Brazilian Journal
of Chemical
Engineering

ISSN 0104-6632
Printed in Brazil
www.scielo.br/bjce

Vol. 34, No. 03, pp. 715 – 726, July – September, 2017
dx.doi.org/10.1590/0104-6632.20170343s20150617

REDUCING THE LEACHABILITY OF NITRATE, PHOSPHORUS AND HEAVY METALS FROM SOIL USING WASTE MATERIAL

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(Submitted: April 14, 2015; Revised: March 17, 2016; Accepted: March 21, 2016)

Abstract – Contaminants like nitrate (NO_3), phosphorus (P) and heavy metals in water are often associated with agricultural activities. Various soil and water remediation techniques have been employed to reduce the risk associated with these contaminants. A study was conducted to examine the extent of leaching of heavy metals (Cd, Ni, Pb and Cr), NO_3 and P. For this purpose sandy and silt loam soils were amended with different waste materials, namely wood ash, solid waste ash, vegetable waste, charcoal, and sawdust. The soils were saturated with wastewater. Irrespective of the waste applied, the pH and EC of the amended soils were found to be greater than the control. Charcoal, sawdust and wood ash significantly decreased heavy metals, nitrate and phosphorus concentrations in the leachate. Treatments were more efficient for reducing Ni than other heavy metals concentrations. Waste amendments differed for heavy metals during the process of leaching. Heavy metals in the soil were progressively depleted due to the successive leaching stages. This research suggests that waste material may act as an adsorbent for the above contaminants and can reduce their leachability in soils.

Keywords: Heavy metals; leachability; nitrate; phosphorus; waste material

INTRODUCTION

The demand for water is continuously increasing in arid and semi-arid countries. Therefore, water of higher quality is preserved for domestic use while lower quality water is recommended for irrigation. Municipal wastewater is less expensive and considered to be an interesting source for irrigation for successful agricultural production. In this context, non-conventional water resources, such as treated wastewater, need to be applied to agriculture (Batarseh et al., 1989). In addition, wastewater is also a valuable source of plant nutrients and organic matter. However, the reuse of wastewater for irrigation may potentially create environmental problems if not properly treated and managed (Bahri and Brissaud, 1996).

With the continuous application of wastewater to field crops, the accumulation of nutrients may cause unfavorable effects on the soil as well as groundwater. Consequently, management of irrigation with wastewater should consider the nutrient content of the specific crop requirements. Application of wastewater to cropland and forested lands is an attractive option for disposal because it can improve the physicochemical properties of soils (Kiziloglu et al., 2007; Angin et al., 2005). Wastewater irrigation provides water, nitrogen (N) and phosphorus (P) as well as organic matter to the soils (Angin et al., 2005) , but there is a concern about the accumulation of potentially toxic elements such as cadmium (Cd), copper (Cu), iron (Fe), lead (Pb) and zinc (Zn) from both domestic and industrial sources. Wastewater treatment and disposal

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practices, depending upon the local conditions, should determine the management practices. Wastewater is composed of 99% water and 1% suspended colloidal and dissolved solids. Municipal wastewater contains organic matter and nutrients (N, P, K), inorganic matter or dissolved minerals, toxic chemicals, and pathogens (Mohammad and Mazahreh, 2003). Nitrate leaching into groundwater, P movement into surface water and groundwater in soil can be associated with the inefficient or excessive application of fertilizers and manures. The most important anthropogenic factor responsible for groundwater pollution is urban and industrial wastewater. Direct release of untreated effluents into the land and water bodies can potentially contaminate surface and groundwater, the soils and eventually the crops grown on these soils that affect the quality of the food produced. Several studies have been conducted to investigate the impact of agricultural, urban and industrial activities on water pollution, which lead to the contamination of the plant-animal-human food chain, and explore possible options for mitigating water pollution (Khurana and Aulakh, 2010; Farooqi et al., 2007; Jeong, 2001).

Most of the industries in Pakistan discharge untreated effluent containing variable concentrations of heavy metals (Manzoor, 2014). Metal concentrations vary with the type of industry in Pakistan. Sources of three metals (Cd, Cr, and Pb) are of particular interest as their concentrations in vegetable fields irrigated with sewage are approaching phytotoxic levels (Khan et al., 2011). Higher concentrations of heavy metals in wastewater also pose a health risk when ingested in higher quantities and can be fatal. Transfer of metals to humans through the food chain may have serious public health consequences (Sardar et al., 2013).

