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# CONTAMINANT LIMIT (c/p index) OF HEAVY METALS IN SPENT OIL CONTAMINATED SOIL BIOREMEDIATED WITH LEGUME PLANTS AND ORGANIC NUTRIENT

**Udom, B. E.,[1] Ano, A. O.,[2] and Chukwu L. I.[2]**

[1]*Department of Crop and Soil Science, University of Port Harcourt, P.M.B. 5323, Port Harcourt, Rivers State, Nigeria.*

[2]*National Root Crops Research Institute, Umudike, P.M.B 7006, Umuahia, Abia State, Nigeria.*

Corresponding author's Email: ebassidy@yahoo.com

## ABSTRACT

Three legume plants (*Gliricidia sepium, Leucaena luecocephala* and *Calapogonium caerulean*) alone or in combination with 0.5% (w/w) poultry manure were tested for their ability to reduce the heavy metals and toxicity criteria of a sandy soil contaminated with 5% (w/w), (equivalent of 50,000 mg/kg) spent lubricating oil, each for two years. The oil and poultry manure led to build-up of Ni, Pb, Zn and Cu in the soils. The contaminant – pollution index (c/p index) calculated for Ni, Pb, Zn and Cu showed that at 3 months after oil contamination, concentration of Ni ranged from 0.03 to 0.024 mg/kg, Pb from 0.01 to 0.18 mg/kg, Zn from 0.27 to 0.60 mg/kg, and Cu from 0.12 to 0.81 mg/kg. The application of oil led to slight contamination of the soil with Pb, moderate to severe contamination with Zn and Cu, whereas, plots treated with poultry manure alone showed very severe contamination with Cu. Within 18 to 36 months, after oil contamination, the *Gliricidia*, *Leucaena* and *Calapogonium* combined with poultry manure reduced the toxicity levels of Ni, Pb, Zn and Cu. The *Gliricidia* was more effective in removal of these metals. At 36 months, the *Gliricidia sepium* combined with poultry manure reduced the Ni, Pb, Zn and Cu concentrations in the soil by 96%, 90%, 42%, and 50% respectively. Therefore, these legume plants are promising species in phytoremediation of oil contaminated sites and for general improvement of soil health. They can bioaccumulate high levels of these metals that could be toxic to other plants or organisms.

**Key words:** contaminant limit, heavy metals, bioremediation, legume plants, organic nutrients

## INTRODUCTION

Heavy metals are widely and usually applied to the elements such as Cd, Cr, Cu, Hg, Ni, and Zn, which are commonly associated with pollution and toxicity problems. It is a general collective term applying to the group of metals and metalloids with an atomic density greater than 6 g/cm$^3$ (Alloway, 1990). However, some of the elements in this group are required by most living organisms in small but critical concentrations for normally healthy growth. Those metals which are unequivocally essential, whose deficiency have adverse effects in normal living conditions include Cu, Mn, Fe and Zn for both plants and animals, Co, Cr and Se for animals, B and Mo for plants.

The toxicity effects caused by excess concentrations of these metals include competition for sites with essential metabolites, replacement of essential ions, and damage to cell membrane (Ernst, 1996). Zinc, Cu, Pb, Cd and Ni are generally the metals of greatest concern. Zinc, Cu and Pb are important because they can be phytotoxic. Whereas, concern for Cd and Ni arises from their possible entry into the food chain (Chaney, 1994). If these metals move too rapidly in a particular soil, they can pollute ground water supplies, especially in areas with high water table. It has been found that limiting Cu-contaminated soils to pH 7 can mitigate the toxicity by reducing the bioavailability of the Cu. (Alloway and Ayres, 1997). Copper is also highly toxic to the soil microbial biomass and this can affect various aspects of soil fertility.

Disposal of petroleum products with high heavy metal burdens on soil could result in nutritional imbalance, phytotoxicity and reduced crop production. Sediments and polluted soils enriched in heavy metals are subjected to erosion, which increase the risk of pollution in the surrounding areas. (Merkl *et al.*,2005). Excessive applications of metals bearing materials to the soil in whatever form have the potential of restricting plant growth and reducing crop yields. Ultimately, yield reduction has been the most important measure of phytotoxicity for agronomic species, since it affects the profitability of crop production and limits the utility of the land. Heavy metal accumulation and possible phytotoxicity are therefore, the most critical long-term hazards associated with disposal of petroleum products to land.

