

Assessing the Immobilization of Heavy Metals in Compost Derived from Organic Fraction of Municipal Solid Waste Amended with Forest-based Biochar

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Abstract

Biochar is characterized by a large specific surface area, porosity, and a large number of functional groups. All of those features causes that biochar can be a potentially good material in the optimization of the process of composting and final compost quality. The aim of this study was to assess the effect of biochar to reduce the bio-availability of heavy metals (HMs) during the composting of organic fraction of municipal solid waste (OFMSW) and to improve the end product quality. Small selected doses of biochar (1%, 3%, and 5% by weight of the OFMSW) were added to the compost piles, and compared with a control without any amendment. The results indicated that the biochar effectively reduced the bio-availability of HMs (Pb, Zn, Ni, Cd, and Cu) compared to control. The combine use of OFMSW + 5% biochar was significantly reduced the Cd content by 71.63% as compare to control. Also, biochar addition at 1% and 3% could reduce the Cd content in final product of the compost respectively by 55.82% and 70.24%. The addition of biochar (even low doses of 1%), lowered the concentrations of HMs to those recommended by compost quality standards. Therefore, the application of biochar as a beneficial additive can improve the quality of compost produced from the OFMSW and provide good conditions for the use of the produced compost as an enhancer of agricultural soil.

Keywords: Organic wastes, Composting, Biochar, Heavy metals.

Introduction

The increasing population growth and industrial development have led to an ever-increasing consumption and the resulting production of solid waste materials (Roman et al., 2021). This has led to the appearance of health and environmental issues particularly in urban environments (Vahidi et al., 2017), which can lead to grave crises in human societies if it is not properly managed (Prodana et al., 2019). As an alternative environmental technology, composting is being increasingly used to handle different kinds of organic waste and minimize environmental pollution (Gong et al., 2021). Composting can be a very effective and economic option for organic fraction of municipal solid waste (OFMSW) since it changes biological structure and eliminates the potential hazards of this waste, leading to a final product capable of improving

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the quality of soil (Akhavan Limoodehi et al., 2017; Ding et al., 2019). One of the main limitations of using the compost is the existence of heavy metals (HMs) (Fischer et al., 2018). HMs are naturally present in the environment, soil, and food and are used extensively in various production processes in the industry. Therefore, they can be present in the compost produced from municipal solid waste (MSW) (Adejumo et al., 2021). The sources of HMs in compost are abundant, e.g., batteries, plastics, dyes, inks, medications, and household insecticide sprays (Abou Jaoude et al., 2020).

The concentration of HMs during the composting process increases due to the microbial decomposition of the organic part and the removal of volatile metals (Yousaf et al., 2022). HMs can remain unchanged during the production stages of compost including the microbial phase and become released after entering the soil due to different factors such as rain, soil characteristics, and plant absorption (Zhang et al., 2021). The most common of these metals include cadmium (Cd), chromium (Cr), nickel (Ni), zinc (Zn), and lead (Pb). The concentration of which in compost varies from micrograms to milligrams in one kilogram of compost (Shapouri and Hassanzadeh Moghimi, 2018). Therefore, the use of compost in soil leads to the accumulation of HMs and nutrients, which in turn degrades the soil texture and finally the seepage of these metals to underground waters (Roohi et al., 2020). Hazards that these elements create for underground waters and ecosystem depend on the biological viability and movement of these elements through soil layers (Lima et al., 2022).

Features of HMs that make them particularly problematic are their non-biodegradable nature, toxicity potential even at low concentrations, and tendency to accumulate in the food chain, in which the human is the final consumer (Joseph et al., 2020). Exposure to HMs can lead to blood disorders, bone disorders, kidney damage, mental capacity reduction, and neurological damages (Kepa Izaguirre et al., 2020). Therefore, extensive research has been done to lower the concentration of HMs in compost made from MSW. In some of these research works, materials such as biochar (Huang et al., 2021), clinoptilolite (Graça et al., 2021), zeolite (Hao et al., 2019), and perlite (Chung et al., 2021) have been used to remove HMs from the compost, and their significant effect on the removal of HMs have also been reported. The primary characteristics of biochar are the surface area and porosity, as well as many other unique functions (Bello et al., 2021). All these characteristics enable biochar to serve as a potential effective material to reduce the mobility of HMs in the composting process and improve the final quality of the produced compost (Awasthi et al., 2021; Bashir et al., 2020).