To curb this environmental deterioration arising from agricultural production, best management practices and remedial techniques have been evaluated and recommended. Among the remediation techniques, the use of chemical amendments to immobilize contaminants of interest has been studied (Cox et al., 2005). The choice of chemical amendments often depends on the availability, contaminant type and chiefly on the cost. The use of industrial by-products or waste materials from local industries offers an alternative and a complementary source.

Land applications of waste material may offer a rational waste management. The use of fertilizers on agricultural fields with elevated phosphorus and nitrate levels needs special management. The potential option for reducing the solubility of elements is the use of waste amendments. There is a need to know the effectiveness of waste material to decrease the leachability of nutrients from soil. Research studies evaluating the impact of the waste material on the reduction of nutrient leaching from soils have not been well reported. Therefore, this study aimed to compare the effectiveness of waste materials (charcoal, sawdust, wood-ash, vegetable waste, burned solid waste) to reduce the leachability of nitrate, phosphorus and heavy metal from soils.

MATERIALS AND METHODS

Samples collection

Five soil amendments: wood ash, solid waste ash, vegetable waste, sawdust and charcoal were collected locally. The materials were applied to assess the leachability of nitrate (NO3), phosphorus (P) and heavy metals [cadmium (Cd), nickel (Ni), lead (Pb) and chromium (Cr)], from two types of soils. The experiment was conducted in the facility of COMSATS Institute of Information Technology, Abbottabad, Pakistan. Municipal wastewater was collected from Abbottabad city. Sandy and silt loam soils were used for the leaching experiment.

Soil analysis

Soils were air-dried and sieved via a 2 mm sieve. Soil pH and electrical conductivity (EC) were measured in soil-water (1:5; w:v) suspensions by a pH meter (Model: HANNA HI 8520) and EC meter (Model: 4320 JENWAY), respectively. Water holding capacity (WHC) of the soil was determined by saturating the soil overnight. The next day the soil was weighed. The difference in the weight determined the WHC of soil. Particle size distribution of the soil was measured by the pipette method (Gee and Bauder, 1986). Water soluble calcium (Ca), magnesium (Mg), potassium (K), sodium (Na) and trace elements were determined using an atomic absorption spectrophotometer (AAS) (Model AAnalyst 700, Perkin Elmer) (Ure et al., 1993).

Leaching experiment

The leaching fraction (LF) of soils was calculated by dividing drained water by applied water. These materials were applied at the rate of 20 t ha-1 on the top of a PVC column (height 50 cm and diameter 15 cm). Each column was filled with 9 kg of soil. The material was mixed in the upper layer of the soil column (0-25 cm). Nitrate, P, and heavy metals were determined during five-stage sequential leaching. Wastewater was applied at a fixed leaching fraction of 0.3-0.4. After each irrigation event, leachate was collected within 24 h and immediately transferred to the laboratory in clean bottles for chemical analysis. The amount of water leached out from each treatment was recorded. The bottles used for collecting leachate were placed on the floor without stretching the drainage pipe. PVC columns were reshuffled after every 48 h to avoid microclimatic effects. After the termination of the experiment, soil samples were collected from each column layer to determine the concentrations of NO3, P, and heavy metals. Moisture contents in the soil were determined by oven drying 50 g of soil. NO3 was determined in the filtrate by UV spectrophotometry at 220 nm. Heavy metal concentrations in the filtrate were determined with an AAS. The P content of soil and leachate was determined colorimetrically by the molybdate blue method using a spectrophotometer at 710 nm. Data were statistically analyzed using Statview software (SAS, 1999), and results were expressed on the oven-dry basis. Means separations were done using LSD at $P < 0.05$.