Contaminant limit (c/p index) has been used for assessment of toxicity risk of heavy metals in a soil site. The limit value is equivalent to maximum permissible risk level. It is intended to indicate the environmental quality to be achieved in a given period. (Kabata-Pendias and Pendias,1984).

Spent lubricating oil includes mono-and multi-grade crankcase oils from petrol engines, together with gear oils and transmission fluids with significant levels of heavy metals and other undesirable properties present in all petroleum products. Atuanya (1987), observed that Nigeria accounts for more than 87 million litres of spent oil annually and that most heavy metals such as Va, Pb, Ni, Cu and Zn which are below detection in unused lubricating oil, showed high values in waste motor oil. Contamination of open vacant plots and farm lands with petrol oils and grease is becoming more widespread problem than crude oil pollution (Anoliefo and Vwioko, 1995; Atuanya, 1987).

The use of plants and organic nutrients to modify soils contaminated with petrol oil and grease will provide a solution for metal stabilization and for minimizing erosion and associated risks. Phyto-remediation has shown great potential as an alternative treatment for remediation of heavy metal-contaminated soils and ground water (Chen and Cutright, 2001, Merkl *et al.*, 2005, Gallizia *et al.*, 2003, Harayama *et al.*, 2004). There is very little information available in the literature on the use of organic nutrients and legume plants to reduce the risk levels of heavy metals in contaminated sites. Moreover, the distinction between contamination and pollution range values of most metals in soils is uncertain. This study will provide valuable input data in the assessment of toxicity risk levels of Ni, Pb, Zn and Cu in soils.

## MATERIALS AND METHODS

The study was carried out at the University of Nigeria, Nsukka, Research Farm (Lat $06^0$ 52' N and Long $07^0$ 24' E) The soil is a *Typic kandiustult* (Nwadialo, 1989), derived from False-Bedded Sandstone (Akamigbo and Igwe, 1990). The mean sand, silt, and clay contents at the 0-30 cm depth were 820, 60 and 120 g $kg^{-1}$ soil respectively, (Table 1). The soil was impacted with equivalent of 50,000 mg $kg^{-1}$ soil (5% w/w) mono- and multi-grade

crankcase oils sourced from petrol and diesel engines. The oil was applied in a single dose each for two years. By the second year of the experiment, oil contaminated plots had equivalent of 100,000 mg $kg^{-1}$ soil, representing a total oil load of 10% (w/w). Some properties of the soil and spent oil used for the experiment are shown in Table 1.

Three (3) legume plants: *Calapogonium Caerulean, Gliricidia sepium,* and *Leucaena leucocephala,* alone or in combination with 0.5% (w/w), (equivalent of 50 mg $kg^{-1}$) of poultry manure were used to enhance biodegradation. The legume seeds and poultry manure were introduced to the plots at seven (7) days after the oil contamination and allowed for incubation, fourteen (14) days, before planting the maize crop. The *Calapogonium caerulean* was planted at 30 x 90 cm spacing, (giving density of 37,000 plants $ha^{-1}$), whereas the *Gliricidia* ssp and *Leucaena* ssp were planted at 1m x 90 cm spacings, (density of 11, 111 plants $ha^{-1}$). FASR-W maize (*zea mays*) variety was used as test crop, planted at 25 x 50 cm spacing, giving a density of 50,000 plants $ha^{-1}$. The legume plants used were regularly pruned to prevent shading of the maize crop and the biomass incorporated into the soil.

The experiment was laid out as a Randomized Complete Block Design (RCBD) with nine (9) treatments, viz: uncontaminated (control) soil (c), 5% spent oil ($A_5$), 5% spent oil + *Calapogonium* spp ($A_5$ + Ca), 5% spent oil + *Gliricidia spp* ($A_5$ + Gl), 5% spent oil + *Leucaena spp* ($A_5$ + Le), 5% spent oil + poultry manure ($A_5$ + Pm), 5% spent oil + *Calapogonium ssp* + 0.5% poultry manure ($A_5$ + Ca + Pm), 5% spent oil + *Gliricidia* ssp + 0.5% poultry manure ($A_5$ + Gl + Pm), 5% spent oil + *Leucaena* ssp + 0.5% poultry manure ($A_5$ + Le + Pm) with five (5) replications. The second application of 5% (w/w) spent oil was done 12 months after the first application.

