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Research Article

Influence of zeolite on heavy metal immobilization in municipal solid waste compost contaminated soil

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Abstract
The application of Municipal solid waste as compost (MSWC) in agricultural fields has become one of the most common practices. Besides its benefits, it poses some harmful effects on soil, as it increases the heavy metal content in MSWC of the soil. It is necessary to find a way to reduce the bioavailability of heavy metals in MSWC before its application into the soil. This study aimed at exploring the efficiency of zeolite as an immobilizer to dwindle heavy metal bioavailability. An incubation experiment was conducted wherein the soil samples were artificially spiked with different rates of MSWC (0, 5, and 10 t ha^{-1}). The zeolite was added to the spiked soil at 5 different levels, namely 0, 5, 10, 15, and 20 %, and their effect on bioavailable heavy metal status was observed during different incubation intervals (0, 15. 30, 60, 90, and 120 days). Results unveiled that applying 10% zeolite significantly ($P<0.05$) reduced the bioavailability of lead (Pb) and nickel (Ni) to Below the detectable limit (Bdl) in all soil samples. Furthermore, the organic carbon status of soil was also enriched by MSWC and 10% zeolite application. The soil pH slightly increased (7.39) with applying 10% zeolite resulting in the immobilization of heavy metals. Hence, 10% zeolite application was one of the most effective immobilizers in eliminating the bioavailability of heavy metals. Therefore, it can be concluded that mixing zeolite with MSWC before applying it to crop fields can reduce the heavy metal overload in soil. Hence, this study highlights the potential of zeolite as an effective choice in dwindling the soil's bioavailability of heavy metal content.

Keywords: Heavy metals, Immobiliser, Municipal Solid Waste Compost (MSWC), Zeolite

INTRODUCTION

Commercial and residential wastes formed in a municipal or notified area in solid or semi-solid form, excluding industrial hazardous wastes but including treated bio-medical wastes, are classified as municipal solid waste. Municipal solid waste generation in various cities of India ranges from 0.3 to 0.6 kg/capita/day. According to estimates, currently India produces 62 million tonnes of Municipal Solid Waste, out of this only 22-28% is treated and processed (MOHUA, 2021). Composting urban waste is a practical solution because it not only solves sanitation issues but also supplies valuable agricultural input in the form of soil conditioners like nitrogen, phosphorus, and potassium (NPK) fertilizers. MSWC improves soil fertility by providing soil organic matter and plant nutrients. It also improves water holding capacity, infiltration, soil aeration, and soil microbial reaction, as well as reducing erosion and improving soil structure (Bouzaiane *et al*., 2014; Weber *et al*., 2014; Lim *et al*., 2015; Almendro- Candel *et al*., 2019). This has a beneficial effect on plant growth (Rajaie and Tavakoly, 2016). However, in some circumstances, only applying MSWC to supply nutrients for crops may not be enough; thus, combining MSWC with inorganic fertilizers can boost soil nutrients and crop yield (Nigussie *et al*., 2015, Machado *and* Hettiarachchi 2020).

MSWC is also utilized to preserve and improve soil structure since its organic matter content can help to reverse the natural deterioration in intensively cultivated soils. It might potentially take the place of typical farm manure, which is sometimes scarce in intensive agriculture areas. Aside from potentially beneficial nutrients, some waste materials may also contain non-essential elements, persistent organic compounds, and bacteria that are potentially toxic to plants (Chukwuji *et al.,* 2005, Kabasiita *et al*., 2022). All varieties of municipal solid waste (MSW) compost include higher levels of heavy metals than the background amounts in soil, and their presence will increase in supplemented soil. The metals zinc (Zn) and lead (Pb) are numerically the most abundant in MSW compost. Heavy metals are found in compost due to a variety of sources, including domestic municipal solid waste products (MSW). Household dust, batteries, throwaway household materials (e.g., bottle caps), plastics, paints and inks, body care products and medicines, and household pesticides (National Household Hazardous Waste Forum, 2000; Bardos, 2004, Dada *et al*., 2022). The presence of harmful heavy metals in municipal solid waste composts (MSWC) raises severe concerns regarding the negative environmental impact. Long-term accumulation of heavy metals in the soil environment is a problem since they can have serious repercussions for human food quality, plant toxicity, and soil microbial processes, and they have very long residence durations in soil once applied. The bioavailable status of heavy metals in MSWC amended soils is higher than that of manure amended soils. MSWC application led to greater transfers of Ni, Pb, and Cd metals in plants (Topcuoglu, 2016).Therefore, it is essential to immobilize the heavy metals present in MSWC when applied to the soil at least during the crop growing period. Physical, chemical, and biological approaches can be used to immobilize heavy metals and minimize their availability in the soil. Chemical immobilization, for example, will reduce the concentration of dissolved pollutants through sorption, lowering the metal availability to plants. Many researchers have suggested that using MSWC in combination with specific modifications could render heavy metals in MSWC immobile. Organic and inorganic amendments are frequently utilized in immobilization technology to speed up the attenuation of metal mobility and toxicity in soils (Selvi *et al*., 2019). The principal function of immobilizing additives is to change the original soil metals to more geochemically stable phases through sorption, precipitation, and complexation processes. Clay, cement, zeolites, minerals, phosphates, organic composts, and microorganisms are the most commonly used amendments. Calcite, goethite, montmorillonite, bentonite, zeolite, and kaolinite are some of the more often utilized clay minerals for heavy metal immobilization (Ou *et al*., 2018). The contribution of each of these clay minerals to heavy metal ion immobilization in the soil varies depending on the heavy metal ion, chemical and physical soil qualities, as well as the clay minerals' features (Radziemska *et al*., 2020).

