS detection limit (0.08mg/kg). Application of natural and Mg- modified zeolite leads to a decreased in DTPA-extractable metals in matured composts (Table 3). Application of natural zeolite in composting process decreased the DTPA-extractable of Pb, Cu, Zn, Mn, Ni and Cd in matured composts by 13%, 44%, 22%, 32%, 28% and 29% in comparison to zeolite-free compost, while in modified zeolite treatments, the decreases were 14%, 47%, 25%, 36%, 43% and 36%, respectively. Mg- modified zeolite was more effective than natural zeolite to reduce DTPA-extractable heavy metals because of higher ionization potential/electronegativity of Mg^{2+} in compare to Na^+ and K^+ (Choi *et al.*, 2016).

Table 3. DTPA-extractable concentration of metals in different matured composts (mg kg^{-1}).

Compost	Cd	Mn	Fe	Zn	Cu	Pb	Ni
C	1.50 a	36.66 a	615.62 a	150.02 a	54.62 a	42.62 a	8.66 a
CNZ5	1.29 b	28.37 b	443.75 b	140.62 ba	36.25 b	40.12 a	7.50 ba
CNZ10	1.05 c	25.01 cb	409.37 cb	125.00 ba	33.01 cb	38.37 a	6.87 bc
CNZ15	0.95 c	21.00 ed	309.37 d	84.37 c	22.25 ed	32.51 b	4.25 e
CMZ5	1.01 c	23.25 cd	393.75 c	137.50 ba	37.50 b	42.51 a	6.25 bc
CMZ10	1.00 c	21.10 ed	363.54 c	112.50 bc	28.12 cd	38.25 a	5.87 dc
CMZ15	0.96 c	18.79 e	312.50 d	87.50 c	19.87 e	31.25 b	4.62 de

In each column, the different letter shows significant different by Duncan's test at $p \leq 0.05$.

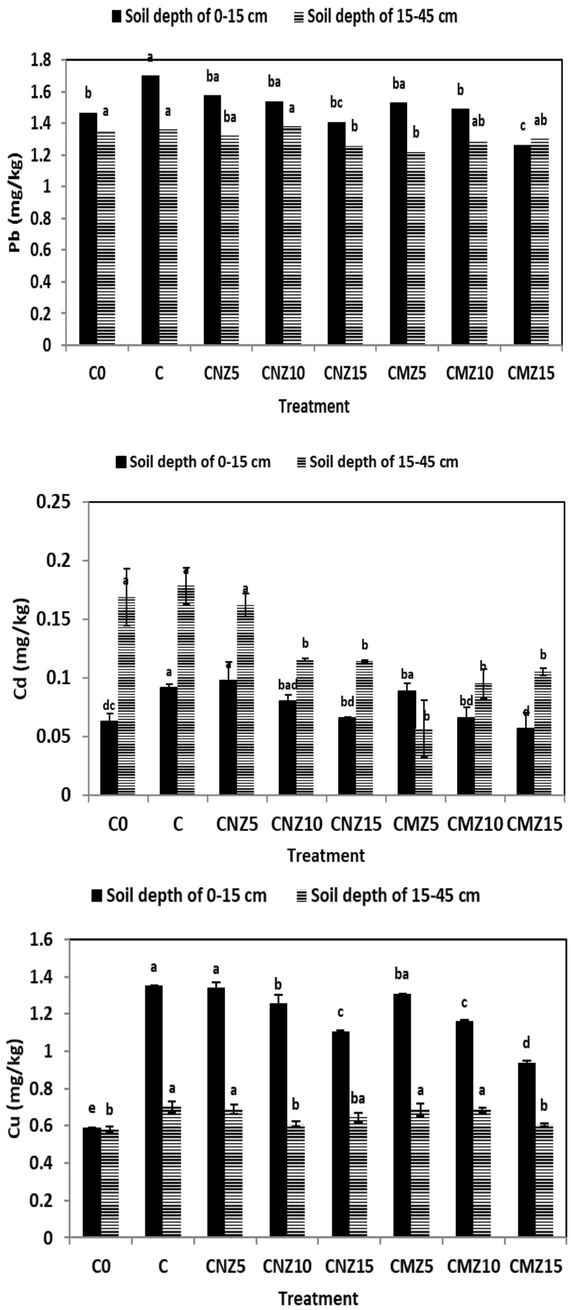
DTPA-extractable metals in soil

Fig. 2 shows the DTPA extractable metal concentrations in soils after corn harvest. Application of MSW compost significantly increased DTPA extractable Cu, Pb, Mn, Ni, and Zn concentrations in the 0-15 cm soil. Cadmium concentration in the 15-45 cm depths of soil increased.

