Pollution, Ecological, and Health Risk Assessments of Heavy Metal Remediated Soil by Compost Fortified with Natural Coagulants

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Abstract

The present study assessed the pollution, ecological, and health risks of heavy metals (HMs) in remediated soils by compost fortified with natural coagulants (Moringa, Neem, and Pawpaw).The polluted soils were mixed with the stabilized composts at varying ratios of 1:200, 1:100, 1:66, and 1:50. Heavy metals(Cu, Zn, Ni, Mn, Cd, and Pb) were determined in compost and soil samples using Atomic Absorption Spectrophotometry. Data collected were analyzed for descriptive and inferential statistics. Removal efficiencies (REs) of HMs by the compost types were also determined. Results showed Mn as the highest observed HM in composts with values ranging from 179.6 ± 1.25 mg kg^{-1} in Neem to 206.3 ± 5.01 mg kg^{-1} in Moringa. The levels of HMs (except Zn) observed in composts were generally less than the target values expected in unpolluted soil. Application of the three composts to contaminated soils reduced the HMs by 5–37, 17–47, 25–43, 43–57,59–71, and 69–75% for Cd, Cu, Ni, Pb, Mn, and Zn, respectively. Removal of HMs in contaminated soils followed the distribution pattern of Zn > Mn > Pb > Cu > Ni > Cd. All the compost types were effective in cleaning up Mn and Zn in contaminated soils. However, Moringa appeared to indicate the highest removal efficiency followed by Neem and Pawpaw, in that order.

Introduction

Soil pollution by heavy metals (HMs) is a major global issue of concern in terms of environmental, ecological, and health effects. Even at low concentrations, HMs are dangerous to humans, animals, plants, and the environment (Taiwo et al. 2021).They are non-biodegradable but undergo eco-biological cycling (Iwegbue et al. 2006; Taiwo et al. 2021).Dumpsite contains high levels of HMs from diverse sources such as industry, municipal and electronic wastes. These metals are eventually released into the soils, with possible infiltration into the groundwater resource through leachates (Liang et al. 2014; Taiwo et al. 2021). Human exposure to HMs could emanate from ingestion of contaminated groundwater, and consumption of plants that had bio-accumulated HMs from the (dumpsite) soil (Duruibe et al. 2007). Other exposure routes to HMs from the soil are inhalation and dermal contact (Orosun et al. 2020).Removal of HMs in contaminated soil is, therefore, necessary for the production of healthy foods and the protection of public health.

HMs can be removed from environmental media (soil, water, sediments) by biological (bioremediation), chemical (precipitation, coagulation, complex formation), and physical (filtration) techniques. Among these, bioremediation is the most economical (low cost) and environment-friendly(natural process) method (Chibuike and Obiora 2014; Taiwo et al. 2020a).Bioremediation involves the use of the living systems (microorganisms and plants) to remove pollutants (organic and/or inorganic) in the environmental media (Taiwo 2011; Taiwo et al. 2016). Although bioremediation processes may not degrade HMs but could transform them from one oxidation state to another (Chibuike and Obiora 2014). During this process, HMs may be transformed to become more volatile, water-soluble, and less toxic. This further allows precipitation, easy removal, and less bioavailability of the pollutant (Garbisu and Alkorta, 2003).During composting, the reduced bioavailability of HMs may result from the formation of stable metal-humus complexes (Huang et al. 2016).

The rate of land encroachment (for residential and farming purposes) around the dumpsite in many cities in developing countries, is very alarming (Sadek and El-Fadel 2000; Badejo et al. 2013; Salukele 2013). It is therefore important to reclaim these inorganic pollutants through proper cleanup in a viable and sustainable manner. Studies on pollution, ecological and toxicological assessments of HMs in contaminated and remediated soils are scarcely and rarely reported. The available research works on HMs were basically conducted on levels and distributions in polluted soils before remediation (e.g., Taiwo et al. 2016; Nejad et al. 2018; Selvi et al. 2019). This is the first study that adopted three different compost types made from natural coagulants (*Moringa oleifera*, *Azadirachta indica*, and *Carica papaya*)to remediate HMs contaminated dumpsite soils. Furthermore, this study also assessed the pollution, ecological, and health risks of HMs in contaminated and remediated dumpsite soils.

Materials And Methods

Collection of contaminated soil

Contaminated soil samples for remediation were collected (at depth 0–30 cm) from four different locations in Saje dumpsite, Abeokuta, Ogun state. This is an abandoned quarry mining site converted to a dumpsite in 1995 (Popoola and Adenuga 2019). On daily basis, about 150 tonnes of municipal wastes are dumped in the Saje dumpsite with a capacity of 680 m^2 and a thickness that varied between 2 and 3 m (Popoola and Adenuga 2019). The polluted dumpsite soils were bulked together to form a composite sample. The composite soil samples were air-dried and sieved through a 2 mm mesh into different plastic containers for the bioremediation experiment (Taiwo et al. 2016).

Experimental design

The experiment took place at a screen house in the Federal University of Agriculture, Abeokuta. The experimental design followed a randomized complete block design of 24 treatments (3 x 4 x 2!). Each treatment was replicated in three samples. The Moringa, Neem and Pawpaw composts and the contaminated soil were mixed together in four separate ratios of 1:200 (5 g composts + 1 kg soil), 1:100 (10 g compost + 1 kg soil), 1:66 (15 g compost + 1 kg soil), and 1:50 (20 g compost + 1 kg soil). Castor oil plant (*Ricinus communis*) seeds were introduced for a four-week integrated (with phytoremediation) experiment (Taiwo et al. 2016). A total of 72 pots were set up for the experiment. Both the 'compost only' and 'compost + castor oil plant' treatments were wetted with distilled water three times per week. Three samples of contaminated soils were analyzed before the introduction of composts and plants.

Compost formulation and composting

The composts were obtained after the degradation of organic wastes (Haouvang et al., 2017) from the poultry manure, leaves of *Moringa oleifera* (Moringa), *Azadirachta indica* (Neem), *Carica papaya* (Pawpaw), and sawdust. The compost materials were mixed from organic wastes such as poultry manure (35 kg), leaves (10 kg), and sawdust (2 kg) following the procedures described in Joyce (2010). The compost underwent a controlled aerobic condition and close monitoring through periodic wetting and turning, for two months. Heavy metal levels in composts were monitored for eight weeks.

Sample analysis

A total of 135 [3 untreated contaminated soil, 72 treated soil, 24 compost (3 per week for 8 weeks), and 36 plant] samples were analyzed for HMs (Cu, Zn, Ni, Mn, Cd and, Pb) using atomic absorption spectrophotometric (AAS) method (Taiwo et al. 2016). Prior to the determination of HMs, the air-dried soil/compost/plant samples were pulverized to < 100 µm. Samples were digested with mixed concentrated acids [perchloric (70% Purity), sulphuric (98% Purity,) and nitric (70% Purity)] using the procedures described in Taiwo et al. (2021).This entails weighing 1.0 g ground samples into a conical flask, followed by an introduction of 1, 2 and 7 mL concentrated perchloric, sulfuric, and nitric acids, respectively. The mixture was heated on a hot plate under gentle heat in a fume cupboarduntil a clear solution was obtained. On cooling, the digested samples were diluted with 50 mL distilled water, filtered into a prewashed 100mL plastic bottle, and made up to mark. The digested samples were transported to the Department of Agronomy Laboratory, University of Ibadan; for determination of Zn, Ni, Mn, Cd, and Pb using AAS (Buck Scientific 210 VGP, USA). Blank samples were run to cancel the matrix background effect of the analytical reagents. All reagents used for extraction were of analytical grade (Sigma-Aldrich Chemie, GmbH). Glassware and plastic bottles were prewashed with 2% $HNO_3$and rinsed thoroughly with distilled water before oven drying at 37^{O}C for 2 hours (Taiwo et al. 2014).