Zhou et al. (2021) examined the effect of biochar on the reduction of ecological risk of HMs in compost and reported that the addition of biochar lowered the bioavailability of HMs of Zn and Cu by 22.9% and 38.6%, respectively, in the final compost product compared with that in the sample without biochar. In another research, Hao et al. (2019) investigated that variation of HMs' concentration during the composting process of poultry manure. The composting process was analyzed in two piles, the control pile (without the addition of biochar) and the pile containing 10% biochar. The results demonstrated that biochar considerably reduced Cu concentration by 90.3% compared with the control compost sample without the addition of biochar, while the reduction trend of Zn was lower, and the presence of biochar only led to 11.7% reduction in this metal. Moreover, Cui et al. (2020) studied the mobility and risks of HMs in compost and reported that adding 10% biochar to the compostable materials reduced heavy metals Zn, Cu, Cd, and Pb by 4.10%, 44.12%, 18.75%, and 30.06%, respectively.

The above findings require verification at specific biochar doses. It is also necessary to assess changes taking place in the pile of organic waste during the process under real conditions, as most existing research has been carried out in laboratory bioreactors. The concentration of HMs in MSW is of particular interest, especially when its compost is used as a fertilizer for soil (Černe et al., 2021). Therefore, an accurate investigation of the concentration of HMs present in compost is essential for regular supervision, risk assessment, and also ecological balance

protection (Cooper et al., 2021). The main novelty of this work consists in studying the impact of a biochar in small doses on the OFMSW composting process under real conditions. The aim of the research was to assess whether and how the biochar application at different doses affected the bio-availability of heavy metals during the composting process of OFMSW (in full scale) and the quality of the final product.

Material and Methods

The description of a study area

In this study, the process of compost production from the OFMSW was conducted under real conditions in the compost plant of the Babol Municipality (CPBM), Mazandaran province, Iran. With a population of over 500 thousand, the city of Babol is considered one of the largest and most populous cities in northern Iran. Currently, around 250 tons of MSW is collected in this city each day, and the per capita waste production for every Babol resident is 820 g per day (Babol Municipality, 2020). Table 1 shows the quantitative and qualitative analysis of MSW produced in Babol. Based on the Table 1, more than 65% of waste produced in Babol is composed of degradable waste, which the ability to be converted into compost. CPBM employs the mechanized processing technology, fermentation under a roofed space, and screw mixer to convert the OFMWS of different districts of Babol and the surrounding villages to compost with the aim of mitigating environmental pollution and improving the waste disposal method (Babol Municipality, 2020).

Table 1. Quantitative and qualitative characteristics of MSW produced in Babol (Babol Municipality, 2020)

Waste component	Composition (%)	Waste component	Composition (%)
Organic matter	65.3	Glass	1.2
Paper and cardboard	8.7	Ferrous metals	1.8
PET	1.1	Non-ferrous metals	2.3
Rubber	0.5	Wood	1.2
Plastic	7.3	Other	9.5
Textile	1.1	Total	100

Biochar preparation and determination of physical and chemical characteristics

The biochar used in this study was purchased from Beshel Biochar Facility which is located in Mazandaran, Iran. The biochar was produced from a mix of forestry residues (including the wood waste of maple, beech, and alder trees) using the pyrolysis method at 700 °C during 8 hours. Table 2 shows the physical and chemical properties of the biochar which were supplied by the manufacturer. Carbon (C) is the most important constituent of various biochars. In this regard, considering a high percentage of C in the biochar sample used in this study (76.1%), it was expected that this material would have a good performance during the research process. The low amount of nitrogen (N) in the biochar (0.71%) occurs due to nitrogen losses in the form of ammonia and/or nitrogen oxides during the carbonization process. In addition, the specific surface area of the biochar sample used in this research was 340 m²/g, indicating its porous structure.