RESULTS AND DISCUSSION

The physicochemical properties of soils are given in Table 1. Soil amendments significantly reduced heavy metal concentrations in the leachates, but the effect of each amendment was different. The cumulative concentration of Cd varied in the order of control > vegetable waste > burned waste > wood ash > charcoal > sawdust during the sequential leaching from both soils. Sandy soil leached out more Cd than silt loam soil. Sawdust, charcoal and burned waste significantly reduced Cd leaching from both soils (Fig. 1). The higher concentration of Cd in the leachate could be attributed to its concentration in the wastewater and lower adsorption sites in sandy soils. Emmerich et al. (1982) also reported a greater risk of Cd uptake by plants or more leaching to the ground water from sandy soil. Organic matter, hydrous oxides of iron, aluminum, manganese and clay minerals are the major components of the soil that may contribute to the retention of Cd in non-mobile or unavailable forms (Sposito and Page, 1985). Soil amendments may reduce the risk of Cd leaching or its uptake by plants (Verloo and Willaert, 1990). The application of inorganic materials such as mining waste and organic waste e.g. sewage sludge, organic manure, have been used to reduce the leaching of heavy metals from soils (Emmerich et al., 1982). Cadmium can exist in soils in several forms, depending upon the type and quantity of soil components, pH and the amount of Cd that has been applied (Christensen, 1984; Brummer, 1986). The amendment incorporation in the soil can fix Cd and can alter the equilibrium between existing forms of Cd, and thus reduce its solubility. Soil pH and organic matter are the soil factors that control Cd availability (Barancikova et al., 2004).

The results of the study clearly showed a significant difference in the leachability of Cr between soil types. In sandy soil, burned solid waste was found to be more effective than other soil amendments. Wood ash and sawdust were efficient in retarding Cr leaching from the silt loam soil (Fig. 2). In the control, sandy soil leached more Cr than silt loam soil. The behavior of Cr in the environment is complex. The availability of Cr is controlled by adsorption–desorption on mineral oxides. Chromium can be adsorbed on mineral oxides with exposed inorganic hydroxyl alkaline materials like wood ash, hydroxyapatite, or CaCO3 that increase soil pH above neutral on the surface since the adsorption increases with decreasing pH. The application of soil amendment can alter Cr mobility and its uptake by vegetation (Rai et al., 2004).

The concentration of Ni was reduced across all amendments as compared to the control soils (Fig. 3). Burned waste reduced Ni significantly as compared to the other waste materials in silt loam soil. Sandy soil did not differ for Ni among the waste treatments. Nickel was reduced in the leachate with enhanced leaching. Silt loam soil released less Ni during the leaching than sandy soil. The amount of Pb leached with water varied considerably in the order of control > vegetable waste > burned solid waste > wood ash > sawdust > charcoal in the sandy soil (Fig. 4). In the case of silt loam soil, charcoal efficiently decreased the Pb concentration in the leachate (Fig. 4). Treatments were systematically more efficient for reducing

Table 1. Physico-chemical properties of soils and wastewater.

Property	Unit	Sandy soil	Silt loam soil	Wastewater
Sand	%	87.0	81.6	-----
Silt	%	7.6	15.1	-----
Clay	%	5.4	3.3	-----
Organic matter	%	0.3	2.6	-----
Water holding capacity	%	14	18	-----
pH (1:5)		7.8	7.6	9.1
EC (1:5)	µ S cm-1	56.7	76.9	439
Ni	mg kg-1	2.6	4.0	0.78 mg L-1
Cd	mg kg-1	1.0	2.1	0.45 mg L-1
Cu	mg kg-1	0.4	0.5	0.24 mg L-1
Ca	cmolc kg-1	9.4	21.6	30.6 mg L-1
Mg	cmolc kg-1	5.4	16.0	21.0 mg L-1
K	cmolc kg-1	5.6	12.6	30.2 mg L-1
Na	cmolc kg-1	3.5	3.5	12.8 mg L-1