Data analysis

Data collected were evaluated for descriptive (mean, standard deviation, minimum and maximum) and inferential (Duncan Multiple Range Test) statistics using the Statistical Package for Social Sciences (SPSS) for Windows (SPSS 22.0).

Removal efficiency (RE)

The portion of HMs remediated by the three compost types and plant + composts in contaminated dumpsite soils was calculated as:

$$\% \text{ RE} = \frac{Cs - Cr}{Cs} \times 100 \quad (1)$$

Where C_s is the concentration of HM in contaminated soil before remediation; C_r is the concentration of HM after remediation.

Pollution and ecological risk assessment

The pollution and ecological risk assessments of HMs in compost/soil samples were estimated for pollution index (PI), Nemerov integrated pollution index (NIPI), and ecological risk index (ERI),using the formulas presented in equations 2–4 (Taiwo et al. 2020a, b).

$$PI = \frac{Cs}{Cf} \quad (2)$$

Where C_s is the concentration of HM in the compost/soil samples, C_f is the reference value of the earth's crust (Wedepohl 1995). PI < 1 indicates low pollution, while PI > 1 illustrates the level of pollution.

$$NIPI = \sqrt{\frac{PIave + PImax}{2}} \quad (3)$$

Where, PI_{ave} is the average pollution index; PI_{max} is maximum pollution index. NIPI ≤ 0.7 = no pollution; 0.7 < NIPI ≤ 1 = warning line of pollution; 1 < NIPI ≤ 2 = low level of pollution; 2 < NIPI ≤ 3 = moderate level of pollution; NIPI > 3 = high level of pollution.

The ecological risk index of HMs in compost/soil samples was estimated as:

$$ERI = \sum RI = \sum T_i \times PI \quad (4)$$

Where, RI is the risk index of HMs in compost/soil samples; T_i is the toxic-response factor given as: 30 for Cd, Cu, Ni and Pb, and 1 for Zn and Mn (Zheng-Qi et al. 2008).ERI < 150 = low ecological risk; 150 < ERI < 300 = moderate ecological risk; 300 < ERI < 600 = considerable ecological risk; ERI > 600 = very high ecological risk.

Bioaccumulation factor (BF)

BF was calculated to assess the amount of heavy metal accumulated by the castor oil plant during the remediation process. BF > 1.0 indicates high metal accumulation, while BF < 1.0 signifies low metal accumulation.

$$BF = \frac{Cm}{Cs} \quad (5)$$

Where Cm is the level of heavy metals determined in castor oil plants; Cs is the concentration of heavy metals in the contaminated soils.

Health risk assessment

The health risk assessment of heavy metals in soils was estimated for carcinogenic (cancer risk) and non-carcinogenic (hazard quotient and hazard index) adverse effects by adopting the models of the United States Environmental Protection Agency (USEPA 2007) presented in equations 6–11.

Estimated Daily Intake- ingestion (EDI_{ing}) = $\frac{CxIR_{ing}xFxEFxEDxCF}{BWxAT}$ (6)

Estimated Daily Intake- inhalation (EDI_{inh}) = $\frac{CxIR_{inh}xFxEFxED}{PEFxBWxAT}$ (7)

Estimated Daily Intake- dermal (EDI_{der}) = $\frac{CxCFxSAxAFxABSxFxEFxED}{BWxAT}$ (8)

Where, EDI_{ing}, EDI_{inh}, and EDI_{der} (mg kg^{-1} day^{-1}) = Estimated daily intake of HMs in soil through ingestion, inhalation and dermal contact, respectively; C = Concentrations of HMs in soil (mg kg^{-1}); CF = Conversion factor = 10^{-6} kg mg^{-1} (Zheng et al. 2015); EF = Exposure frequency = 250 days $year^{-1}$ (Li et al. 2009); ED = 30 years for carcinogenic effects (USEPA 2001); AT = Averaging time/life expectancy; for non-carcinogenic effects, AT = ED, while for carcinogenic effects, AT = 2190 days for children and 19,893 days for adults (enHealth 2012; WHO 2015); IR_{ing} = Ingestion rate of soil = 200 and 100 mg day^{-1} for children and adults, respectively; IR_{inh} = Inhalation rate of soil = 5 and 20 mg $kg^{-1}day^{-1}$ for children and adults, respectively (Taiwo et al. 2017); ABS = Absorption factor = 0.001 (Du et al. 2013); AF = Adherence factor = 0.07 mg cm^2 (USEPA, 2001); BW = Body weight = 15 and 60 kg for children and adults, respectively (USEPA 2001); F = Fraction of time spent outside per day = 6.94‰ (Li et al. 2009); PEF = Particle emission factor = 1.36 x 10^9 m^3 kg^{-1} (USEPA, 2001); SA = Exposed skin surface area = 5,000 cm^2 day^{-1} (Taiwo et al. 2020b).

Hazard quotient (HQ) = $\frac{EDI}{RfD}$ (9)

Non-cancer hazard index (HI) = $\sum_{i=1}^{n} HQ$ i = 1…n (10)

Where, EDI = Estimated daily intake of HMs in soil (mg kg^{-1} day^{-1}); RfD = Reference dose of HMs(mg kg^{-1} day^{-1}) obtained from Integrated Risk Information System (USEPA, 2007); n = numbers of analyzed HMs; the HQ > 1 indicates deleterious health effect, while the HQ < 1 illustrates no ill health effect.

Cancer risk (CR) = EDI x CSF (11)

Where, EDI = Estimated daily intake of HMs in soil (mg kg^{-1} day^{-1}); CSF = Cancer slope factor of HMs (mg^{-1} kg day) (USEPA 2007).

Results And Discussion

Levels of heavy metals (HMs) in composts

Table 1 shows the levels of HMs in the matured composts after the eighth week of preparation. Mn was the highest measured heavy metal in the composts, having values that varied from 179.6 ± 1.25 $mgkg^{-1}$ in Neem compost to 206.3 ± 5.01 mg kg^{-1} in Moringa compost. There were no statistical ($p > 0.05$) variations between Mn concentrations of Moringa and Pawpaw composts. However, the Mn levels of these two composts were significantly ($p < 0.05$) higher than those observed in Neem compost. Zn was the second most abundant heavy metal determined in all the composts. The highest significant ($p < 0.05$) level of Zn was observed in Neem compost (147.3 ± 6.05 mg kg^{-1}). Cd was determined below the detection limit (0.01 mg kg^{-1}) of the analytical instrument in all the compost types. The levels of Mn, Cu, Ni, Cd, and Pb in composts were less than the target values expected in unpolluted soil (Denneman and Robberse1990), indicating safe application for agricultural production of crops. However, the Zn values in all the composts were higher than the target value of 50 mg kg^{-1} expected in unpolluted soil (Denneman and Robberse 1990). The high levels of Zn in composts may suggest no detrimental effects because Zn is an important metal required by plants for growth (Andresen et al. 2018). The distribution pattern of HMs in composts followed the pattern of Mn > Zn > Cu > Pb > Ni > Cd.

