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Effect of Sewage Sludge Addition on Soil Quality in Terms of Metal Concentrations

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Municipal authorities in developing countries often handle large volumes of sewage sludge with limited resources at their disposal. Increased industrialization and urbanization has further exacerbated the problem during the last few decades. High energy requirements for sewage incineration, scarcity of landfill sites and the phasing out of other environmentally unacceptable disposal options, such as ocean disposal have led many of these countries to look at options such as disposal of sewage sludge on agricultural land (Hue, 1988). Agricultural land application improves certain soil chemical properties, such as soil pH, soil organic matter (OM) content and nutrients (Tsadilas et al., 1995; Lopez Mosquera et al., 2000; Sastre et al., 2001). Sewage sludge has been reported to be effective as a fertilizer by increasing dry matter yield of various crops (Tsadilas et al., 1995). Sewage sludge can also improve soil physical properties, such as porosity, aggregate stability, bulk density and water retention capacity (Karapanagiotis et al., 1991).

There is an extensive body of research indicating that land application of sewage sludge has several benefits with relatively low environmental risks (National Research Council, 1996). However, concerns remain in the scientific community about potential effects of metals in the sewage sludge on soil quality (McBride, 1995). Heavy metals in sewage sludge originate from a number of different sources such as industrial and commercial wastes as well as domestic household wastes (e.g., feces, cleaners, paints, and wear and tear of utensils and equipment), and runoff from roads and roofs. Repeated application of heavy metal enriched sewage sludge can significantly increase the concentration of toxic metals in agricultural soils. Metal transfer from sewage sludge to soil and subsequently to plants pose potential health risks since this allows heavy metals to enter the food chain. Since metals persist in the soil for long periods of time, long-term health effects of sewage sludge application is a major cause for concern (McGrath, 1987). Most of the studies on the effects of heavy metals from sewage sludge application have been relatively short-term (1 to 5 year range). However, uncertainty exists about the long-term fate of heavy metals in sewage sludge. One of the possible long-term effects could be a gradual reduction of metal availability to plants due to their progressive immobilization into less soluble forms (Brown et

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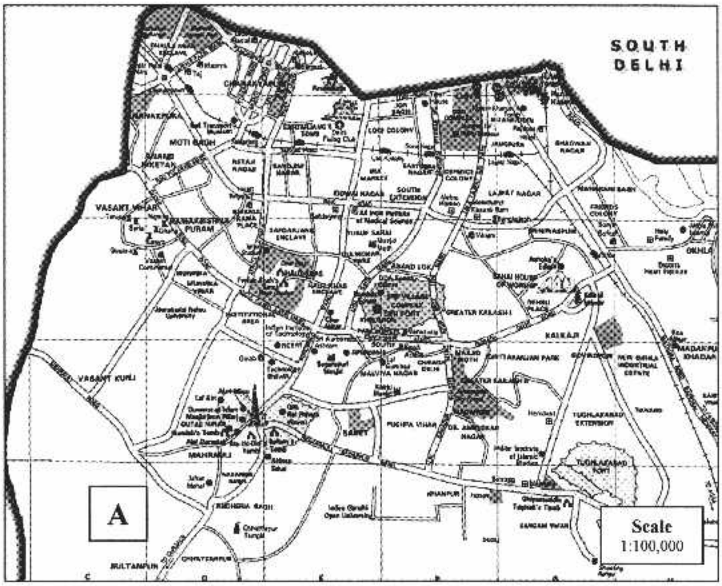

Figure 1. Site map showing the sampling location in South New Delhi ("A" marks the agricultural soil amended with sewage sludge)

al., 1998). The other possible effect could be an increase in metal bioavailability and leaching through OM mineralization in sewage sludge (Zhao et al., 1997). The current study was undertaken to evaluate the long-term effects of sludge application on the quality of an agricultural soil in terms of total concentrations of heavy metals. Since majority of the sludge metal regulations around the world are based on total concentrations of sludge metals (NSW EPA, 1997), evaluation of total metal levels may be useful as a global index of contamination. In this study, concentrations of seven "typical" sludge metals (Co, Cu, Pb, Zn, Ni, Cr, and Cd) were analyzed in a control soil (not affected by sewage sludge), and in soils amended with sewage sludge for over 50 years. The values were compared to the maximum allowable levels (MAL) of metals in soils, established by Kloke (1980) primarily on the basis of the negative effects of such high concentrations on the growth and development of plants. Finally, an interpretation of the soil quality was made by ranking the sludge-amended soils into five contamination classes (very slight to very severe contamination) and five pollution classes (slight to excessive pollution).

MATERIALS AND METHODS

Soil samples were collected from the surface horizon of an agricultural field in South New Delhi, India (Figure 1). This area was historically amended with domestic sewage sludge of unknown source or chemical composition for approximately 50 years. Although this practice has been stopped since 1990,

Table 1. Physicochemical properties of the control soil and the soils amended with sewage sludge.