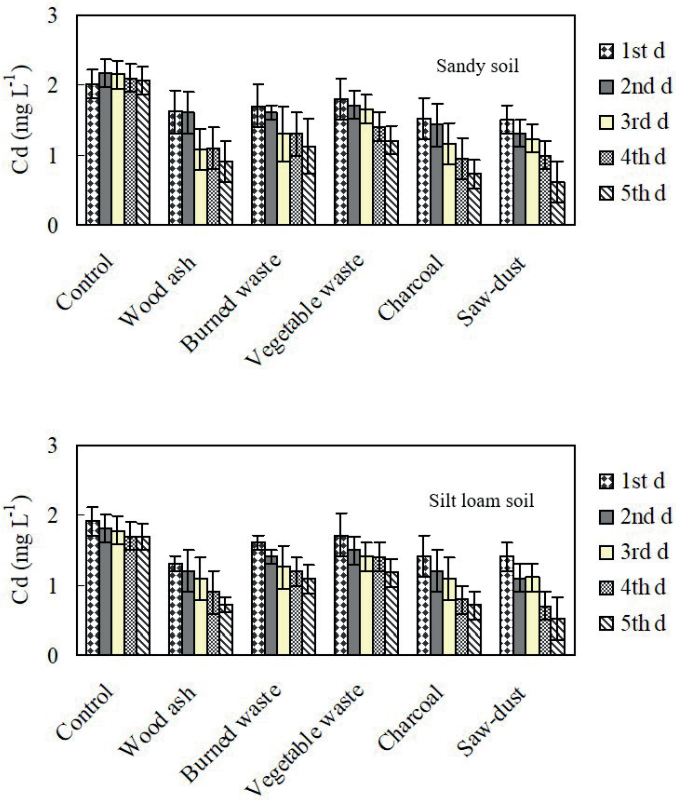

Figure 1. Leachability of Cd as affected by waste amendments after five-stage sequential leaching.

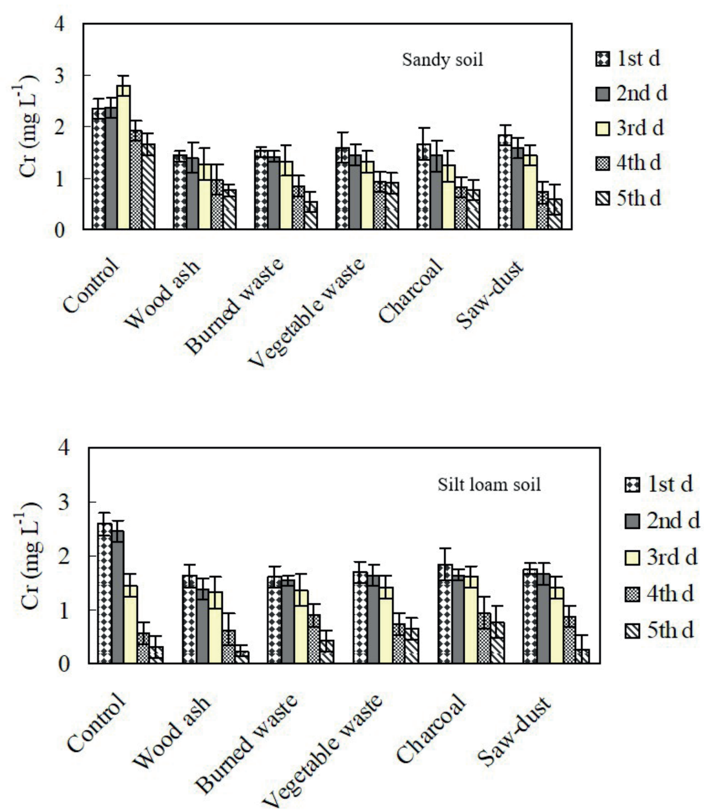

Figure 2. Leachability of Cr as affected by waste amendments after five-stage sequential leaching.

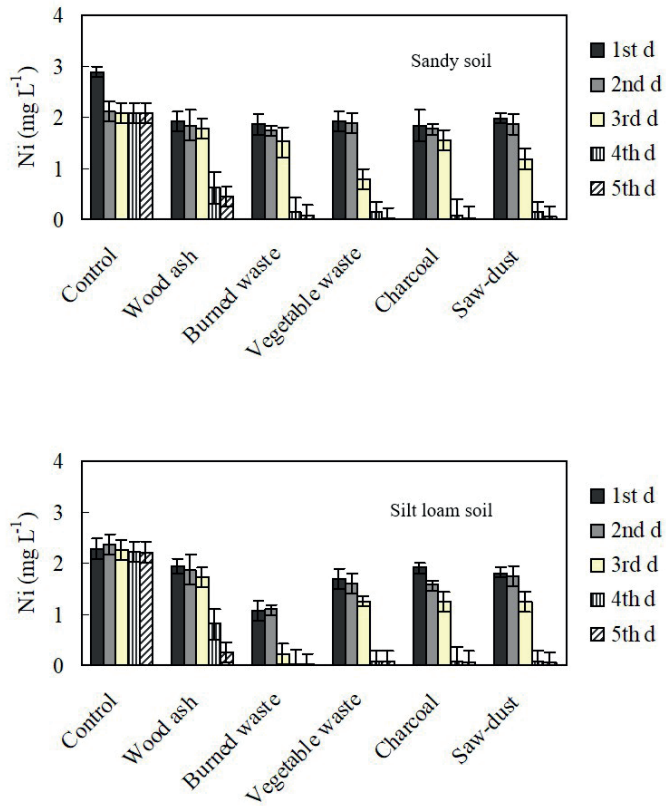

Figure 3. Leachability of Ni as affected by waste amendments during five-stage sequential leaching .