Effects of composts on contaminated soils

Table 1
Metal contents in matured (8weeks) composts

		N	Mean	Std. Deviation	Minimum	Maximum	*Target value in soil (Denneman and Robberse 1990)
			mg kg^{-1}				
Cu	Moringa Compost	3	11.53a	0.26	11.25	11.75	36
	Neem Compost	3	12.08a	1.33	10.55	12.95	36
	Pawpaw Compost	3	14.32b	0.66	13.60	14.90	36
Zn	Moringa Compost	3	130.3a	5.01	126.5	136.0	50
	Neem Compost	3	147.3b	6.05	142.5	154.0	50
	Pawpaw Compost	3	126.8a	4.48	124.0	132.0	50
Ni	Moringa Compost	3	3.73a	0.56	3.10	4.15	35
	Neem Compost	3	6.55b	0.61	6.15	7.25	35
	Pawpaw Compost	3	4.32a	0.58	3.70	4.85	35
Mn	Moringa Compost	3	206.3b	5.01	202.50	212.0	476
	Neem Compost	3	179.6a	1.25	178.20	180.5	476
	Pawpaw Compost	3	214.7b	8.01	206.50	222.5	476
Cd	Moringa Compost	3	< 0.01a		< 0.01	< 0.01	0.8
	Neem Compost	3	< 0.01a		< 0.01	< 0.01	0.8
	Pawpaw Compost	3	< 0.01a		< 0.01	< 0.01	0.8
Pb	Moringa Compost	3	7.10a	0.13	7.00	7.25	85

Similar alphabets along the columns are not significant at $p > 0.05$.

	N	Mean	Std. Deviation	Minimum	Maximum	*Target value in soil (Denneman and Robberse 1990)
Neem Compost	3	5.62a	1.90	3.75	7.55	85
Pawpaw Compost	3	5.08a	0.52	4.50	5.50	85
Similar alphabets along the columns are not significant at $p > 0.05$.						

The levels of Cu, Zn, Ni, Cd, and Pb measured in the three composts were lower than the respective values of 59.9, 405, 6.64, 1.15, and 67.3 mg kg^{-1} reported by Reyes Pinto et al. (2020) in cow compost manure. Furthermore, higher values of these metals were also observed in the compost of tree litters (Reyes Pinto et al, 2020). Compare to the present study, Wierzbowska et al. (2018) documented higher levels of Cu (297 mg kg^{-1}), Zn (831 mg kg^{-1}), Mn (274 mg kg^{-1}), Pb (178 mg kg^{-1}), and Ni (35.2 mg kg^{-1}) in compost made from unsorted municipal wastes.

Analysis of compost produced from urban green wastes by Wierzbowska et al. (2018)also showed higher values of Cu (34.1 mg kg^{-1}), Zn (133 mg kg^{-1}), Mn (327 mg kg^{-1}), Pb (29.1 mg kg^{-1}) and Ni (15.5 mg kg^{-1}) than the corresponding HMs observed in this study. However, our past study on compost made from poultry manure and water hyacinth showed lower concentrations of Mn (4.38 mg kg^{-1}), Cu (0.18 mg kg^{-1}), and Zn (0.76 mg kg^{-1}) than those observed in this study. Similarly, Abubakari et al. (2017) reported low levels of Zn (1.8–3.2 mg kg^{-1}), Pb (0.7–4.2 mg $kg^{-1,}$) and Cu (1.1–6.8 mg kg^{-1}) in different composts from Ghana.

The formulated composts showed the HM values, which could possibly be applied for the safe agricultural production of food crops (Taiwo et al. 2022). The composts also indicated good suitability for remediation experiments.

Effects of composts on contaminated soils

The HMs determined in polluted dumpsite soils before and after remediation processes are presented in Table 2. The polluted soil revealed the highest concentration of Zn (1236 ± 256 mg kg^{-1}).All the HMs observed in the contaminated soils had concentrations greater than the target values expected in unpolluted soil (Denneman and Robberse 1990). Furthermore, the levels of HMs in polluted dumpsite soils were greater than the permissible limits of 10, 50, 200, and 250 mg $kg^{-1}$for Cd, Cu, Pb, and Zn, respectively (UNEP (2013). The distribution pattern of heavy metals in contaminated soil followed the decreasing order of Zn > Mn > Pb > Cu > Ni > Cd. The application of the three different composts resulted in a reduction of HMs in the contaminated soils. The HM data shown represented the average values of HMs from different treatments at 1:200 (5 g composts + 1 kg soil), 1:100 (10 g compost + 1 kg soil), 1:66 (15 g compost + 1 kg soil), and 1:50 (20 g compost + 1 kg soil). Moringa compost reduced the concentrations of Cu (219 ± 42.9 to 123 ± 24.7 mg kg^{-1}), and Mn (906 ± 34.6 to 258 ± 24.3 mg kg^{-1}) in the polluted soils.

Table 2
Levels of heavy metals in contaminated soils before and after remediation by composts and plant

	Treatment	N = 75	Mean	SD	Min	Max	*Target value in soil	+Permissible limit
			mg kg^{-1}					
Cu	Polluted Soil	3	219e	42.9	189	268	36	50(ER)
	Polluted Soil + Moringa Compost	12	123bc	24.7	98	191	36	50(ER)
	Polluted Soil + Moringa Compost + Castor oil plant	12	89.1a	9.39	70.4	108	36	50(ER)
	Polluted Soil + Neem Compost	12	158d	25.8	121	184	36	50(ER)
	Polluted Soil + Neem Compost + Castor oil plant	12	106b	15.9	77	134	36	50(ER)
	Polluted Soil + Pawpaw Compost	12	132c	15.5	110	152	36	50(ER)
	Polluted Soil + Pawpaw Compost + Castor oil plant	12	97.8a	15	75.4	126	36	50(ER)
Zn	Polluted Soil	3	1236d	256	953	1450	50	250(ER)
	Polluted Soil + Moringa Compost	12	338c	23.8	295	373	50	250(ER)
	Polluted Soil + Moringa Compost + Castor oil plant	12	265ab	21.2	234	309	50	250(ER)
	Polluted Soil + Neem Compost	12	316bc	38.8	263	377	50	250(ER)
	Polluted Soil + Neem Compost + Castor oil plant	12	224a	29.5	197	291	50	250(ER)
	Polluted Soil + Pawpaw Compost	12	355b	25.6	302	391	50	250(ER)
	Polluted Soil + Pawpaw Compost + Castor oil plant	12	263ab	37.7	206	305	50	250(ER)
Ni	Polluted Soil	3	15.6f	1.71	14.2	17.5	35	100(ER)
	Polluted Soil + Moringa Compost	12	11.6e	1.42	9.75	15.3	35	100(ER)

SD- Standard deviation, Min – Minimum, Max – Maximum, Similar alphabets along the columns are not significant at $p < 0.05$, * The target value indicates the maximum metal levels expected in unpolluted soil, ER –Ecological risk, HR- Health risk, *Denneman and Robberse (1990), +UNEP (2013)