***Soil sample and measurement of heavy metal***

Soil samples were collected from 0 – 30cm depth at 3, 12, 18, 24, 30 and 36 months after oil contamination, air-dried and crushed to pass through a 2 mm sieve. Heavy metals (Ni, Pb, Zn and Cu) were measured by atomic absorption spectrophotometer (AAS), after digesting 3g air-dried soil sample in concentrated $HClO_4$ – $HNO_3$ as described by (Carter, 1993). The values were compared with the widely used normal and critical levels set by Kabata – Pendias and Pendias (1984). The contaminant limit (c/p index) was calculated as the ratio between the heavy metal content in the soil and the toxicity criteria (the tolerable levels) and classified according to Lacatusu (1998) as: very slight (c/p index < 0.1), slight (0.1 – 0.25), moderate (0.26 – 0.50), severe (0.51 – 0.75) and very severe contamination) 0.76 – 1.00), and that of pollution range as: slight (1.1 – 2.0) , moderate (2.1– 4.0), severe (4.1 – 8.0), very severe (8.1 – 16.0) and excessive pollution (>16.0). The distinction between contamination and pollution range of heavy metals was established according to Lacatusu (1998). The legume plants used in this study are good bioaccumulators of heavy metals (Merkl *et al*. 2005), fast growing with massive root system, which penetrate the soil for several metres.

## RESULTS AND DISCUSSION

The soil is sandy loam with pH of 4.7 and low in total nitrogen (Table 1). The spent oil has high levels of Pb, Zn, and Cu and specific gravity of 0.87.

***Heavy metal concentrations***

The heavy metal concentrations pH values of the soil are shown in Table 2. There were build-up of Ni, Pb, Zn and Cu in plots contaminated with spent oil and similar build-up in plots treated with poultry manure. $P^H$ values ranged from 3.1 to 3.7 in spent oil-contaminated plots leading to increase in soil acidity between 2 and 36 percent relative to the control. This confirmed that spent oil and poultry manure are sources of heavy metals contamination in soils (Udom *et al.*, 2004,

Amadi *et al.*, 1993). In 3 months, Pb, Zn and Cu showed significant ($P < 0.05$) increases in the oil contaminated plots relative to control (Table 1). Plots treated with poultry manure (Pm) alone, showed the highest values of 17, 48, 43.6 and 48.3 mg/kg of Pb, Zn and Cu respectively, and similar trend at 6 months after oil contamination. In 12 months, the increase in Ni, Pb, Zn and Cu concentrations in the contaminated plots ($A_5$), were 158%, 702%, 118% and 446% respectively compared to the control (Table I). The high levels of these metals in the contaminated plots $A_5$ is an indication that Ni, Pb, Zn and Cu have been introduced to the soil via the spent oil and poultry manure applied. This confirmed the observations of Amadi *et al.* (1993) that most heavy metals such as Va, Pb, Ni and Fe which are below detection in unused lubricating oil showed high values in waste motor oil, and when disposed to soil, lead to contamination of the soil.

At high concentrations, these metals can block essential functional groups in the soil, displacing other metals ions and modify the active conformation of biological molecules in soil and plants, causing reduction in growth (Vangronsveld and Clijsters, 1994, Ernst, 1996).

Within 18 to 36 months, after oil application, the *Gliricidia*, *Leucaena* and *Calopogonuim* combined with poultry manure showed reductions in Ni, Pb, Zn, and Cu. At 36 months, the *Gliricidia sepium* combined with poultry manure significantly reduced the Ni, Pb, Zn and Cu concentrations in the soil by 96%, 90%, 42% and 50%, respectively, relative to the $A_5$ soil. This implies that these legume plants belong to the small group of plants reported by Brown *et al.* (1995), that can tolerate high levels of these metals.