Zeolites are hydrated aluminosilicates of alkali and alkaline earth cations that occur naturally as crystalline, hydrated aluminosilicates. They occur naturally when high-pH, high-salt-content water mixes with volcanic ash, resulting in fast crystal formation. They have a silicate framework made up of interconnecting SiO_4 and AlO_4 tetrahedrons. The negatively charged aluminosilicate structure attracts positively charged cations, giving the zeolite a high cation exchange capacity (CEC). Large cation groups (sodium, potassium, barium, and calcium) and even relatively enormous molecules and cation groups (water, ammonia, carbonate ions, and nitrate ions) can penetrate the zeolites due to the large free space. In some zeolites, these spaces are joined and form vast channels of varying diameters, depending on the mineral. The resident ions and molecules can easily move in and out of the structure because of these channels. The capacity of zeolites to lose and absorb water without causing damage to their crystal structures is one of its most essential properties. Zeolites are excellent chemicals for soil remediation because of their features. Heavy metals like Pb(II), Cu (II), Cd(II), and Cr(III) were thought to counteract the tetrahedral Al's negative charge in the unreacted zeolite. (El-Eswed *et al*., 2015, Belviso, 2020). Many studies have shown that zeolite is effective in heavy metal immobilisation of soil. This study was undertaken with the objective to elucidate the potential of zeolite as an immobilizer of contaminants in MSWC under a laboratory incubation experiment.

MATERIALS AND METHODS

Collection and characterisation of MSWC

MSWC was collected from United Phosphorus Limited (UPL), Vellalore, Coimbatore, Tamil Nadu and subjected to laboratory characterization. The physico-chemical properties such as pH (Jackson, 1973), organic carbon (Walkley and Black, 1934), and Heavy metals

Table 1. Characterization of MSWC and soil

Properties	MSWC	Initial soil	Spiked Soil
pH	7.43	7.15	7.12
Organic carbon (%)	11.0	0.34	0.36
Available Nitrogen (kg ha^{-1})	-	152	189
Available Phosphorus (kg ha^{-1})	-	11	19
Available Potassium (kg ha^{-1})	-	630	685
Lead (mg kg^{-1})	65.0	Bdl	63
Nickel (mg kg^{-1})	25.0	Bdl	20

(Jackson, 1973 were analyzed and depicted in Table.1.

Soil collection and characterisation

The soil samples were collected from Madampatti village of Thondamuthur block in the Coimbatore district. The soil samples were air-dried, ground, and passed through a 2mm sieve before further analysis and their initial characteristics were analyzed before and after spiking with MSWC and are given in Table 1.

Incubation experiment

A soil incubation experiment was conducted to study the effect of zeolite on heavy metal immobilization in MSWC. Two hundred grams of collected soil were taken in a plastic container and different rates of MSWC, *i. e*., 0, 5, and 10 t ha^{-1} were spiked to soil and incubated for 3 days for stabilization. The field capacity was maintained in the incubated soils. Based on weight loss, distilled water was added to the container to maintain the moisture content throughout the incubation experiment. After 3 days of equilibration, Zeolite was added at five different rates *i. e*., 0, 5, 10, 15, and 20%. Three replicates of each treatment were prepared, randomly placed, and incubated in the laboratory at 25± 2°C for 60 days. The samples were collected in triplicate for laboratory analysis at 0, 30, 60, 90, and 120 Days After Incubation (DAI). The collected samples were then analyzed for pH (Jackson, 1973), Organic carbon (Walkley and Black, 1934), and the water-soluble fraction of lead (Pb) and nickel (Ni) (US Environmental Protection Agency, 1979).