However, in natural and modified zeolite compost treatments, Cd and Ni were concentrated in the depth of 0-15 cm and did not move to the lower depth as compared to Co and C treatments. The concentration of metals in CNZ- and CMZ- amended soils was less than the other treatments.

Increasing the zeolite dosage and using modified zeolite in applied compost significantly reduced the availability of heavy metals in soil.

Thus, the minimum values of bioavailable metals were recorded in the soils treated with compost containing 15% zeolite specially CMZ15. Zeolites are applied for stabilization of heavy metals because of their unique pore characteristics, excellent ion exchange ability, and high particular surface area (Castaldi *et al.*, 2005; Koshy & Singh, 2016; Wen *et al.*, 2016).



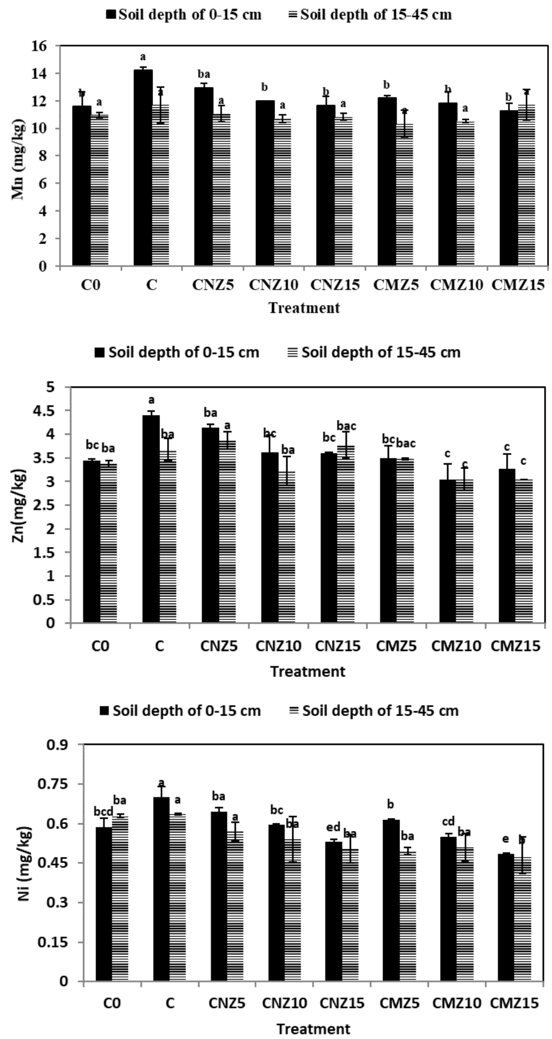
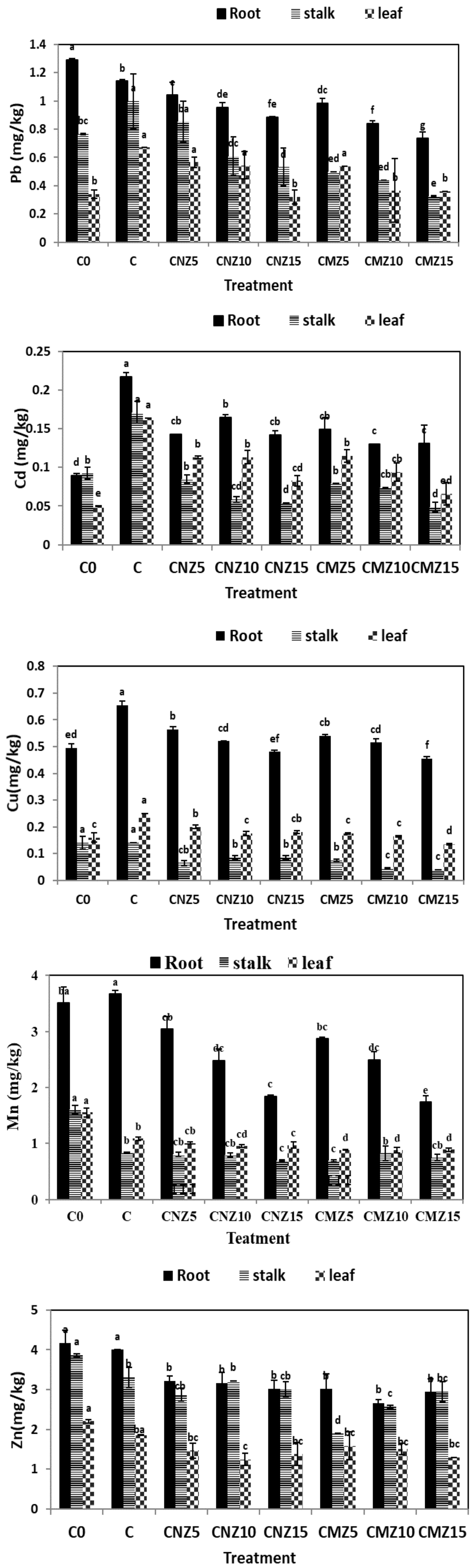