**Table 1: Some characteristics of the soil (0-30cm depth), poultry manure and spent oil used in the experiment**

| Parameters | Unit | Soil | Poultry manure | Spent oil |
|---|---|---|---|---|
| Sand (200-50µm) | g $kg^{-1}$ | | | |
| Silt (50-2µm) | g $kg^{-1}$ | | | |
| Clay(< 2µm) | g $kg^{-1}$ | | | |
| Texture | - | Sandy loam | - | - |
| Organic carbon | g $kg^{-1}$ | 6.84 | 28.6 | 31.5 |
| Total N | g $kg^{-1}$ | 0.76 | 4.5 | 2.79 |
| pH ($H_2O$) | - | 4.7 | 6.5 | - |
| Specific gravity | - | - | - | 0.87 |
| Pb | mg $kg^{-1}$ | 1.48 | BDL | 286[b] |
| Zn | mg $kg^{-1}$ | 18.6 | 182.8 | 478[b] |
| Cu | mg $kg^{-1}$ | 7.0 | 46.1 | 164[b] |

BDL – Below detection limit b – Values in $mgl^{-1}$

### *Contaminant-pollution index*

The contaminant-pollution index(c/p index) calculated for Ni, Pb, Zn, and Cu concentrations in the soil are shown in Table 2. At 3 months after oil contamination, the contaminant-pollution index of Ni ranged from 0.003 to 0.024 mg/kg, and Pb from 0.12 to 0.81 mg/kg. The oil led to slight contamination of the soil with Pb, moderate to severe contamination with Zn and Cu, whereas $A_5$ + Pm showed severe contamination of the soil with Cu. This indicated that Zn and Cu are the major contaminant risk in spent oil impacted soils. At 12 months, Zn and Cu showed moderate to severe contamination in the $A_5$ plots, and slight to very slight contamination with Pb.

After 18 months when additional 5% and 0.5% levels of spent oil and poultry manure respectively, were applied to the soil, the $A_5$ soil showed severe risk levels of these heavy metals. This indicated that Ni, Pb, Zn, and Cu are commonly associated with contamination and toxicity problems in soils as earlier reported by Alloway and Ayres (1997). Copper at this level of concentration has been reported to inhibit plant growth, and interfered with several cellular processes in plants (Devez *et al.*, 2003), and Pb and Zn at these levels can suppress homeostatic mechanism in microorganisms (Ernst, 1996).

Within 18 to 36 months, after oil contamination, the *Gliricidia*, *Leucaena* and *Calapogonium* reduced the c/p index Pb, Zn and Cu (Table 3). The *Gliricidia sepium* alone was more effective in reducing toxicity levels of these heavy metals. The c/p index of Pb, Zn, and Cu in the treated soils showed gradual reduction in 18, 24, 30 and 36 months. This is an indication that these legume plants are promising in phytoremediation of heavy metal contaminated soils.