Statistical analysis

Each treatment in this study was applied in a randomized design. Statistical Package of Social Sciences (SPSS) was employed for statistical analysis. The data recorded were analyzed statistically by analysis of variance techniques appropriate for Factorial Completely Randomised Design (FCRD) as suggested by Gomez and Gomez (1984). Means were compared by the least significant difference test (CD < 5%).

RESULTS AND DISCUSSION

Effects of zeolite on soil pH

Different zeolite and MSWC application rates showed very slight variations in the soil pH, as depicted in Fig 1. The soil pH showed an increasing trend along the

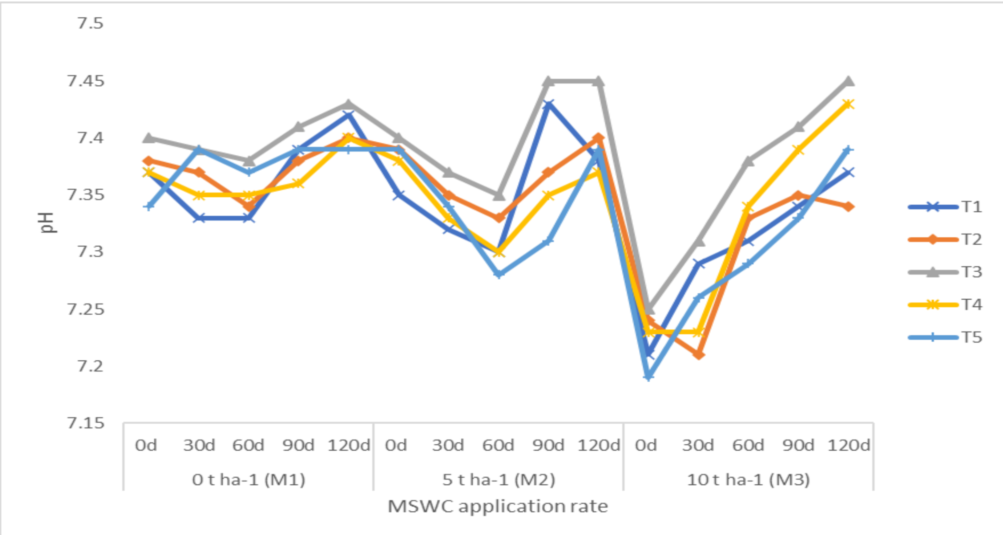

Fig. 1. *Effect of zeolite and MSWC application on soil pH. M_1 – MSWC @ 0 t ha^{-1}, M_2 – MSWC @ 5 t ha^{-1}, M_3 – MSWC @ 10 t ha^{-1}. T_1 – Control , T_2 – 5 % Zeolite, T_3 – 10 % Zeolite, T_4 – 15 % Zeolite, T_5 – 20 % Zeolite*

subsequent days of incubation under the influence of zeolite application. The MSWC application decreased the soil pH in subsequent days of incubation. The application of 10 t ha^{-1} showed a lesser soil pH (7.31) than that of 5 t ha^{-1} (7.36). The zeolite application @ 10% rate showed a notable increase in soil pH (7.39) at all stages under all rates of MSWC application. The effect of 10% zeolite application on soil pH was found to be on par with the 15% and 20% application rates. Even though the increase in MSWC application decreased the soil pH, the zeolite application at 10% reduced the effect of MSWC on soil pH. Li *et al*. (2009) reported that an increase in the dose of zeolite application significantly increased the soil pH, which promotes the chemical immobilization of heavy metals by the metal sorption process. The application of zeolite increases the solution pH and therefore increases the efficiency of heavy metal removal. This is due to the competition between the hydrogen ions and heavy metal cations for the same exchange sites and electrostatic repulsion between the heavy metal cations in the solution (Elboughdiri and Garcia, 2020). The ion exchange process increases with an increase in pH up to a maximum value (Argun, 2008) concluded that pH values between 5 and 7 are the best heavy metal removal efficiency value. Applying zeolite increases the alkalinity by increasing soil pH and Na+ release, which increases soil EC and increases heavy metal adsorption capacity. This also increases the negative charge and adsorbs heavy metals by complex levels (Shi *et al*., 2009).