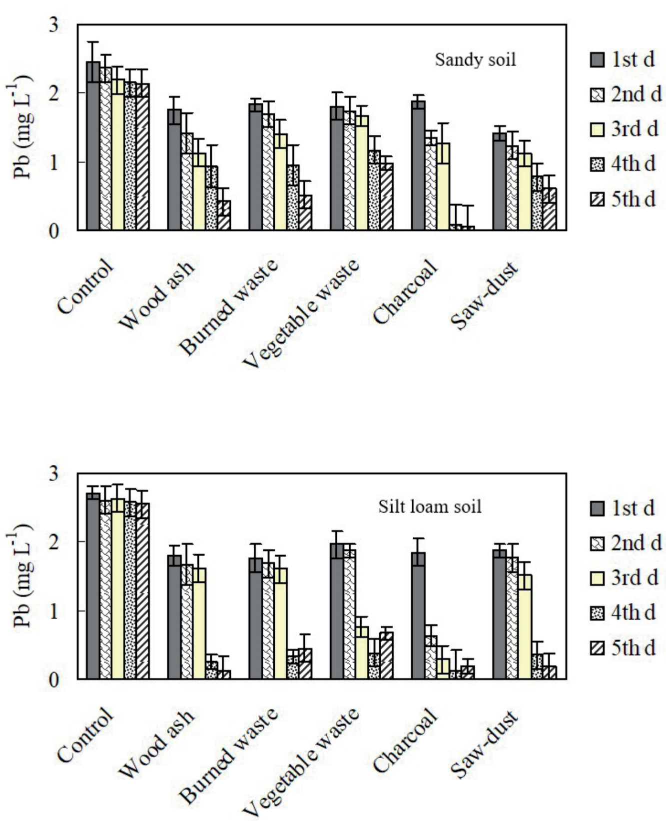

Figure 4. Leachability of Pb as affected by waste amendments during five-stage sequential leaching.

Ni as opposed to other heavy metals. Although waste amendments were different for different metals, there was a gradual change in the results during the leaching process. The most soluble heavy metals of the control soil were progressively depleted due to the continuous leaching during the experiment.

Cadmium reacts with organic matter (OM) by forming stable coordination complexes (Stevenson, 1994). Nickel, on the other hand, is related to the OM to a lesser extent and complexes mostly with inorganic substances (Hickey and Kittrick, 1984). The higher pH related to ash application and its potential to solubilize and mobilize OM make OM-facilitated metal transport a potentially significant mechanism for metal conveyance in the soil. The OM-complexed Cu is more mobile in soil than simple Cu under alkaline conditions (Chirenje and Ma, 1999). The leachability of heavy metals has been reported to be highly correlated with dissolved organic carbon leaching from the soil (Chirenje, 2000; Chirenje and Ma, 1999). The leachability of trace metals was reduced by changes in the pH of the soil columns, which reduced the solubilization and subsequent mobilization of OM from the soil profile.

Contaminant immobilizing amendments decreased trace element leaching and their bioavailability by inducing various sorption processes: adsorption to mineral surfaces, the formation of stable complexes with organic ligands, surface precipitation, and ion exchange. Precipitation as salts and co-precipitation can also contribute by reducing the contaminant mobility. Different sorption/dissolution processes were influenced by the factors: pH, redox potential, type of soil constituents, cation exchange capacity and a single mechanism rarely accounts for the immobilization of elements in soil (Bolan et al., 2003). Heavy metals are related to various soil components in different ways and these associations determine their mobility, and availability. The sorption behavior of Cd, Cr, Ni, and Zn in soils varied from soil to soil and was influenced by soil properties such as pH, organic matter, cation exchange capacity (CEC), and clay contents (McBride and Martinez, 1994).