**Table 2: Heavy metal content of the top 0 – 30cm soil of oil contaminated site as influenced by the treatments.**

| Treatment | $P^H(H_2O)$ | $P^H$ (KCl) | Ni | Pb | Zn | Cu |
|---|---|---|---|---|---|---|
| | | | ← | mg kg$^{-1}$ | → | |
| | | | | **3rd Month** | | |
| $A_5$ | 3.7 | 3.3 | 2.4 | 15.3 | 31.4 | 39.0 |
| $A_5$ + Gl | 3.8 | 3.3 | 2.2 | 15.2 | 30.9 | 30.1 |
| $A_5$ + Le | 4.0 | 3.8 | 2.2 | 15.9 | 30.4 | 30.6 |
| $A_5$ + Ca | 3.9 | 3.5 | 2.1 | 15.0 | 30.1 | 29.2 |
| $A_5$ + Pm | 4.0 | 3.7 | 2.1 | 17.5 | 43.6 | 48.3 |
| $A_5$ + Gl + Pm | 4.2 | 4.0 | 2.1 | 17.2 | 42.1 | 32.6 |
| $A_5$ + Le + Pm | 4.4 | 4.0 | 2.1 | 17.3 | 41.5 | 33.1 |
| $A_5$ + Ca + Pm | 4.1 | 3.8 | 2.1 | 17.3 | 41.8 | 30.9 |
| C | 4.0 | 3.5 | 0.3 | 1.0 | 18.6 | 7.1 |
| LSD (0.05) | 0.67 | 0.53 | NS | 0.5 | 0.6 | 7.0 |
| | | | | **6th Month** | | |
| $A_5$ | 3.1 | 3.0 | 2.3 | 15.1 | 32.7 | 30.29 |
| $A_5$ + Gl | 3.6 | 3.4 | 1.1 | 15.0 | 30.8 | 28.3 |
| $A_5$ + Le | 3.8 | 3.5 | 1.2 | 14.8 | 31.0 | 28.6 |
| $A_5$ + Ca | 3.8 | 3.6 | 1.4 | 15.0 | 31.6 | 28.7 |
| $A_5$ + Pm | 4.1 | 3.7 | 2.1 | 17.1 | 44.7 | 36.0 |
| $A_5$ + Gl + Pm | 4.4 | 4.2 | 1.7 | 16.0 | 38.1 | 30.2 |
| $A_5$ + Le + Pm | 4.3 | 4.0 | 1.7 | 16.2 | 39.5 | 29.7 |
| $A_5$ + Ca + Pm | 4.4 | 4.0 | 1.9 | 15.2 | 39.2 | 29.1 |
| C | 4.3 | 4.0 | 0.2 | 1.2 | 18.3 | 7.1 |
| LSD (0.05) | 0.36 | 0.28 | 0.3 | 0.1 | 0.8 | 1.8 |
| | | | | **12th Month** | | |
| $A_5$ | 3.2 | 3.0 | 3.9 | 8.2 | 40.8 | 38.8 |
| $A_5$ + Gl | 4.1 | 3.8 | 2.4 | 6.9 | 33.1 | 26.9 |
| $A_5$ + Le | 3.8 | 3.6 | 2.4 | 6.8 | 34.5 | 27.7 |
| $A_5$ + Ca | 4.5 | 4.1 | 2.4 | 7.0 | 33.8 | 27.1 |
| $A_5$ + Pm | 3.8 | 3.6 | 2.5 | 9.9 | 46.4 | 33.4 |
| $A_5$ + Gl + Pm | 4.8 | 4.2 | 2.2 | 7.1 | 38.1 | 28.1 |
| $A_5$ + Le + Pm | 4.2 | 4.0 | 2.3 | 7.2 | 38.4 | 29.4 |
| $A_5$ + Ca + Pm | 4.1 | 4.0 | 2.4 | 7.1 | 38.5 | 29.5 |
| C | 4.3 | 3.9 | 0.2 | 1.0 | 18.7 | 7.1 |
| LSD (0.05) | 0.85 | 0.36 | 0.4 | 0.2 | 0.6 | 0.4 |