Effects of zeolite on soil organic carbon

The incorporation of zeolite has remarkably improved the soil's organic carbon status. The soil organic content showed an increasing trend along the subsequent days of incubation due to the addition of MSWC. Different zeolite and MSWC application rates showed variation in the soil organic content, as depicted in Fig 2. The increase in the rate of MSWC application has significantly raised the soil organic carbon in subsequent stages of the incubation experiment. Comparing the different MSWC application rates, 10 t ha^{-1} of MSWC showed a higher organic carbon content (0.55%) than that of 5 t ha^{-1} (0.49%). 10% zeolite application showed a higher organic carbon content (0.53 %). On comparing the treatments, the effect of 10% zeolite application on soil organic content was found to be on par with 15% and 20% application rates at all stages under all rates of MSWC application. Similar positive impact of zeolite on soil nutrient status and maize yield were obtained by Aslam *et al*. (2021). The application of 10% zeolite increased the soil organic carbon, organic matter, water retention capacity, and decreased bulk density. It also reduces the oxidation of organic matter by forming complexes with them. The application of zeolite increases the soil organic carbon status by improving soil water retention and aggregate stability (Cairo *et al*., 2017). Truc and Yoshida, (2011) reported the increased accumulation of carbon in zeolite applied plots due to the complexation between the oxy of zeolite and organic acids in soil, resulting in the strong organo- metallic complex. This reduces the speed of organic matter decomposition and increases the soil's organic carbon status.

Effects of zeolite on soil heavy metal status

The changes in the water-soluble fraction of heavy metals - pb and nickel due to zeolite application were examined along the subsequent days of incubation. The concentration of lead and nickel significantly differed

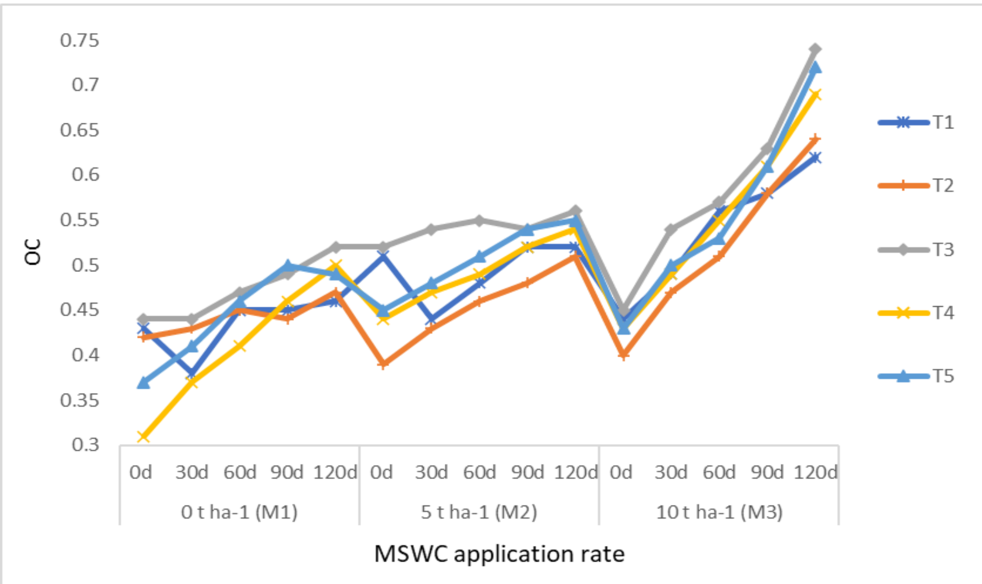

Fig. 2. *Effect of zeolite and MSWC application on soil OC.M_1 – MSWC @ 0 t ha^{-1}, M_2 – MSWC @ 5 t ha^{-1}, M_3 – MSWC @ 10 t ha^{-1}.T_1 – Control , T_2 – 5 % Zeolite, T_3 – 10 % Zeolite, T_4 – 15 % Zeolite, T_5 – 20 % Zeolite*

Table 2. Effect of zeolite application on Pb concentration in soil at different time intervals