Fig. 2. DTPA extractable metal concentrations in different treatments For each soil depth, bars the different letter shows significant different ($p<0.05$).

Distribution of heavy metals in different parts of corn plant

The concentrations of heavy metal in root, stalk, and leaves of corn plants are shown in Fig. 3. remarkable differences in heavy metal concentration were found among the treatments. Increasing the zeolite dosage and using modified zeolite in applied compost significantly reduced most of the heavy metals concentrations in corn roots, stalks and leaves compared to C treatment (Fig. 3).

Thus minimum values of metals concentration in corn plants were recorded in CMZ15 treatment. the zeolitic material has an extraordinary exchange capacity that could decrease the solvable metal concentration (Querol *et al.*, 2006).



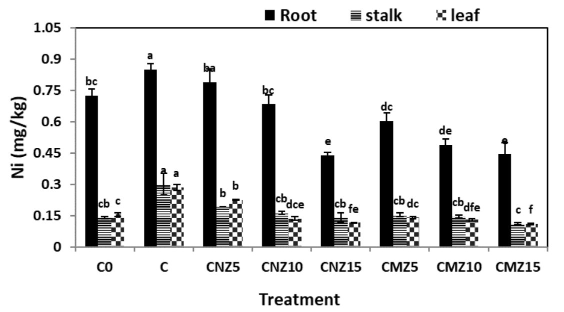


Fig. 3. Metal concentrations (mg kg^{-1} d.w.) in root, stalk and leaf of the corn plants grown in the control and the amended soils. Different letters for each plant tissue indicate a significant difference at $p < 0.05$.

Bioaccumulation in different parts of plant

The bioaccumulation factors (BAF) for different parts of plant at the harvesting time are shown in Table 4. Among the soil amendments, composts containing Mg-modified zeolite was more effective than natural zeolite and zeolite-free compost to reduce BAF_r, BAF_s and BAF_l of Pb, Cu, Mn and Ni. Zeolite takes up most of the heavy metals, that are bounded to the most mobile forms while the rest remain to an inert form so the BAF is reduced (Zorpas, 2011). For Cd, BAF_r in all treatments, BAFs in C and Co treatments and BAF_l in all treatments except Co were more than one, illustrating that Cd is distributed in the plant aerial parts and adding compost containing zeolite reduced the BAFs. With increasing the zeolite in CNZ and CMZ treatments, BAF_r for Cd increased but BAF_l and BAF_s decreased. It showed that zeolite prevented the bioaccumulation of Cd in plant aerial parts. For Zn BAF_r, between the C, CNZ and CMZ treatments were not significantly different, while with increasing the zeolite dosage in CMZ treatment, BAFs was increased, and BAF_l in CMZ treatment was more than that in C and CNZ treatments.