| | | | | | | |
|---|---|---|---|---|---|---|
| | | | | **18th Month** | | |
| $A_5$ | 3.1 | 3.0 | 3.9 | 28.0 | 45.8 | 42.6 |
| $A_5$ + Gl | 4.6 | 4.3 | 1.1 | 15.1 | 24.8 | 37.9 |
| $A_5$ + Le | 4.1 | 3.8 | 1.1 | 15.3 | 31.2 | 36.2 |
| $A_5$ + Ca | 4.3 | 4.1 | 1.3 | 15.2 | 31.6 | 31.3 |
| $A_5$ + Pm | 3.7 | 3.5 | 1.9 | 16.2 | 43.4 | 40.8 |
| $A_5$ + Gl + Pm | 4.8 | 4.4 | 1.0 | 15.3 | 39.1 | 36.7 |
| $A_5$ + Le + Pm | 4.6 | 4.4 | 1.3 | 15.4 | 30.4 | 39.2 |
| $A_5$ + Ca + Pm | 4.8 | 4.4 | 1.2 | 15.4 | 31.2 | 38.1 |
| C | 4.2 | 4.0 | 0.2 | 1.5 | 17.6 | 10.0 |
| LSD (0.05) | 0.81 | 0.3 | 0.1 | 0.1 | 0.4 | 0.1 |
| | | 3.0 | | | | |
| | | 4.3 | | **24th Month** | | |
| $A_5$ | 3.1 | 4.1 | 4355 | 3.9 | 28.1 | 44.7 | 38.7 |
| $A_5$ + Gl | 4.8 | 4.1 | 3866 | 0.9 | 15.0 | 21.5 | 26.0 |
| $A_5$ + Le | 4.3 | 3.5 | 3894 | 0.9 | 15.2 | 26.0 | 27.0 |
| $A_5$ + Ca | 4.4 | 4.4 | 4011 | 0.9 | 15.2 | 26.8 | 27.1 |
| $A_5$ + Pm | 3.8 | 4.6 | 4024 | 0.3 | 16.3 | 33.9 | 33.2 |
| $A_5$ + Gl + Pm | 4.8 | 4.5 | 3668 | 0.8 | 14.2 | 30.6 | 27.7 |
| $A_5$ + Le + Pm | 4.8 | 4.0 | 3699 | 0.8 | 14.3 | 36.6 | 28.4 |
| $A_5$ + Ca + Pm | 4.6 | 0.22 | 3681 | 0.8 | 14.6 | 30.1 | 28.0 |
| C | 4.2 | | 3079 | 0.3 | 1.4 | 17.7 | 7.1 |
| LSD (0.05) | 0.26 | | 27.9 | 0.0 | 0.1 | 0.2 | 0.4 |
| | | | | | **30th Month** | | |
| $A_5$ | 3.4 | 3.2 | 4429 | 4.0 | 28.0 | 44.3 | 39.0 |
| $A_5$ + Gl | 4.8 | 4.6 | 3776 | 0.8 | 19.7 | 21.2 | 21.2 |
| $A_5$ + Le | 4.6 | 4.3 | 3801 | 0.8 | 10.8 | 26.1 | 25.1 |
| $A_5$ + Ca | 4.8 | 4.7 | 3874 | 0.8 | 10.9 | 26.6 | 25.3 |
| $A_5$ + Pm | 3.7 | 3.6 | 3882 | 1.3 | 10.2 | 30.7 | 28.1 |
| $A_5$ + Gl + Pm | 4.8 | 4.6 | 3364 | 0.7 | 11.0 | 27.4 | 22.0 |
| $A_5$ + Le + Pm | 4.6 | 4.5 | 3386 | 0.7 | 10.1 | 29.3 | 22.6 |
| $A_5$ + Ca + Pm | 4.8 | 4.7 | 3400 | 0.1 | 10.0 | 29.1 | 23.9 |
| C | 4.2 | 4.0 | 3057 | 0.2 | 1.1 | 17.9 | 7.3 |
| LSD (0.05) | 0.14 | 0.22 | 22.9 | 0.1 | 0.1 | 0.1 | 0.6 |
| | | | | | **36th Month** | | |
| $A_5$ | 3.4 | | 3946 | 4.0 | 28. | 44.5 | 38.3 |
| $A_5$ + Gl | 4.8 | 3.3 | 3674 | 0.8 | 10.5 | 20.9 | 20.6 |
| $A_5$ + Le | 4.8 | 4.7 | 3689 | 0.8 | 10.6 | 25.3 | 22.1 |
| $A_5$ + Ca | 4.7 | 4.5 | 3880 | 0.8 | 10.7 | 25.7 | 22.7 |
| $A_5$ + Pm | 3.8 | 4.5 | 3981 | 1.0 | 10.1 | 30.0 | 27.0 |
| $A_5$ + Gl + Pm | 5.0 | 3.6 | 3119 | 0.6 | 2.8 | 25.9 | 19.1 |
| $A_5$ + Le + Pm | 4.8 | 4.8 | 3321 | 0.7 | 3.1 | 27.6 | 19.3 |
| $A_5$ + Ca + Pm | 5.1 | 4.5 | 3472 | 0.6 | 3.3 | 27.9 | 19.1 |
| C | 4.1 | 5.0 | 3015 | 0.2 | 1.0 | 17.8 | 7.3 |
| LSD (0.05) | 0.37 | | 121.3 | 0.0 | 0.0 | 0.1 | 0.1 |
| | | | | 100[a] | 100[a] | 70[d] | 60[a] |

a = Threshold tolerable limit (Kabata-Pendias and Pendias, 1984).