Application of waste material to the silt loam soil resulted in a marked reduction in nitrate as compared to the sandy soil (Fig. 5). For soil amendment, the concentration of nitrate varied in the order of control < vegetable waste < burned solid waste < wood ash < sawdust < charcoal. A higher P level in the leachate was observed from control soil, and the lowest amount of P was seen in wood ash and sawdust amended sandy soil and burned solid waste amended silt loam soil (Fig. 6). Leaching of P enhanced up to 3 days of leaching and then reduced significantly. Results showed that waste amendments effectively reduced nitrate leaching from the soils. In the case of charcoal, nitrate concentration was higher in the upper layer of sandy soil and silt loam soil. A limited number of studies showed that charcoal from certain materials and processes can, in certain soils and over certain time scales, reduce the loss of nitrate from the soil by leaching (Ding et al., 2010) and also suppress N2O emissions (Yanai et al., 2007). Mechanistic explanations include increased ammonium adsorption through increased cation exchange capacity (CEC) (Liang et al., 2006); altered nitrification rates in forest soils expressing no net nitrification (DeLuca et al., 2006). Nitrate concentration was lower in the silt loam soil amended with sawdust. In the case of sandy soil, the concentration was higher. Burned solid waste and vegetable waste were also successful in retarding nitrate leaching but with vegetable waste the concentration was lower as compared to other amendments. In burned solid waste, a higher nitrate concentration was observed in the upper layer of sandy soil and the lower layer of silt loam soil. Wood ash contains very little carbon and nitrogen. Therefore, its application to the soil may reduce the total contents of C and N by increasing the solubility of organic carbon and the nitrification rate (Weber et al., 1985). As such, wood ash can be regarded as an N-free fertilizer, and is particularly suited for forest soils where nutrient unbalances, especially increased N/P ratios, can be expected due to acid depositions. Wood ash is essentially a direct source of other major elements, notably P, Ca, Mg and especially K in soils (Unger and Fernandez, 1990).

The heavy metal accumulation was more in the upper layer (0-25cm) than the lower layer of the silt loam soil (Table 2). The result was opposite for sandy soil due to the leaching of the heavy metals. Waste material increased the water holding capacity of the soil and retarded the leaching of metals and nutrients from the soils. Chromium concentration was higher in the upper layer and lower in the lower layer. All amendments retarded Cr leaching from both soils. Chromium concentration was greater in the silt loam in both layers as compared to the sandy soil. All amendments effectively retarded Cd leaching from the soil in both layers. Nickel and Pb concentrations were similar among soil layers. However, slight differences were noted between charcoal, sawdust and wood ash. These materials were effective in reducing the solubility of Ni and Pb. Therefore, higher concentrations in the extract were obtained from the amended soils.

Phosphorus accumulation occurred in the upper layer (0-25 cm) of the soil column after a leaching process (Table 3). Sawdust and charcoal were found to be efficient for P retention in the sandy soil but in silt loam soil wood ash and charcoal were found to be more efficient. The higher surface charge density of charcoal enables the retention of ions by its higher cation exchange capacity (Liang et al., 2006), and the higher surface area, internal porosity, and the presence of both polar and non-polar surface sites enabled charcoal to adsorb nutrients. The study concluded that the treatment process with waste amendments decreased P solubility; therefore, less leaching occurred.

Electrical conductivity and soil pH were enhanced in the lower zone of the soil column (Table 4). The increases

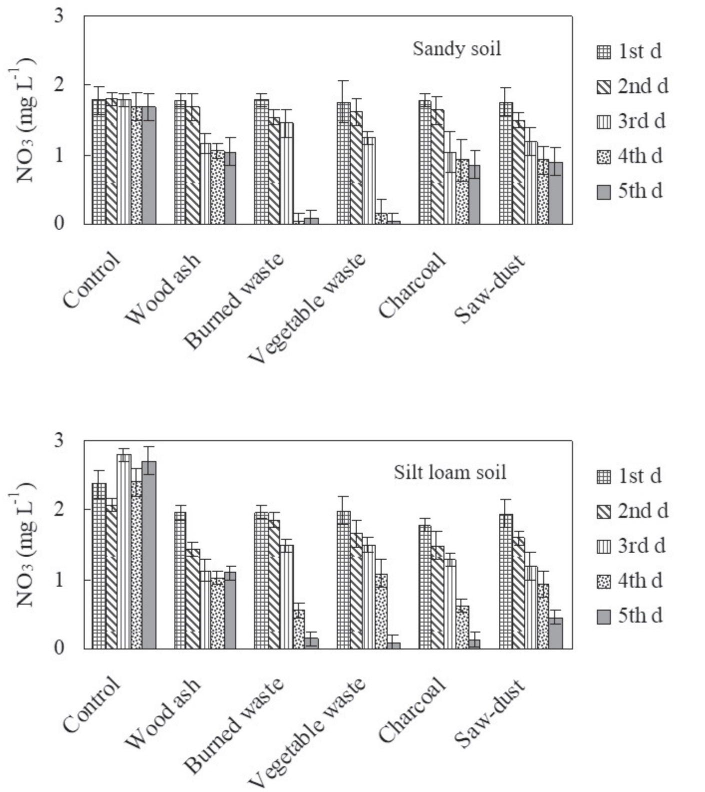

Figure 5. Leachability of NO3 as affected by waste amendments during five-stage sequential leaching.