The Composting Process

The duration of the composting process was nine weeks (63 days), starting in Saturday, June 12, 2021 and ending in Friday, August 13, 2021. During the composting process, the weather

condition of the research location was stable, and since the processing, fermentation, and final production halls of CPBM were roofed, the compostable materials were not exposed to rain, moisture from rainwater accumulation, or other environmental and weather phenomena. The produced biochar from forest residues was added at 0% (control sample), 1%, 3%, and 5% by weight to the piles of materials (windrows with a width and height of 1 m and a length of 3 m) inside the roofed fermentation halls of the CPBM (Fig. 1). Table 3 gives the name and combination of different compostable materials. In the thermophilic phase of composting (first three weeks), the materials were aerated periodically with an air current rate of 0.2 m³/min. The mixing of the compostable materials during the research period was performed using a box turner, and aeration was achieved using fans present in the fermentation hall. Temperature feedback was scanned using an automatic control system. The aeration was performed continuously in the first 12 hours, after which the system was turned on at 65 °C and turned off at 60 °C. To control this range, a programmable logic controller (PLC) was used. After the completion of the composting period at the end of week nine, the compostable materials were transferred to the final processing hall (final processing) and sifted and prepared using a trommel screen (with 20-mm mesh), vibrating screen, and glass separator.

Table 2. Physicochemical properties of the biochar used in the compost production process

Parameters	Unit	Biochar
Pyrolysis temperature	°C	700
Pyrolysis time	hour	8
Particle size	mm	1.18-2.36
pH	-	8-8.5
C	%	76.1
Moisture	%	6
N	%	0.71
Specific surface area	m ² /g	340



Figure 1. Composting windrows inside the roofed fermentation hall of the CPBM

Table 3. Feedstock ratio of compost piles at setup

Pile	Feedstock ratio (percent by weight)
BC0	OFMWS + 0% biochar
BC1	OFMWS + 1% biochar
BC3	OFMWS + 3% biochar
BC5	OFMWS + 5% biochar

Sampling and experimental analyses

Sampling the compost piles was conducted using the mixes sampling at two different times, the beginning and end of the compost production process (days 0 and 63). Before and after each sampling stage, the pile of compostable materials was completely blended and mixed using a loader.

The sampling at each point was performed at three different depths, a sample from the surface, a sample from the center, and a sample from the middle depth between the surface and center of the pile. Then, the collected samples were placed in a clean plastic bucket. After the end of sampling, the content of each bucket was completely blended, such that no layer was formed based on the particle size. Altogether, 24 samples, including 6 samples of control compostable materials (without the addition of biochar) and 18 samples of compostable materials combined with different doses of biochar, were collected and immediately transferred to the laboratory. After the preparation, measurement of pH, moisture content (MC), bulk density, organic material (OM), C, N, and HMs concentration was performed for control samples that had been collected at the beginning of the composting process (day 0). In the remaining samples (those containing different doses of biochar), the concentrations of HMs were also determined.

To measure pH, samples were extracted at room temperature (25 °C) with water at solid to water ratio of 1:10 (dw/v) using a horizontal shaker at 200 rpm for 3 h followed by centrifugation at 3000 RPM for 20 min and filtration with 0.45 µm syringe filters. pH was measured electrometrically in slurry after the shaking step, with smartCHEM-LAB Laboratory Analyser.

The MC was determined by oven drying at 105±2 °C until weight became constant as reported above. OM was determined by loss on ignition (LOI) of dried and ground mass in the muffle furnace at 550±15 °C for 4 h.