**Table 3: C/p index of the soil and some heavy metals as modified by the treatments**

| Treatment | Ni | Pb | Zn | Cu |
|---|---|---|---|---|
| | | **3rd Month** | | |
| $A_5$ | 0.024[a] | 0.16[b] | 0.45[c] | 0.65[d] |
| $A_5$ + Gl | 0.022[a] | 0.15[b] | 0.44[c] | 0.50[c] |
| $A_5$ + Le | 0.022[a] | 0.15[b] | 0.44[c] | 0.52[d] |
| $A_5$ + Ca | 0.021[a] | 0.15[b] | 0.43[c] | 0.47[c] |
| $A_5$ + Pm | 0.021[a] | 0.18[b] | 0.63[d] | 0.81[e] |
| $A_5$ + Gl + Pm | 0.021[a] | 0.17[b] | 0.60[d] | 0.55[d] |
| $A_5$ + Le + Pm | 0.21[a] | 0.18[b] | 0.59[d] | 0.55[d] |
| $A_5$ + Ca + Pm | 0.21[a] | 0.17[b] | 0.60[d] | 0.52[d] |
| C | 0.003[a] | 0.01[a] | 0.27[c] | 0.12[b] |
| | | **6th Month** | | |
| $A_5$ | 0.023[a] | 0.15[b] | 0.47[c] | 0.51[d] |
| $A_5$ + Gl | 0.011[a] | 0.15[b] | 0.44[c] | 0.47[c] |
| $A_5$ + Le | 0.012[a] | 0.15[b] | 0.44[c] | 0.48[c] |
| $A_5$ + Ca | 0.015[a] | 0.15[b] | 0.45[c] | 0.48[c] |
| $A_5$ + Pm | 0.021[a] | 0.17[b] | 0.64[d] | 0.60[d] |
| $A_5$ + Gl + Pm | 0.017[a] | 0.16[b] | 0.55[d] | 0.51[d] |
| $A_5$ + Le + Pm | 0.018[a] | 0.16[b] | 0.57[d] | 0.49[c] |
| $A_5$ + Ca + Pm | 0.019[a] | 0.15[b] | 0.56[d] | 0.49[c] |
| C | 0.002[a] | 0.02[a] | 0.26[c] | 0.12[b] |
| | | **12th Month** | | |
| $A_5$ | 0.039[a] | 0.08[a] | 0.58[d] | 0.65[d] |
| $A_5$ + Gl | 0.024[a] | 0.06[a] | 0.47[c] | 0.45[c] |
| $A_5$ + Le | 0.024[a] | 0.07[a] | 0.49[c] | 0.46[c] |
| $A_5$ + Ca | 0.024[a] | 0.07[a] | 0.48[c] | 0.45[c] |
| $A_5$ + Pm | 0.025[a] | 0.10[a] | 0.66[d] | 0.56[d] |
| $A_5$ + Gl + Pm | 0.022[a] | 0.07[a] | 0.55[d] | 0.47[c] |
| $A_5$ + Le + Pm | 0.023[d] | 0.07[a] | 0.55[d] | 0.49[c] |
| $A_5$ + Ca + Pm | 0.024[a] | 0.07[a] | 0.55[d] | 0.49[c] |
| C | 0.003[a] | 0.01[a] | 0.27[c] | 0.12[b] |
| | | **18th Month** | | |
| $A_5$ | 0.039[a] | 0.28[c] | 0.63[d] | 0.71[d] |
| $A_5$ + Gl | 0.011[a] | 0.15[b] | 0.36[c] | 0.63[d] |
| $A_5$ + Le | 0.011[a] | 0.15[b] | 0.45[c] | 0.60[d] |
| $A_5$ + Ca | 0.013[a] | 0.15[b] | 0.45[c] | 0.52[d] |
| $A_5$ + Pm | 0.019[d] | 0.16[b] | 0.62[d] | 0.68[d] |
| $A_5$ + Gl + Pm | 0.010[a] | 0.15[b] | 0.56[d] | 0.61[d] |
| $A_5$ + Le + Pm | 0.013[a] | 0.15[b] | 0.44[c] | 0.65[d] |
| $A_5$ + Ca + Pm | 0.012[a] | 0.16[b] | 0.45[c] | 0.65[d] |
| C | 0.003[a] | 0.02[a] | 0.25[b] | 0.17[b] |
| | | **24th Month** | | |
| $A_5$ | 0.039[a] | 0.28[c] | 0.64[d] | 0.65[d] |
| $A_5$ + Gl | 0.009[a] | 0.15[b] | 0.31[c] | 0.44[c] |
| $A_5$ + Le | 0.009[a] | 0.15[b] | 0.37[c] | 0.45[c] |
| $A_5$ + Ca | 0.009[a] | 0.16[b] | 0.38[c] | 0.45[c] |
| $A_5$ + Pm | 0.003[a] | 0.14[b] | 0.49[c] | 0.55[d] |
| $A_5$ + Gl + Pm | 0.008[a] | 0.15[b] | 0.45[c] | 0.46[c] |
| $A_5$ + Le + Pm | 0.008[a] | 0.15[b] | 0.44[c] | 0.48[c] |
| $A_5$ + Ca + Pm | 0.008[a] | 0.15[b] | 0.43[c] | 0.47[c] |
| C | 0.003[a] | 0.02[a] | 0.25[b] | 0.12[b] |