Metals translocation

Translocation factors (TF) for different parts of plant at the harvesting time are shown in Table 5. Metal immobilization is considered in plant roots whenever TFs is less than 1. The results showed TF_s <1 for all metals except Cd, offering a little translocation from roots to plant aerial parts. With adding zeolite specially modified form in compost, TF_s of Pb, Cu, Cd and Ni decreased significantly as compared to zeolite-free compost, while zeolite in applied compost caused a significant increase in TF_s of Zn that is useful for plants growth. For Mn and Zn, with increasing the zeolite dosage in CNZ and CMZ treatments, TF_s was increased.

Compost containing zeolite decreased the TF_l of Mn, Zn and Ni in comparison with the zeolite-free compost. For Pb were not significant difference in the fraction of TF_l between the C, CNZ and CMZ treatments.

Table 4. Bioaccumulation factors (BAF) in some parts of the corn plant grown in different treatments.

	Co	C	CNZ5	CNZ10	CNZ15	CMZ5	CMZ10	CMZ15
					BAF_r			
Pb	0.88 a	0.67 b	0.66 b	0.62 cb	0.63 cb	0.64 cb	0.57 c	0.59 c
Cd	1.48 a	2.39 b	1.55 a	2.06 b	2.17 b	1.73 b	2.05 b	2.83 b
Cu	0.84 a	0.49 b	0.42 c	0.42 c	0.43 c	0.41 c	0.44 c	0.48 b
Mn	0.32 a	0.32 a	0.28 ba	0.23 b	0.17 bc	0.28 ba	0.24 b	0.15 c
Zn	1.21 a	0.91 b	0.78 b	0.91 b	0.84 b	0.86 b	0.90 b	0.90 b
Ni	1.25 a	1.23 a	1.23 a	1.15 ba	0.83 c	0.99 b	0.89 bc	0.92 b
					BAF_s			
Pb	0.52 ba	0.54 a	0.52 ba	0.40 bc	0.38 c	0.36 c	0.33 c	0.30 c
Cd	1.49 b	1.89 a	0.90 cd	0.73 d	0.83 cd	0.90 cd	1.17 cb	0.91 cd
Cu	0.24 a	0.10 b	0.05 c	0.07 cb	0.08 cb	0.06 c	0.04 c	0.04 c
Mn	0.15 a	0.08 b	0.08 b	0.08 b	0.07 b	0.07 b	0.08 b	0.07 b
Zn	1.12 a	0.75 cb	0.70 c	0.90 b	0.84 cb	0.71 c	0.86 cb	0.90 b
Ni	0.24 cb	0.43 a	0.29 b	0.27 cb	0.26 cb	0.25 cb	0.26 cb	0.23 c
					BAF_l			
Pb	0.23 cb	0.42 a	0.36 a	0.35 ab	0.23 cb	0.31 cab	0.22 c	0.25 cb
Cd	0.82 c	1.80 a	1.23 b	1.42 ba	1.27 b	1.33 b	1.43 ba	1.17 bc
Cu	0.27 a	0.19 b	0.15 c	0.14 c	0.16 cb	0.13 c	0.14 c	0.14 c
Mn	0.15 a	0.10 b	0.09 b	0.09 b	0.09 b	0.09 b	0.08 bc	0.07 c
Zn	0.64 a	0.42 cb	0.35 c	0.36 c	0.38 cb	0.44 cb	0.49 b	0.40 cb
Ni	0.27c	0.48 a	0.35 b	0.23 d	0.22 d	0.23 d	0.24 d	0.23 d

In each column, the different letter shows significant different by Duncan's test at $p \leq 0.05$.

Table 5. Translocation factors of metals in stalk (TF_s) and leave (TF_l) after harvesting.