The C content was determined using an ELTRA CHS 580 analyzer in a furnace preheated to 1350 °C, where the sample was burnt and flue gas was directed to the measuring cuvettes. The N content was determined using an ELTRA N 580 analyzer in an oven preheated to 950 °C. The OM and N losses were calculated from the initial (X_1) and final (X_2) ash contents according to the equations (1-2) by Paredes et al. (1996).

$$OM \text{ loss } (\%) = 100 - 100 \frac{X_1(100 - X_2)}{X_2(100 - X_1)} \quad (1)$$

$$N \text{ loss } (\%) = 100 - 100 \frac{X_1 N_2}{X_2 N_1} \quad (2)$$

Where N_1 and N_2 are the initial and final N concentrations, respectively. HMs contents in dry mass (d.m.) were measured using the ICPOES model 5100 SVDV inductively coupled plasma mass spectrometry system.

Before the measurement compost was mineralized with aqua regia. Wet bulk density was estimated based on mass in the known volume.

Statistical analysis

Statistical analysis of the obtained results was made using the IBM SPSS-v.21 software package. A two-factor analysis of variance was performed in order to check the significance of the selected physicochemical properties and HMs contents in samples collected at different times from piles with different biochar contents.

Results and discussion

Characteristics of OFMSW used in the compost production

Table 4 gives the analysis results for the physicochemical characteristics and concentration of HMs in raw materials (OFMSW) used in this research to produce the compost. According to this table, pH of the OFMSW is 7.88 ± 0.1 , which sits in the alkali range. In a similar study conducted by Awasthi et al. (2015) in the compost facility of Jabalput, India, the pH of the compostable materials was reported in a similar range and equal to 7.27. However, Romero de Leon et al. (2021) measured the pH of the compostable materials used in a compost plant located in Mexico City in the acidic range equal to 5.5. This difference between the reported values is generally attributed to the different compositions of compostable materials, which mainly results from the manner of implementing waste separation from source program and the different efficiencies of processing and removal systems in compost facilities (Fernández-Delgado et al., 2020; Frimpong et al., 2021; Godlewska et al., 2017).

According to Table 4, MC in the OFMSW is $59.2 \pm 2.3\%$. This value corresponds to the moisture range measured for the compostable materials in the compost production line of Ireland (58.7%), as reported by Graca et al. (2021). On the other hand, the MC of the OFMSW of Gipuzkoa, Spain, was reported as 41% (Kepa Izaguirre et al., 2020). The main reason for the different reported MCs is the differences of organic waste materials and the diverse climatic and weather conditions of the mentioned regions (D'Hose et al., 2020; Cui et al., 2022).

Based on the contents of HMs measured in the OFMSW (Table 4), it is observed that the concentrations of some HMs such as Zn, Cu, and Pb were relatively high and determined as 1497.96 ± 35.4 mg/kg, 299.94 ± 18 mg/kg, and 101.78 ± 7.3 mg/kg, respectively. In a study by Asquer et al. (2017), the concentrations of Cu and Zn were respectively 17.76 mg/kg and 56.64 mg/kg, which are much lower than the total average of these metals in the OFMSW in this study. The main reason for the different reported values is differences between the collection and transportation methods of MSW (Guo et al., 2021). The lack of waste separation from source (Jiang et al., 2021) and mixed waste transportation to the compost facility production lines in CPBM lead to the entrance of materials such as dyes, batteries, electronic devices, plastics, newspapers, domestic cleaning agents such as soaps and detergents, cosmetics, packaging, and medications to the compostable materials and thus increases the concentration of HMs in them. A similar condition is also observed in other cities of Iran. For instance, Rupani et al. (2019) measured the concentration of Cr in OFMSW collected from compost facility of Aradkooh, Tehran, equal to 81.7 mg/kg, which is around 2.5 times greater the measured concentration of this HM in the OFMSW of CPBM (35.5 ± 1.7 mg/kg).