| | | | | |
|---|---|---|---|---|
| | | **30th Month** | | |
| $A_5$ | $0.040^a$ | $0.28^c$ | $0.63^d$ | $0.65^d$ |
| $A_5$ + Gl | $0.008^a$ | $0.11^b$ | $0.31^c$ | $0.36^c$ |
| $A_5$ + Le | $0.008^a$ | $0.12^b$ | $0.37^c$ | $0.42^c$ |
| $A_5$ + Ca | $0.003^a$ | $0.11^b$ | $0.38^c$ | $0.42^c$ |
| $A_5$ + Pm | $0.013^a$ | $0.10^b$ | $0.44^c$ | $0.47^c$ |
| $A_5$ + Gl + Pm | $0.007^a$ | $0.11^b$ | $0.39^c$ | $0.37^c$ |
| $A_5$ + Le + Pm | $0.007^a$ | $0.10^b$ | $0.42^d$ | $0.38^c$ |
| $A_5$ + Ca + Pm | $0.007^a$ | $0.10^b$ | $0.42^c$ | $0.40^c$ |
| C | $0.002^a$ | $0.011^a$ | $0.26^c$ | $0.12^b$ |
| | | **36th Month** | | |
| $A_5$ | $0.04^a$ | $0.28^c$ | $0.64^d$ | $0.64^d$ |
| $A_5$ + Gl | $0.01^a$ | $0.11^b$ | $0.30^c$ | $0.35^c$ |
| $A_5$ + Le | $0.01^a$ | $0.11^b$ | $0.36^c$ | $0.37^c$ |
| $A_5$ + Ca | $0.01^a$ | $0.11^b$ | $0.37^c$ | $0.38^c$ |
| $A_5$ + Pm | $0.01^a$ | $0.10^b$ | $0.43^c$ | $0.45^c$ |
| $A_5$ + Gl + Pm | $0.01^a$ | $0.03^a$ | $0.37^c$ | $0.32^c$ |
| $A_5$ + Le + Pm | $0.01^a$ | $0.03^a$ | $0.40^c$ | $0.32^c$ |
| $A_5$ + Ca + Pm | $0.01^a$ | $0.03^a$ | $0.40^c$ | $0.32^c$ |
| C | $0.002^a$ | $0.01^a$ | $0.26^c$ | $0.12^b$ |

a = Very slightly contaminated
b = Slightly contaminated
c = Moderately contaminated
d = Severely contaminated
e = Very severely contaminated

## CONCLUSION

It is indicated that *Gliricidia sepium*, *Leucaena leucocephala* and *Calapogonium cerulean* can mitigate toxicity levels of Ni, Pb, Zn and Cu and also reduce soil acidity. Within 18 to 36 months, there was general reduction in c/p index for Pb, Zn, and Cu in plots treated with legume plants. Consequently, they are excellent bioremediators of heavy metal contaminated soils and can be exploited in clean-up of heavy metal contaminated soils. However, the absence of any adverse growth effect on these plants highlight the danger of these metals being bioavailable to consuming animals or humans through the food chain.

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