	Treatment	N = 75	Mean	SD	Min	Max	*Target value in soil	+Permissible limit
	Polluted Soil + Moringa Compost + Castor oil plant	12	9.57cd	1.12	8.22	12.5	35	100(ER)
	Polluted Soil + Neem Compost	12	9.90d	1.34	8.20	12.6	35	100(ER)
	Polluted Soil + Neem Compost + Castor oil plant	12	8.40bc	1.43	5.76	11.1	35	100(ER)
	Polluted Soil + Pawpaw Compost	12	7.96b	1.44	6.20	10.5	35	100(ER)
	Polluted Soil + Pawpaw Compost + Castor oil plant	12	6.09a	0.68	5.24	7.00	35	100(ER)
Mn	Polluted Soil	3	906e	34.6	876	944	476	
	Polluted Soil + Moringa Compost	12	258b	24.3	226	321	476	
	Polluted Soil + Moringa Compost + Castor oil plant	12	218a	10.8	195	235	476	
	Polluted Soil + Neem Compost	12	286c	35.5	239	380	476	
	Polluted Soil + Neem Compost + Castor oil plant	12	221a	24.4	186	258	476	
	Polluted Soil + Pawpaw Compost	12	351d	24.6	319	399	476	
	Polluted Soil + Pawpaw Compost + Castor oil plant	12	266bc	24	226	293	476	
Cd	Polluted Soil	3	11.3f	2.80	8.63	14.2	0.8	10(ER)
	Polluted Soil + Moringa Compost	12	10.3e	0.55	9.26	11.2	0.8	10(ER)
	Polluted Soil + Moringa Compost + Castor oil plant	12	9.51d	0.47	8.84	10.3	0.8	10(ER)
	Polluted Soil + Neem Compost	12	8.06c	0.53	7.30	8.75	0.8	10(ER)
	Polluted Soil + Neem Compost + Castor oil plant	12	6.40bc	0.52	5.25	7.15	0.8	10(ER)

SD- Standard deviation, Min – Minimum, Max – Maximum, Similar alphabets along the columns are not significant at $p < 0.05$, * The target value indicates the maximum metal levels expected in unpolluted soil, ER –Ecological risk, HR- Health risk, *Denneman and Robberse (1990), +UNEP (2013)

	Treatment	N = 75	Mean	SD	Min	Max	*Target value in soil	+Permissible limit
	Polluted Soil + Pawpaw Compost	12	6.83b	0.50	6.10	7.75	0.8	10(ER)
	Polluted Soil + Pawpaw Compost + Castor oil plant	12	5.83a	0.50	5.21	6.80	0.8	10 (ER)
Pb	Polluted Soil	3	245b	85	159	329	85	200 (HR)
	Polluted Soil + Moringa Compost	12	107a	35	69.5	179	85	200 (HR)
	Polluted Soil + Moringa Compost + Castor oil plant	12	80.3a	16.9	45.5	98.3	85	200 (HR)
	Polluted Soil + Neem Compost	12	106a	29.3	67.8	166	85	200 (HR)
	Polluted Soil + Neem Compost + Castor oil plant	12	76.6a	7.30	69.3	94.5	85	200 (HR)
	Polluted Soil + Pawpaw Compost	12	101a	35.1	36.8	174	85	200 (HR)
	Polluted Soil + Pawpaw Compost + Castor oil plant	12	82.7a	10.6	64.8	101	85	200 (HR)

SD- Standard deviation, Min – Minimum, Max – Maximum, Similar alphabets along the columns are not significant at $p < 0.05$, * The target value indicates the maximum metal levels expected in unpolluted soil, ER –Ecological risk, HR- Health risk, *Denneman and Robberse (1990), +UNEP (2013)

Neem compost was most effective in cleaning up Zn in the contaminated soils by reducing the concentration from 1236 ± 256 to 316 ± 38.8 mg kg^{-1}. Pawpaw compost exhibited the highest removal ability for Ni (15.6 ± 1.71 to 7.96 ± 1.44 mg kg^{-1}), Cd (11.3 ± 2.80 to 6.83 ± 0.50 mg kg^{-1}), and Pb (245 ± 85 to 101 ± 35.1 mg kg^{-1}) in polluted soil samples. The trends of HMs reduction followed a similar pattern for the 'compost + castor oil plant' experimental setup. The castor oil plant further reduced the HMs in polluted soil, indicating the promising and complementary remediation potential of the plant. The pattern of metal removal by combined compost and plant was similar to the past study of Taiwo et al. (2016), where compost and kenaf plants were used to remediate HMs such as Cu, Zn, Mn, Fe and Cr in industrially contaminated soils.

After treatment, the Cu remediated in soils by different compost types and castor oil plants revealed levels greater than the permissible limit of 50 mg kg^{-1}. Similar trends were observed for Zn (except in Neem compost + castor oil plant treatment, 224 ± 29.5 mg kg^{-1}), and Cd in Moringa compost treated soil (10.3 ± 0.55 mg kg^{-1}), which had the values greater than the acceptable limits of 250 and 10 mg kg^{-1}, respectively. The high residual contents of Cu and Zn in remediated soil may pose no threats to plants and humans because they are essential metals required for plants' biological and metabolic processes

(Singh et al. 2011). It should be noted that Pb, Mn, and Cd (except in Moringa compost treated soil) were reduced in the contaminated soil to a safe level by the composts and the phyto-remediating plant. This study demonstrated the effectiveness of these three composts to clean up toxic HMs such as Cd and Pb from polluted soil.

Removal efficiency by composts and castor oil plants

Figure 1 shows the fractions of HMs remediated in polluted soils by ‘compost only’ and ‘compost + castor oil plant’ treatments. Moringa compost exhibited the highest removal efficiency (RE) for Cu (40–47%), Pb (54–57%), and Mn (69–71%). There were little variations observed in REs of HMs at various mixing ratio treatments of soil and composts. With the introduction of the castor oil plant, the REs of Moringa compost for Cu, Pb, and Mn increased to 60–63%, 66–77%, and 76–77%, respectively. This study showed that Moringa compost was more effective in removing Mn and Pb than Cu in polluted soil. In comparison to the previous study by Taiwo et al. (2016), Moringa compost was two times more effective in removing Cu from the soil than Water hyacinth compost. However, both studies showed similarities in the REs of Cu for combined ‘compost and plant’ treated soil. Neem compost revealed the highest REs for Zn (73–75%); the values that rose to 78–83% in the ‘compost + castor oil plant’ treatment. Cd and Ni were mostly removed by Pawpaw compost having REs of 34–37% and 40–43%, respectively. In the ‘compost + castor oil plant’ treatment, the RE followed the incremental pattern of 43–48% for Cd and 56–60% for Ni. The pattern of HMs removal by the composts followed the decreasing order of Zn > Mn > Pb > Cu > Ni > Cd; while the effectiveness of composts for cleaning up HMs followed the trend of Moringa > Neem > Pawpaw.

Pollution risk assessment

The data of different pollution risk indices of HMs in compost, contaminated, and remediated soil samples are presented in Fig. 2. The pollution index (PI), Nemerov integrated pollution index (NIPI), and ecological risk index (ERI)values were observed below the permissible limits in all the compost types. This indicates the safe utilization of the composts as potential soil biofertilizer/amendment for agricultural purposes (Ayilara et al. 2020). The contaminated soil revealed the PI values of 0.32, 1.3, 8.8, 16.6, 19, and 112 for Ni, Mn, Cu, Pb, Zn, and Cd, respectively. The dumpsite soils depicted significant contamination levels by HMs except for Ni. Cd showing the highest PI value is the major HM of environmental concern in the dumpsite soils. Our past studies have similarly shown high PI values for Cd in road dust from major traffic hotspots in Abeokuta, Osogbo, and Lagos, southwestern Nigeria (Taiwo et al. 2017; Taiwo et al. 2020a, b).Pb also exhibited a high PI value, which is about nine times greater than the recommended limit of 1.0. High PIs had been documented for Cd and Pb in mining and agricultural soils (Olayinka et al. 2017; Yahaya et al. 2021). The lowest PI values were observed in the soils treated with Pawpaw compost, and combined Pawpaw compost + castor oil plant.