Table 4. Physicochemical properties of raw materials used for the compost production

Parameters	Unit	OFMSW
MC	%	59.20 ± 2.3
OM	%	50.48 ± 3.1
Bulk density	kg/m ³	519.14 ± 32
C	%	31.02 ± 6.5
N	%	2.18 ± 0.2
C/N	-	14.23 ± 1.8
pH	-	7.88 ± 0.1
Cd	mg/kg	1.02 ± 0.3
Cr	mg/kg	35.5 ± 1.7
Cu	mg/kg	299.94 ± 18
Ni	mg/kg	46.15 ± 3
Pb	mg/kg	101.78 ± 7.3
Zn	mg/kg	1497.96 ± 35.4

Mean \pm standard error of mean (n=3)

Effect of biochar on HMs concentration in compost production process from OFMSW

Table 5 demonstrates the concentrations of HMs in the produced composts at the end of week nine from the start of the process. Table 5 shows that the BC0 (control pile) has the highest and the BC5 has the lowest concentrations of HMs among the piles of compostable materials. Pb, Zn, Ni, Cd, and Cu demonstrated a significant concentration reduction in all the compost piles containing biochar (BCs 1, 3, and 5) compared with the control pile (without biochar addition). Regarding Cr, no significant difference was observed between the piles containing biochar and the control pile. In this regard, Cr concentration was measured as 37.14 ± 0.7 mg/kg in the final product of the control pile while it was measured as 35.23 ± 0.5 , 34.41 ± 0.6 , and 34.27 ± 0.6 mg/kg in the final product of piles containing 1%, 3% and 5% biochar, respectively. Reductions in Cr concentration in the piles with 1%, 3%, and 5% biochar was 5.15%, 7.36%, and 7.73%, respectively, which is not significant.

Among different HMs, Cd concentration in the pile containing 5% biochar declined by 71.63% compared with that in the control pile, indicating the highest reduction trend. In addition, in the piles containing 1% and 3% biochar, the concentration of this metal declined by 55.82% and 70.24%, respectively, relative to that in the control pile. As can be seen in Table 5, the variations of Cd concentration in the piles containing 3% and 5% biochar are similar. This is also the case for Cu and Zn, for which the rise in the biochar content of the compostable materials from 3% to 5% failed to further decrease the concentrations of these metals at the end of the process. Regarding Pb, the addition of 1%, 3%, and 5% biochar to the compost pile lowered the concentration of this metal by 12.6, 13.6, and 25.14 respectively, compared with the control pile. This indicates that increasing the content of biochar in the compostable materials from 1% to 3% led to no significant change in Pb concentration. The variation of Ni in the piles containing biochar followed a completely different trend compared with other metals. The concentration of this metal in the control pile was 46.19 ± 2.3 mg/kg, which declined to 38.49 ± 2 , 38.49 ± 1.7 , and 37.47 ± 2.3 mg/kg in the piles containing 1%, 3%, and 5% biochar, respectively. Ni concentration declined by 16.68% in the piles with 1% and 3% biochar and by 16.72% in the pile with 5% biochar. Indeed, increasing the weight percentage of biochar mixed with the compostable materials had no positive effect on the reduction of Ni concentration.

Table 5. Changes in the concentrations of HMs with the addition of biochar in the composting process of OFMSW

Pile	Heavy metals (mg/kg d.m.)					
	Pb	Zn	Ni	Cd	Cr	Cu
BC0	104.31±7	1536.07±41	46.19±2.3	2.15±0.22	37.14±0.7	339.00±18
BC1	91.17±5.8	1077.86±38	38.49±2	0.95±0.11	25.23±0.5	259.72±25
BC3	90.13±6.4	1023.54±39	38.49±1.7	0.64±0.19	34.41±0.6	240.36±19
BC5	78.09±7.3	994.16±35	38.47±2.3	0.61±0.24	34.27±0.6	218.75±15

Mean ± standard error of mean (n = 3)

Based on the biomass nature, the compost can contain different concentrations of HMs (Liang et al., 2020). The composting process affects the bioavailability of HMs. In biologic process, a decrease in the concentration of OM in biomass leads to a rise in the concentration of HMs (Gondek et al., 2018). Bonds are formed between HMs and OM through the formation of complexes, which lower the solubility of metals and thus reduce their bioavailability (Vandecasteele et al., 2013). The results of various research works have shown that biochar demonstrates a significant affinity to HMs (Inyang et al., 2016). Biochar is used for the treatment of aquatic environments and immobilization of HMs in soil and sediments (Ahmad et al., 2014). Therefore, it can be inferred that the addition of biochar to compostable materials during the process also leads to the stabilization of HMs in the final product (Awasthi et al.