	Co	C	CNZ5	CNZ10	CNZ15	CMZ5	CMZ10	CMZ15
					TF_s			
Pb	0.59 b	0.80 a	0.78 a	0.63 ab	0.60 ab	0.55 bc	0.58 b	0.51 c
Cd	1.04 a	0.79 b	0.59 cb	0.35 d	0.38 cd	0.53 cd	0.56 cbd	0.41 cd
Cu	0.28 a	0.21 b	0.11 ed	0.16 cbd	0.18 cb	0.14 ced	0.09 e	0.09 e
Mn	0.75 a	0.45 c	0.41 c	0.50 bc	0.58 ba	0.37 c	0.50 bc	0.66 ba
Zn	0.93 ab	0.83 b	0.89 ab	1.03 a	1.01 a	0.83 b	0.97 a	1.01 a
Ni	0.19 c	0.35 a	0.24 b	0.24 b	0.32 a	0.34 a	0.30 a	0.25 b
					TF_l			
Pb	0.44b	0.81 ba	0.72 ba	0.93 a	0.62 ba	0.88 a	0.64 ba	0.83 ba
Cd	0.55c	1.12 b	1.35 b	1.96 a	1.52 b	1.45 b	1.27 b	1.31 b
Cu	1.17c	1.79 cb	3.22 a	2.12 b	2.18 b	2.34 b	3.69 a	3.38 a
Mn	0.97c	1.30 a	1.24 ba	1.20 ba	1.39 a	1.27 ba	1.08 bc	1.19 ba
Zn	0.57ba	0.57 ba	0.52 ba	0.39 b	0.47 ba	0.61 a	0.58 a	0.44 ba
Ni	1.11a	1.12 a	1.18 a	0.83 b	0.88 b	0.90 b	0.91 b	1.01 ba

In each column, the different letter shows significant different by Duncan's test at $p \leq 0.05$.

Biomass yield

The biomass data are demonstrated in Fig. 4. The results clearly indicate that all compost treatments had capacity to increase the corn yield. The order of affirmative effect of the tested composts on plant growth were CMZ10> CMZ15> CNZ15> CMZ5> CNZ10> CNZ5> C> Co. In this regard, maximum dry matter production was obtained.

From the CMZ10% treatment. Type of zeolite (natural and modified) and content of zeolite in applied compost affected biomass yield. Plant dry weight was increased by 11.8% in modified zeolite application as compared to the natural zeolite. Using Mg-modified zeolite in compost may also increase Mg^{2+} level in topsoil that increased the nitrogen uptake and corn growth.

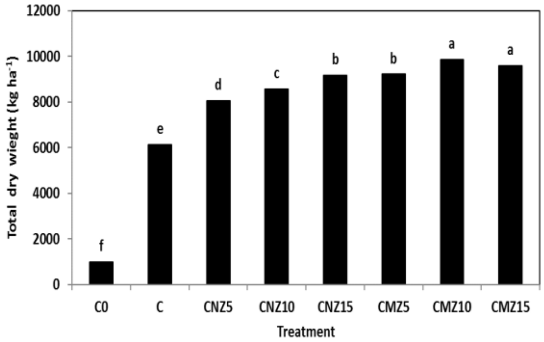


Fig. 4. Total dry yield (kg ha^{-1}) for all treatments. Bars with different letters shows significant different by Duncan's test at $p \leq 0.05$.

Conclusions

Application of natural zeolite in composting process decreased the DTPA-extractable of Cu, Pb, Zn, Ni, Cd and Mn in matured composts by 44%, 13%, 22%, 28%, 29% and 32% in comparison to zeolite-free compost, while in modified zeolite treatments, the decreases were 47%, 14%, 25%, 36%, 36% and 43% respectively. Pb, Cu, Zn, Ni, Mn and Cd in CNZ-amended soil decreased by 11%, 8%, 14%, 16%, 14% and 11% as comparison to zeolite-free compost, respectively, while in CMZ treatments, the decreases were 16%, 16%, 26%, 21%, 17% and 22%, respectively.

Amended soils with compost containing Mg-modified zeolite decreased the heavy metal concentrations by 39%, 61% and 62%, in the roots, stalks, and leaves of corn, respectively, as compared to zeolite-free compost. Compost containing zeolite (specially Mg-modified forms) caused a decrease of 30% to 60% in bioaccumulation factor of Pb, Cu, Mn and Ni cations in all parts of corn. With adding zeolite to compost, TF_s of Pb, Cu, Cd and Ni cations decreased significantly compared to zeolite-free compost. The amount of dry shoot weight in CNZ- and CMZ-amended soils were approximately 40% and 56% higher than those soil amended with zeolite-free compost.

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