Despite the treatments by the three compost types, and assisted castor oil plants; Cd, Cu, Ni, and Pb still retained the PI > 1.0, indicating a level of pollution. The reason might be due to the relatively low levels of the corresponding metals in the earth’s crust (Wedephol 1995).These data were adopted as background in the estimation of the PI. If the permissible limit values of these metals were used as background data; this would have reduced the PI values to the level below the pollution line. Therefore, the high PI values observed in the treated soils might not suggest the non-effectiveness of the composts for bioremediation of the dumpsite soils.

The Nemerov integrated pollution index (NIPI) is usually utilized to evaluate the risk pollution potentials of metals in soils (Zhao and Li 2013; Yang et al. 2013). The NIPI data of HMs in composts were less than 0.7 and thus portrayed no pollution risk, except for Zn (NIPI = 1.52) in Neem compost, which indicated a low pollution level. The NIPIs for Cu, Zn, Pb, and Cd in polluted soils were greater than 3.0, thus establishing a high level of pollution. After remediation, the NIPI decreased in all the treatments by 'compost only' and 'compost + castor oil plant', however, Cd and Pb still exhibited NIPIs > 3.0 in 'compost only' treated soil, while NIPIs < 3.0 were observed in 'compost + castor oil plant' treatment. High NIPI has been reported in polluted soil due to smelting and mining activities (Yang et al. 2013).

The activities of the past mining as well as indiscriminate disposal of unsorted wastes such as metallic substances at the dumpsite might be responsible for the high NIPI values observed for Cd. However, Mn (with NIPI < 1.0) was completely reduced to no pollution baseline level by the bioremediation and phytoremediation processes. Cd is the only HM with high ecological risk in both contaminated and remediated soils. Despite treatment, the ERI values > 600 were documented in all the samples. Pb was the second-highest HM of ecological risk having ERI < 150 in all the untreated and treated soil samples. This was similar to the recent study of Yahaya et al. (2021) at five villages in Zamfara state, northern Nigeria, where Cd and Pb made up 98.6% of the total potential ecological risk. High ERI was documented in agricultural soil analysed in China (Qi et al. 2020). In Kronum and Amakom dumpsite soils in Ghana, high levels of ERI > 600 were observed for Pb and Cd, indicating a very high ecological risk (Akanchise et al. 2020).

Bioaccumulation factor

Figure 3 presents the bioaccumulation factor (BF) of HMs in castor oil plants grown under the influence of the compost-spiked dumpsite soils. BF is usually adopted to assess the effectiveness of plants to accumulate and translocate metals from the soil or water (Koleli et al. 2015). The BF values were generally less than 1.0, indicating low metal translocation and accumulation in the castor oil plants. This, therefore, suggests that the castor oil plant has low tolerance and accumulation for the observed HMs (Yanqun et al., 2005). Pawpaw compost integrated with castor oil plant revealed the highest BF values for Cd (0.56–0.95) and Ni (0.62–0.91), while Neem compost + castor oil plant treatment showed the greatest accumulating affinity for Pb (0.13–0.30) and Zn (0.35–0.89).Moringa fortified compost + castor oil plant had the highest BF levels for Cu (0.16–0.28) and Mn (0.15–0.31).

The BF followed the decreasing order of Cd > Ni > Zn > Mn > Pb > Cu. The implication of this is that castor oil plant may be best adopted for phytoremediation of Cd, Ni, and Zn in polluted soil; showing the BF values close to 1.0 at compost/soil mixing ratio of 1:50 (i.e., 20 g compost + 1 kg soil). The highest levels of BF were observed mostly in the compost/soil mixing ratio of 1:50, while the lowest BFs were documented in the treatment, 1:200 (i.e., 5 g compost + 1 kg soil). This indicates that the application rate of compost could affect the bio-accumulating potential of the plants during the integrated remediation process (Singh and Prasad 2015).

Health risk assessment

The health risk assessment for hazard quotient (HQ) and hazard index (HI) for HMs in contaminated and remediated soils are shown in Table 3. The data represent the combined toxicological risk due to ingestion, inhalation, and dermal contact exposure routes for adults and children. The HQs from the individual routes of entry by adults and children are presented in Tables S1-S6 (in the supplementary material). Ingestion was the main route of exposure to HMs in the toxicological risk assessment data for contaminated and remediated soils, similar to the past studies (Taiwo et al. 2017; Taiwo et al. 2020a, b). The HQs of Cu, Zn, Cd, Ni, and Mn were less than 1.0 in both contaminated and remediated soils for children and adults, indicating no adverse health effects. However, Pb (in the contaminated soil exposed to by adults, and contaminated/ remediated soil exposed to by children) exhibited HQs > 1.0, therefore establishing non-carcinogenic adverse health effects. Pb is a pediatric toxicant with arrays of non-carcinogenic adverse health effects including low intelligence quotients, poor development in children, inhibition of haemoglobin, structural and functional defects to unborn babies (teratogenesis), damage to the central nervous system and peripheral nervous system, psychosis, damage to the kidney and gastrointestinal tract (GIT), (Duruibe et al., 2007; Taiwo et al., 2010; Taiwo and Awomeso 2017).

Table 3

Hazard quotient and hazard index values of heavy metals in contaminated and remediated soils

		RfD **mg kg^{-1} day^{-1}**	**Mean**	**Std. Deviation**	**Mean**	**Std. Deviation**
			HQ			
			Children		Adults	
Cu	Polluted Soil	0.037	0.375	0.073	0.026	0.005
	Polluted Soil + Moringa Compost	0.037	0.210	0.042	0.014	0.003
	Polluted Soil + Moringa Compost + castor oil plant	0.037	0.153	0.016	0.011	0.001
	Polluted Soil + Neem Compost	0.037	0.271	0.044	0.019	0.003
	Polluted Soil + Neem Compost + castor oil plant	0.037	0.181	0.027	0.012	0.002
	Polluted Soil + Pawpaw Compost	0.037	0.226	0.027	0.016	0.002
	Polluted Soil + Pawpaw Compost + castor oil plant	0.037	0.168	0.026	0.012	0.002
Zn	Polluted Soil	0.30	0.261	0.054	0.018	0.004
	Polluted Soil + Moringa Compost	0.30	0.071	0.005	0.005	0.000
	Polluted Soil + Moringa Compost + castor oil plant	0.30	0.056	0.004	0.004	0.000
	Polluted Soil + Neem Compost	0.30	0.067	0.008	0.005	0.001
	Polluted Soil + Neem Compost + castor oil plant	0.30	0.047	0.006	0.003	0.000
	Polluted Soil + Pawpaw Compost	0.30	0.075	0.005	0.005	0.000
	Polluted Soil + Pawpaw Compost + castor oil plant	0.30	0.056	0.008	0.004	0.001
Ni	Polluted Soil	0.02	0.049	0.005	0.003	0.000
	Polluted Soil + Moringa Compost	0.02	0.037	0.005	0.003	0.000
	Polluted Soil + Moringa Compost + castor oil plant	0.02	0.030	0.004	0.002	0.000
	Polluted Soil + Neem Compost	0.02	0.031	0.004	0.002	0.000
	Polluted Soil + Neem Compost + castor oil plant	0.02	0.027	0.005	0.002	0.000