ID	pH	OM (%)	CEC (cmol/kg)	Clay (%)	Fe+Al (ox) (mg/kg)
B-control	9.50	1.96	206.8	51.7	437
B1	8.84	2.23	267.5	42.1	1408
B2	8.65	2.15	264.4	42.4	1383
B3	8.43	2.11	281.8	43.4	768
B4	8.78	1.98	237.3	41.2	1727
B5	8.43	1.96	250.8	41.4	941
B6	8.60	1.71	262.2	42.3	774
B7	8.45	2.07	254.5	40.9	967
B8	8.67	2.06	249.2	39.9	918
B9	8.85	2.17	245.9	39.7	1325
B10	8.53	1.99	256.7	40.0	780

continued presence of high levels of metals in the soil is possible. Uncontaminated control soil was collected from a residential garden in the vicinity of the agricultural field. Grab samples were collected from the surface horizon (0– 15 cm) of 10 randomly picked sites. Handling of the soil samples followed the 1981 EPA/CE-81-1 protocol (Plumb, 1981). Collected samples were dried at 105°C, homogenized, and passed through a 2 mm sieve. Soils were characterized for pH using EPA Method 9050 (USEPA, 1986). Organic matter content was determined using the loss-on-ignition method and cation exchange capacity (CEC) by ammonium acetate method (Sparks, 1996). Amorphous (oxalate-extractable) Fe and Al oxide contents were determined using the Tamm's reagent (Sparks, 1996).

Total concentrations of heavy metals were determined by digesting the soil samples following EPA Method 3050B (USEPA, 1996). The digests were analyzed for Co, Cu, Pb, Zn, Ni, Cr, and Cd by ICP-MS. All analyses were carried out in triplicates and mean values are reported. Replicates had to fall within 95-105% to be considered acceptable. Recoveries of 90-110% of spikes and external standards were considered acceptable. Analyses that did not satisfy these QA/QC protocols were reanalyzed.

One-way analysis of variance (ANOVA) was performed to decipher if the metal concentrations in the soil samples were significantly different. Tukey's HSD multiple comparison tests were conducted to locate significant differences amongst the sampling sites. All statistical analyses were performed at a 0.05 significance level.

RESULTS AND DISCUSSION

Soil samples were typically clayey in nature (Table 1). The pH of the control soil was 9.5; application of sewage sludge decreased soil pH by approximately 1 pH

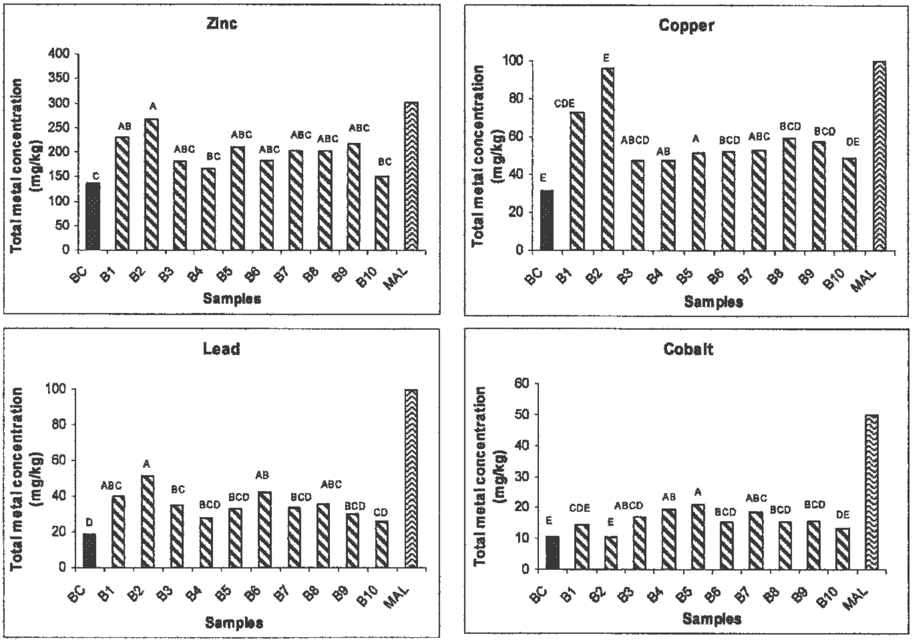

Figure 2. Total metal (Co, Pb, Cu and Zn) concentrations in sludge amended and control soil samples in comparison with the maximum allowable limit (MAL). Values not labeled with the same letter are significantly different from each other.

unit (average pH of treated soils was 8.6), which is considered to be better for plant growth compared to the high pH of the native soil. Soil OM content was only slightly higher in sludge-amended soils compared to the control (Table1), which is expected, since this area has been historically amended with sewage sludge which stimulates soil microbial activity. Since sewage sludge application was stopped more than a decade ago, much of the added OM is likely to have decomposed with time. There was also an insignificant increase in cation exchange capacity in the soils amended with sewage sludge compared to the control (Table1). The marginal increase in the CEC of soils could be reflective of the slightly higher OM content, because CEC of soils has been reported to be a direct function of OM content (Brady and Weil, 2001). There is a significant increase in oxalate-extractable Fe and Al contents (reflective of amorphous Fe/Al-oxide concentrations) due to addition of sewage sludge (Table1).