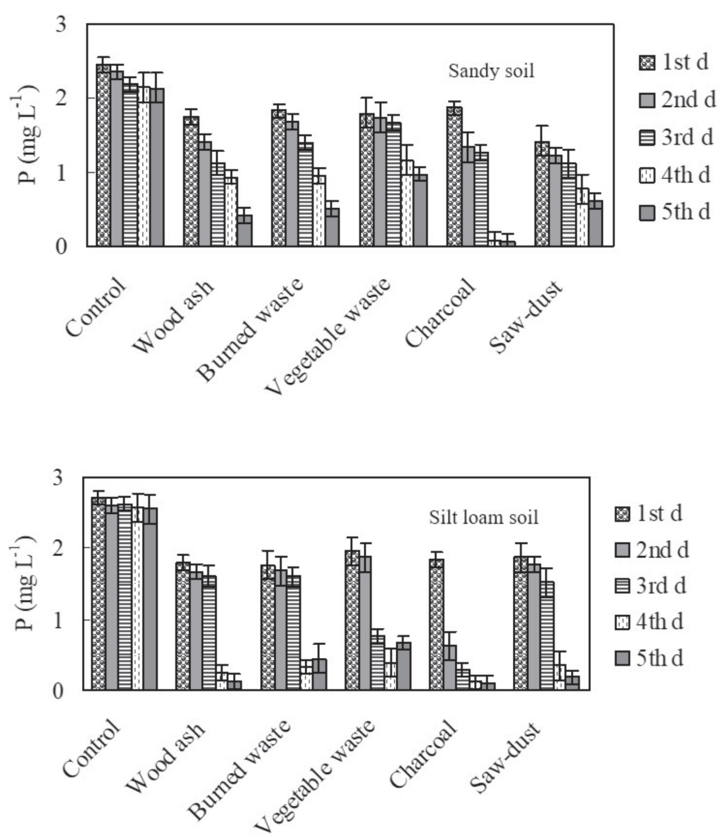

Figure 6. Leachability of P as affected by waste amendments during five-stage sequential leaching.

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Table 2. Heavy metal concentrations (mg kg-1) in soils after leaching.

Soil type	Waste material	Cr		Cd		Ni		Pb	
		0~25 cm	25~50 cm	0~25 cm	25~50 cm	0~25 cm	25~50 cm	0~25 cm	25~50 cm
Sandy	Control	1.38	1.54	1.71	1.43	1.66	1.46	1.56	1.66
	Wood ash	1.41	1.87	1.89	1.71	1.92	1.59	1.88	1.91
	Burned waste	1.41	1.91	1.99	1.52	1.74	1.61	1.72	1.88
	Vegetable waste	1.68	1.76	1.87	1.61	1.82	1.66	1.69	1.72
	Charcoal	1.97	1.73	2.13	1.98	2.31	1.99	1.88	2.33
	Sawdust	1.71	1.81	2.01	1.81	2.11	1.91	1.92	2.21
	LSD (0.05)	0.12	0.15	0.09	0.20	0.16	0.13	0.10	0.15
Silt loam									
	Control	1.14	1.99	1.15	1.09	1.43	1.57	1.62	1.71
	Wood ash	2.13	1.02	1.95	1.99	1.74	1.88	1.99	1.99
	Burned waste	2.54	2.12	1.82	1.65	1.92	1.68	1.71	1.82
	Vegetable waste	2.31	2.41	1.51	1.43	1.71	1.71	1.69	1.81
	Charcoal	2.54	2.14	2.33	1.86	2.13	2.51	1.99	2.37
	Sawdust	2.76	2.51	2.12	1.99	2.31	2.31	2.31	2.22
	LSD (0.05)	0.13	0.13	0.11	0.15	0.12	0.16	0.12	0.14

Table 3. Nitrate and phosphorus concentrations (mg kg-1) in soils after leaching.