		RfD mg kg^{-1} day^{-1}	Mean	Std. Deviation	Mean	Std. Deviation
	Polluted Soil + Pawpaw Compost	0.02	0.025	0.005	0.002	0.000
	Polluted Soil + Pawpaw Compost + castor oil plant	0.02	0.019	0.002	0.001	0.000
Mn	Polluted Soil	0.14	0.410	0.016	0.028	0.001
	Polluted Soil + Moringa Compost	0.14	0.117	0.011	0.008	0.001
	Polluted Soil + Moringa Compost + castor oil plant	0.14	0.099	0.005	0.007	0.000
	Polluted Soil + Neem Compost	0.14	0.130	0.016	0.009	0.001
	Polluted Soil + Neem Compost + castor oil plant	0.14	0.100	0.011	0.007	0.001
	Polluted Soil + Pawpaw Compost	0.14	0.159	0.011	0.011	0.001
	Polluted Soil + Pawpaw Compost + castor oil plant	0.14	0.120	0.011	0.008	0.001
Cd	Polluted Soil	0.001	0.715	0.178	0.049	0.012
	Polluted Soil + Moringa Compost	0.001	0.653	0.035	0.045	0.002
	Polluted Soil + Moringa Compost + castor oil plant	0.001	0.603	0.030	0.042	0.002
	Polluted Soil + Neem Compost	0.001	0.511	0.034	0.035	0.002
	Polluted Soil + Neem Compost + castor oil plant	0.001	0.406	0.033	0.028	0.002
	Polluted Soil + Pawpaw Compost	0.001	0.433	0.032	0.030	0.002
	Polluted Soil + Pawpaw Compost + castor oil plant	0.001	0.370	0.032	0.025	0.002
Pb	Polluted Soil	0.0035	4.439	1.541	0.306	0.106
	Polluted Soil + Moringa Compost	0.0035	1.942	0.633	0.134	0.044
	Polluted Soil + Moringa Compost + castor oil plant	0.0035	1.455	0.306	0.100	0.021
	Polluted Soil + Neem Compost	0.0035	1.912	0.531	0.132	0.037
	Polluted Soil + Neem Compost + castor oil plant	0.0035	1.388	0.132	0.096	0.009
	Polluted Soil + Pawpaw Compost	0.0035	1.825	0.637	0.126	0.044

		RfD mg kg^{-1} day^{-1}	Mean	Std. Deviation	Mean	Std. Deviation
	Polluted Soil + Pawpaw Compost + castor oil plant	0.0035	1.498	0.192	0.103	0.013
HI	Polluted Soil		6.251		0.430	
	Polluted Soil + Moringa Compost		3.031		0.209	
	Polluted Soil + Moringa Compost + castor oil plant		2.396		0.165	
	Polluted Soil + Neem Compost		2.922		0.201	
	Polluted Soil + Neem Compost + castor oil plant		2.149		0.148	
	Polluted Soil + Pawpaw Compost		2.743		0.189	
	Polluted Soil + Pawpaw Compost + castor oil plant		2.231		0.154	

The sum of HQs or the hazard index also indicated a value > 1.0, which suggests adverse health impacts through cumulative exposure to HMs in soil. Pb (64–71%) and Cd (11–25%) were the largest contributors to the adverse health effects. Correspondingly, the past study of Ogundele et al. (2019) demonstrated HI of 1.1 for children exposed to dumpsite soils from Ibadan, Oyo State, Nigeria. The study also showed Cd and Pb as the highest contributors to the HI. Furthermore, the HI > 1.0 was documented by Huang et al. (2015) for HMs (As and Cr) in soil samples from an abandoned dumpsite in China. The HQs and HIs (established for adults and children) of HMs in the contaminated soil were 2–3 times higher than those obtained from the remediated soil. This establishes the promising potential of the composts and castor oil plant for bioremediation of polluted soils. However, there is a necessity for further bioremediation of the treated soils to safeguard public health. This could possibly be achieved through the extension of the degradation period by two to four weeks. This study showed that children were more vulnerable to exposure to HMs in contaminated and remediated soils than adults. This, therefore, corresponds to the observations in the past studies (Taiwo et al. 2017; Sulaiman et al. 2019; Taiwo et al. 2020; Karimian et al. 2021).

Table 4 presents the combined (ingestion + inhalation + dermal contact) cancer risk (CR) values of HMs in contaminated and remediated soils. The CRs due to the individual route of exposure (i.e., ingestion, inhalation, dermal contact) are presented in Tables S7-S12 (in the supplementary information). Similar to the non-carcinogenic HQ data, ingestion was the predominant exposure medium to the carcinogenic metals. The CRs for Ni and Cd in both contaminated and remediated soils were generally higher than the safe limit of 1.0 x 10^{-4}, thereby establishing the carcinogenic effects on adults and children. The CR for Pb in contaminated soil was also higher than the permissible limit of 1.0 x 10^{-4}. Cd constituted 82–86%

of the total carcinogenic effects. The recent study by Karimian et al. (2021) established the carcinogenic effects ($CR > 1.0 \times 10^{-4}$) of HMs in landfill soil from Teran, Iran. Unlike the present study, Karimian et al. (2021) documented higher CR values for HMs exposed to in landfill soil by children than adults.

Table 4
Combined cancer risk values of heavy metals in contaminated and remediated soils

		SF kg mg^{-1}day^{-1}	Mean	Std. Deviation	Mean	Std. Deviation
			Children		Adults	
Ni	Polluted Soil	0.84	8.31E-04	9.11E-05	4.16E-04	4.56E-05
	Polluted Soil + Moringa Compost	0.84	6.26E-04	7.56E-05	3.13E-04	3.79E-05
	Polluted Soil + Moringa Compost + castor oil plant	0.84	5.10E-04	5.97E-05	2.55E-04	2.99E-05
	Polluted Soil + Neem Compost	0.84	5.27E-04	7.14E-05	2.64E-04	3.57E-05
	Polluted Soil + Neem Compost + castor oil plant	0.84	4.47E-04	7.62E-05	2.24E-04	3.81E-05
	Polluted Soil + Pawpaw Compost	0.84	4.24E-04	7.67E-05	2.12E-04	3.84E-05
	Polluted Soil + Pawpaw Compost + castor oil plant	0.84	3.24E-04	3.62E-05	1.62E-04	1.81E-05
Cd	Polluted Soil	6.1	4.36E-03	1.08E-03	2.18E-03	5.42E-04
	Polluted Soil + Moringa Compost	6.1	3.98E-03	2.13E-04	1.99E-03	1.06E-04
	Polluted Soil + Moringa Compost + castor oil plant	6.1	3.68E-03	1.82E-04	1.84E-03	9.10E-05
	Polluted Soil + Neem Compost	6.1	3.12E-03	2.05E-04	1.56E-03	1.03E-04
	Polluted Soil + Neem Compost + castor oil plant	6.1	2.48E-03	2.01E-04	1.24E-03	1.01E-04
	Polluted Soil + Pawpaw Compost	6.1	2.64E-03	1.93E-04	1.32E-03	9.68E-05
	Polluted Soil + Pawpaw Compost + castor oil plant	6.1	2.26E-03	1.93E-04	1.13E-03	9.68E-05
Pb	Polluted Soil	0.0085	1.32E-04	4.58E-05	6.61E-05	2.29E-05
	Polluted Soil + Moringa Compost	0.0085	5.78E-05	1.88E-05	2.89E-05	9.43E-06