Treatments	0 DAI			30 DAI			60 DAI			90 DAI			120 DAI		
	M_1	M_2	M_3	M_1	M_2	M_3	M_1	M_2	M_3	M_1	M_2	M_3	M_1	M_2	M_3
T_1	Bdl	51	62	Bdl	49	60	Bdl	47	59	Bdl	45	57	Bdl	45	56
T_2	Bdl	50	61	Bdl	42	42	Bdl	31	21	Bdl	18	13	Bdl	Bdl	Bdl
T_3	Bdl	49	63	Bdl	22	38	Bdl	13	19	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl
T_4	Bdl	47	60	Bdl	31	31	Bdl	10	11	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl
T_5	Bdl	48	60	Bdl	19	29	Bdl	9	9	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl

M_1 – MSWC @ 0 t ha^{-1}, M_2 – MSWC @ 5 t ha^{-1}, M_3 – MSWC @ 10 t ha^{-1};T_1 – Control, T_2 – 5 % Zeolite, T_3 – 10 % Zeolite, T_4 – 15 % Zeolite, T_5 – 20 % Zeolite.

Table 3. Effect of zeolite application on Ni concentration in soil at different time intervals

Treatments	0 DAI			30 DAI			60 DAI			90 DAI			120 DAI		
	M_1	M_2	M_3	M_1	M_2	M_3	M_1	M_2	M_3	M_1	M_2	M_3	M_1	M_2	M_3
T_1	Bdl	12	18	Bdl	10	15	Bdl	9	12	Bdl	8	8	Bdl	5	4
T_2	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl
T_3	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl
T_4	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl
T_5	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl	Bdl

M_1 – MSWC @ 0 t ha^{-1}, M_2 – MSWC @ 5 t ha^{-1}, M_3 – MSWC @ 10 t ha^{-1}; T_1 – Control, T_2 – 5 % Zeolite, T_3 – 10 % Zeolite, T_4 – 15 % Zeolite, T_5 – 20 % Zeolite.

under the MSWC and zeolite application in their subsequent stages of incubation, as depicted in Table 2 and 3. The increase in MSWC application rate increased the lead and nickel status. The application of 10 t ha^{-1} of MSWC showed a higher pb and nickel content than that of 5 t ha^{-1}. The reduction in bio-available heavy metal content was more profound under the application of 10% zeolite. But its effect was statistically comparable with 15% and 20% zeolite application under all rates of MSWC application. From the results, it can be justified that increasing the zeolite application have not shown any significant variations in immobilising heavy metals. Therefore, the application of Zeolite @10% can be considered as effective immobilizer of heavy metals even under the higher MSWC application rate of 10 t ha^{-1}. The results indicated that the immobilising potential of 10% zeolite in MSWC is as effective as its potential in soil. Contin *et al*. (2019) reported that 10% zeolite efficiently reduced heavy metals such as Cu, Cd, Ni, and Zn in soil due to the simultaneous occurrence of organic complexation, pH rise, sorption by surface complexation, and cation-exchange retention. Wyszkowski (2019) reported that 2.5% zeolite effectively reduces the heavy metal uptake in sewage sludge-contaminated soil. Zeolite can act as an efficient immobilizer of multi-minerals because of its higher cation exchange capacity than other soil minerals. The number of toxic elements desorbed from zeolite was lower compared to other minerals, therefore, it can hold more than 70% of heavy metals in non-exchangeable form (Pannucio *et al*., 2009). Mahabdi *et al*. (2007) reported that 9% zeolite effectively reduced Cd solubility and 10% reduced pb solubility. Zeolite follows an ion-exchange mechanism by which some ions are forced to move through the mass of zeolite and its canals based on its hydration radius (Azogh *et al*., 2021).

Conclusion

The present study proved that zeolite can be effectively used as an amendment for immobilizing the heavy metals of MSWC. The soil incubated with MSWC at 0, 5 and 10 t ha^{-1} showed that MSWC application could increase the heavy metal status of soil, particularly the bio-available fraction of Pb and Ni. This heavy metal hike in the soil is not acceptable, as the long-term application of MSWC can pollute the soil and human health. Hence there is a need to design a methodology for using MSWC to soil with affecting soil health. Therefore, using an immobilizing agent along with MSWC can reduce its heavy metal overload in soil. Hence the results of the study suggest that zeolite @10% application rate can reduce the bioavailability of heavy metals in soil. This captures the heavy metals in MSWC, reducing their impact on soil. The effects of zeolite @ 15 and 20% were statistically comparable with zeolite @ 10 %. So, it can be concluded that 10% zeolite is an effective immobilizer for both 5 and 10 t ha^{-1} MSWC application rates. Instead of applying MSWC alone in soil, it is advisable to use it along with zeolite to protect the soil health and quality for sustainable agriculture.

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Conflict of interest

The authors declare that they have no conflict of interest.

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