Soil type	Waste material	NO3		P	
		0~25 cm	25~50 cm	0~25 cm	25~50 cm
Sandy	Control	1.13	1.44	0.75	0.79
	Wood ash	1.87	1.89	1.74	1.65
	Burned waste	1.72	1.77	1.44	1.35
	Vegetable waste	1.66	1.68	1.46	1.86
	Charcoal	2.13	2.11	1.89	1.72
	Sawdust	2.01	1.99	1.79	1.61
	LSD (0.05)	0.10	0.11	0.07	0.10
Silt loam					
	Control	1.42	1.46	0.84	0.89
	Wood ash	1.84	1.91	1.89	1.85
	Burned waste	1.72	1.79	1.54	1.43
	Vegetable waste	1.68	1.72	1.49	1.38
	Charcoal	2.31	2.12	1.94	1.88
	Sawdust	2.26	2.19	1.81	1.76
	LSD (0.05)	0.11	0.12	0.08	0.10

in EC level with wastewater application could be due to the addition and accumulation of salts. Earlier results obtained by Eneji et al. (2001) also showed an increase in the EC of soils. The pH of the amended soils was greater than the control. Biochar is postulated to function as a soil conditioner and fertilizer by increasing the cation exchange capacity (CEC), pH, and water retention, and by sequestering toxic heavy metals and gradually releasing limiting nutrients (Steiner et al., 2007). Various studies have shown that application of wood ash increases soil pH and decreases the exchangeable Al content of acid soils (Lerner and Utzinger, 1986). Electrical conductivity is a measure of the salt concentrations in the soil solution. This has been shown to increase with an increase in waste water application (Chang et al., 1991).

Baziramakenga et al. (2001), Chang et al. (1991) and Hernando et al. (1989) reported an increased soil water holding capacity after application of urban wastes. Chang et al. (1991) also noted increased hydraulic conductivity. Edwards et al. (2000) found that potatoes, sawdust and

Table 4. Electrical conductivity (EC) and pH of soil after leaching.

Soil type	Waste material	pH		EC (mS m-1)	
		Soil column layer			
Sandy		0~25 cm	25~50 cm	0~25 cm	25~50 cm
	Control	7.2	7.9	47.1	48.3
	Wood ash	7.3	8.1	50.1	48.1
	Burned waste	7.5	8.1	52.1	5.2
	Vegetable waste	7.8	8.3	48.2	50.1
	Charcoal	7.9	8.5	50.3	54.1
	Sawdust	7.7	8.4	49.1	52.4
	LSD (0.05)	0.23	0.18	1.12	2.43
Silt loam					
	Control	7.3	8.1	47.1	50.3
	Wood ash	8.1	8.3	48.2	53.1
	Burned waste	7.9	8.1	42.6	50.4
	Vegetable waste	8.1	7.9	53.1	58.3
	Charcoal	8.4	8.6	53.1	58.5
	Sawdust	8.4	8.7	50.2	58.1
	LSD (0.05)	0.12	0.15	1.14	1.24

manure increased soil moisture over untreated soil. In a non-aggregated soil, any effects on water retention are likely to be due to the properties of the material itself. However, in a more structured soil changes in both aggregation and pore size and continuity may affect the water holding capacity. Using thin sections, Giusquiani et al. (1995) also found that the stability of the pore system was improved in the treated soils and total porosity increased linearly with waste application rates.

CONCLUSIONS

It is concluded that waste amendments significantly altered the leaching behaviors of heavy metals, phosphorus and nitrate from soils. The variation of the pH and EC in the amended soil may be related to the solubility of nutrients. Charcoal, sawdust and wood ash acted as adsorbents that fixed heavy metals such as Pb, Ni, and Cd and reduced their leachability. Waste amendments differed for heavy metals leaching: charcoal < wood-ash < sawdust < burned solid waste. Application of waste material in the silt loam soil increased the nitrate concentration in soil as compared to the sandy soil. For soil amendment, the concentration of nitrate in the leachate varied in the order of control > wood ash > burned solid waste > sawdust > charcoal. The study concluded that the treatment process with waste amendments decreased phosphorus concentration in the leachate. This study indicated that waste amendments beneficially improve soil characteristics and have the potential to be applied as effective amendments in contaminated soils.

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