		SF kg mg^{-1}day^{-1}	Mean	Std. Deviation	Mean	Std. Deviation
	Polluted Soil + Moringa Compost + castor oil plant	0.0085	4.33E-05	9.12E-06	2.17E-05	4.56E-06
	Polluted Soil + Neem Compost	0.0085	5.69E-05	1.58E-05	2.85E-05	7.90E-06
	Polluted Soil + Neem Compost + castor oil plant	0.0085	4.13E-05	3.94E-06	2.07E-05	1.97E-06
	Polluted Soil + Pawpaw Compost	0.0085	5.43E-05	1.89E-05	2.72E-05	9.48E-06
	Polluted Soil + Pawpaw Compost + castor oil plant	0.0085	4.46E-05	5.70E-06	2.23E-05	2.85E-06
ΣCR	Polluted Soil		5.32E-03		2.66E-03	
	Polluted Soil + Moringa Compost		4.66E-03		2.33E-03	
	Polluted Soil + Moringa Compost + castor oil plant		4.23E-03		2.12E-03	
	Polluted Soil + Neem Compost		3.70E-03		1.85E-03	
	Polluted Soil + Neem Compost + castor oil plant		2.97E-03		1.48E-03	
	Polluted Soil + Pawpaw Compost		3.12E-03		1.56E-03	
	Polluted Soil + Pawpaw Compost + castor oil plant		2.63E-03		1.31E-03	

Conclusion

This is a novel study that applied the three compost types formulated from the leaves of natural coagulants such as Moringa, Neem and Pawpaw mixed with poultry manure, and sawdust to clean up HMs in contaminated dumpsite soil. The composts revealed lower HM concentrations than the target value expected in soil. The composts were able to remove HMs in contaminated soil following the order of Zn > Mn > Pb > Cu > Ni > Cd. The pattern of HMs' reduction in polluted by the three composts followed the trend: Moringa > Neem > Pawpaw. The pollution and ecological risks of HMs in polluted and remediated dumpsite soils indicated high pollution levels having pollution index greater than 1.0 for Cd, Pb, Cu, and Zn. A high ecological risk index greater than 600 was observed for Cd. The health risk data of HMs in both contaminated and remediated soils through the combined route of exposure (ingestion + inhalation + dermal contact) revealed Pb (HQ > 1.0) as the major HM of health concern. The cancer risk values for Cd, Pb, and Ni in contaminated soil were higher than the permissible limit of 1.0 x 10^{-4}, thereby establishing carcinogenic effects. This study, therefore, recommends further treatment of the remediated soil to protect public health, and environmental safety. The present waste management policies in the study area should also be reviewed by the appropriate authority.

Declarations

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Conflicts of interest/Competing interests

The authors declare that they have no conflict of interest.

Availability of data and material (data transparency)

All data generated or analyzed during this study are included in this published article [and its supplementary information files].

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Figures

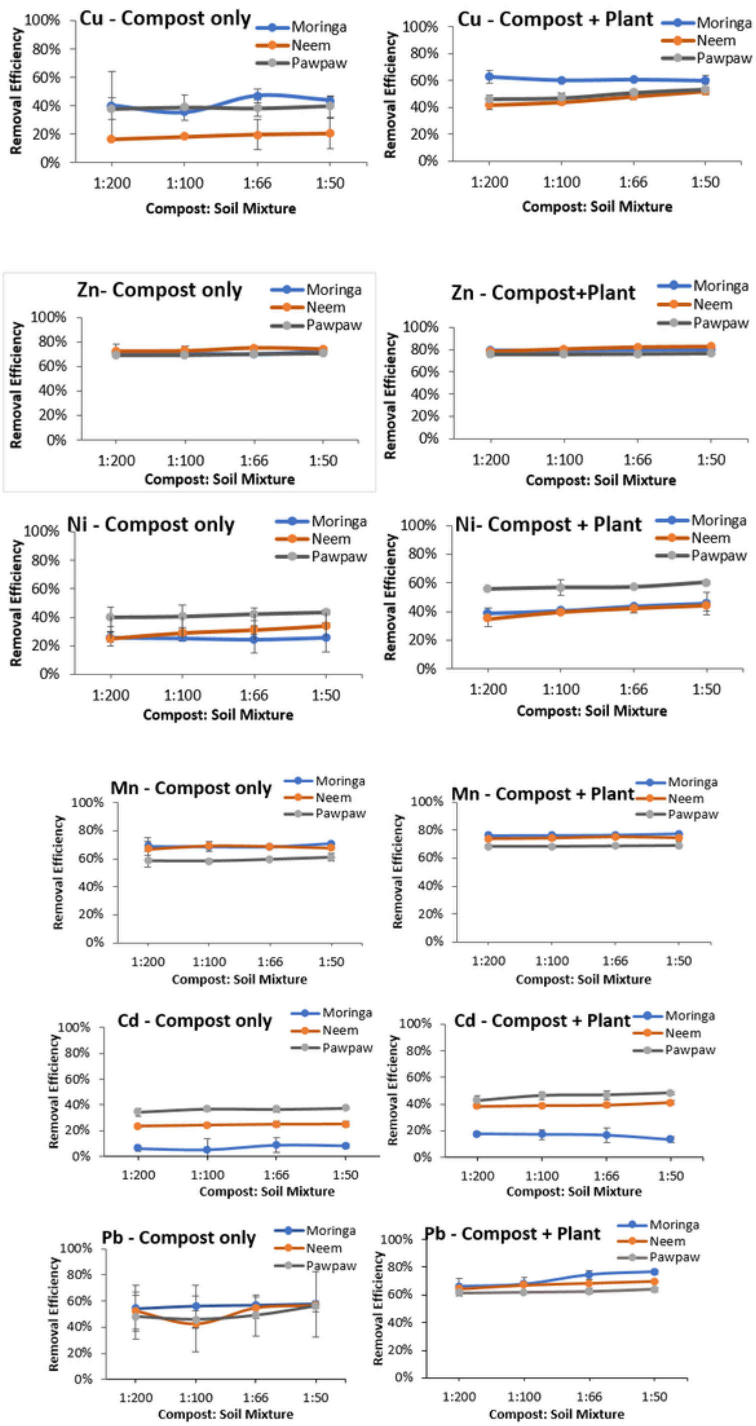

Figure 1

Removal efficiency of heavy metals in polluted soil by different composts and castor plant