2020). HMs do not disappear during the composting process, but their mobility significantly decreases after the addition to soil (Deng et al., 2021). Therefore, the risk of their transfer into the environment (through leaching from soil and accumulation in organisms) declines, particularly when OFMSW or other waste containing HMs are used to produce compost (Ignatowicz, 2017). The reduced mobility of heavy metals in the compostable materials containing biochar is achieved through the following mechanisms (Hale et al., 2021):

- Physical absorption by biochar,
- Complexation at the biochar surface,
- Ion exchange with ions on the biochar surface, and
- Electrostatic reaction with electrical charge at the biochar surface.

In composting processes with different initial materials, reduced concentrations of HMs have been observed in the final product of compostable materials combined with biochar compared with compostable materials going through the process without the addition of biochar. For instance, Duan et al. (2021) conducted the research to investigate the variation of HM concentrations during the composting process from sheep manure. It was reported that the concentration of Zn in the control treatment (without biochar combination) was 211.9 mg/kg, while the concentration of this metal in compost treatments containing 2.5%, 5%, 7.5%, 10%, and 12.5% biochar reached 186.47, 175.71, 171.04, 164.87, and 161.458 mg/kg, respectively. The findings of another study showed that combining 1.5% and 5% biochar with OFMSW as the compostable materials lowered the concentrations of Cd by 49.5% and 55.72%, respectively (Malinowski et al., 2019). Awasthi et al. (2016) assessed the effect of biochar on the bioavailability of HMs during the composting process from sewage sludge and reported that in the control pile, the concentration of Ni and Zn increased gradually from 28.34 to 46.16 mg/kg and from 270.2 to 485.1 kg/mg, respectively. However, the concentration of these metals in the compost pile containing 12% biochar showed a decreasing trend. In addition, Wang et al. (2022) reported that in the composting process of pig manure, the addition of biochar lowered the concentration of Zn and Cu by 9.9% and 24.8%, respectively.

Comparative analysis of quality standards for compost

To further investigate, HM concentrations in the compost are provided in Table 6 in accordance with the international standards of World Health Organization (WHO) and Unites States Environmental Protection Agency (EPA). Comparing the HM concentration of the final produced compost in the piles tested (Table 5) with international standards given in Table 6 indicates that the concentrations of Zn and Pb in the compost produced from the control pile (without biochar addition) were above the standard range and failed to meet them. However, the addition of biochar even at low doses (1%) decreased the concentration of these metals. Although the concentrations of some HMs (Ni, Cd, and Cr) in the control pile are below limits specified by international standards, it is desirable to have even lower concentrations of these metals.

Table 6. International standards of the compost quality in terms of HMs concentration (mg/kg d.m.) (Hao et al., 2019; Zhang et al., 2021)

HMs	WHO	EPA
Cd	15.40	10
Cr	-	100
Cu	90-260	200
Ni	-	200
Pb	200-400	100
Zn	800-1200	2000

Conclusions

The continuous increase in the volume of MSW necessitates the use of methods such as composting to reduce the volume of waste. Since the organic fertilizer produced in this process has agricultural and gardening applications and is somewhat related to the health of population, investigating the qualitative characteristics of compost and efforts to improve its quality must always be considered. The results of the present study indicate that using biochar as a beneficial additive material can improve the quality of the final product of the composting process in real conditions through lowering the concentration of HMs. According to the results, adding 5% of biochar to the compostable materials decreased the concentrations of Cd, Cu, Zn, Pb, Ni, and Cr by 71.63%, 35.48%, 35.38%, 25.14%, 16.72%, and 7.73%, respectively. The results of this research can therefore be used to better use biochar in the production of compost from organic waste.

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