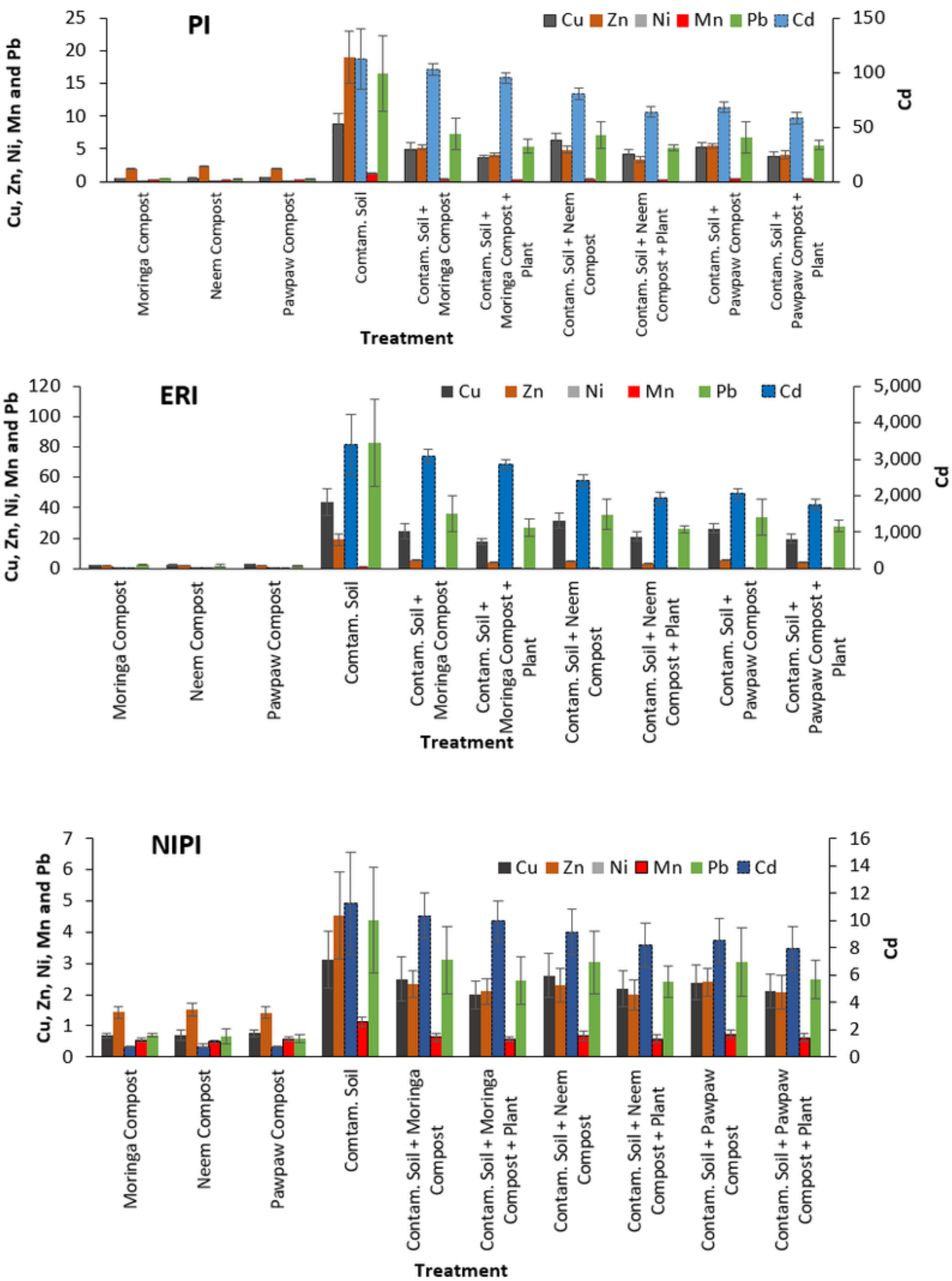

Figure 2

Pollution and ecological risk data of soil/compost samples

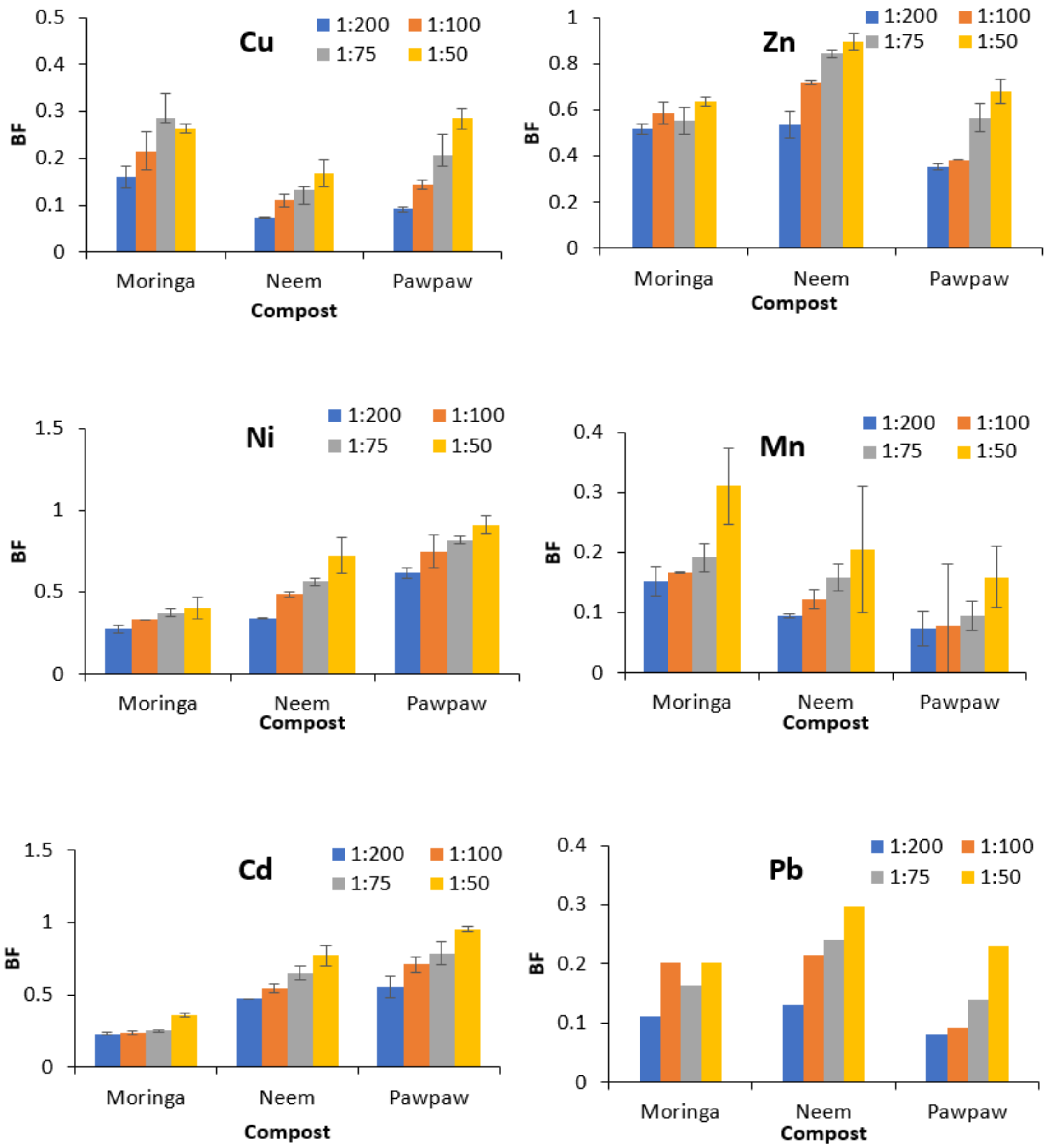

Figure 3

Bioaccumulation factors of heavy metals in plants under the influence of different compost type

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