Total concentrations of metals in soils are reported in Figures 2 and 3. Concentrations of metals in sludge-amended soils (B1-B10) have been compared to the untreated control (BC) and the maximum allowable limit (MAL) for such metals as established by Kloke (1980), primarily based on growth and development of plants (Lacatusu, 1998). It is evident from Figures 2 and 3 that the concentrations of all metals studied were much greater in sludge-amended soils compared to the control soil. Such elevated metal levels probably reflect the metal

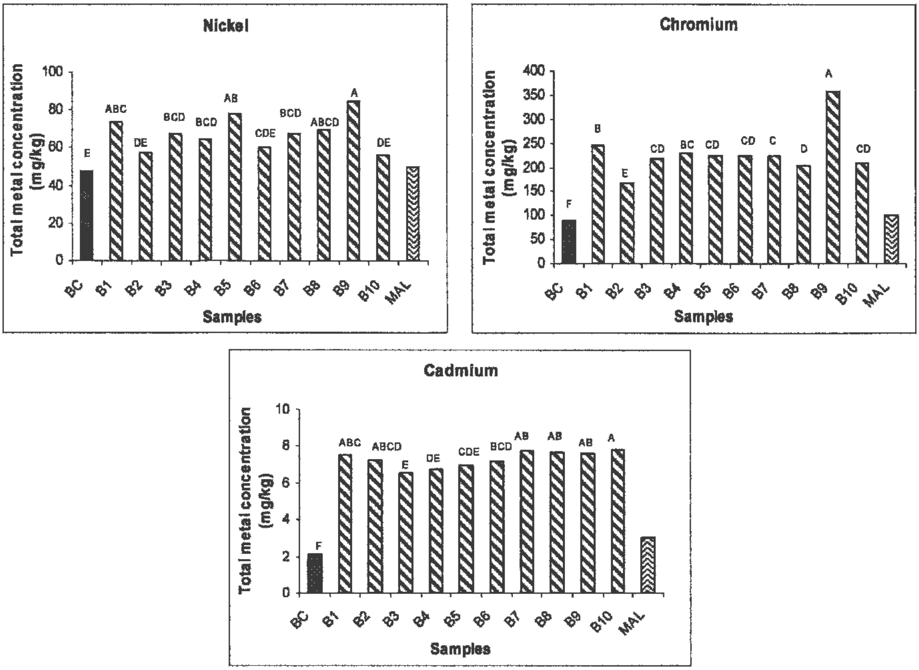

Figure 3. Total metal (Ni, Cr, Cd) concentrations in sludge amended and control soil samples in comparison with the maximum allowable limits (MAL). Values not labeled with the same letter are significantly different from each other.

composition of sewage sludge. However, this hypothesis could not be verified due to the unavailability of sludge samples, since the practice of sludge-amendment ended more than a decade ago. Total concentrations of Zn in the sludge-amended soil samples ranged between 150 and 266 mg/kg (Figure 2). Even at the highest concentration, soil Zn values are lower than the MAL. However, in the majority of the cases, Zn concentration in the treated soils was significantly higher than that in the control soil (135 mg/kg). Moreover, Zn concentrations in the sludge-amended soils were different between the sampling sites, which reflect a non-uniform mode of sludge application. Similar results were obtained for Cu, Pb, and Co (Figure 2). In all cases, total metal concentrations in sludge-amended soils were greater than that in control soils, but lower than the MAL. Hence, although sewage sludge was the source of additional metals in the treated agricultural soil, its effect on plant growth and development has not been tremendously detrimental. On the other hand, total concentrations of Ni in the sludge-amended soils varied between 56 and 84 mg/kg, which are higher than the MAL (50 mg/kg) (Figure 3). Concentration of Ni in the control soil was lower than the MAL. Similar patterns were observed for Cr and Cd (Figure 3); the sludge-amended soils had higher concentrations of these metals than the control soil and the MAL (100 mg/kg for Cr and 3 mg/kg for Cd). In order to classify soil quality based on metal concentrations in soils, Lacatusu (1998) defined the term "soil

Table 2. Schematic presentation of contamination/pollution index(c/p) for metals in soils (Lacatusu, 1998).

C/p	Classification	Sludge amended soils (mean C/p)
<0.1	Very slight contamination	
0.1-0.25	Slight contamination	
0.26-0.50	Moderate contamination	Co (0.31), Pb(0.35)
0.51-0.75	Severe contamination	Zn (0.67), Cu (0.58)
0.76-1.00	Very severe contamination	
1.1-2.0	Slight pollution	Ni (1.35)
2.1-4.0	Moderate pollution	Cd (2.43), Cr (2.31)
4.1-8.0	Severe pollution	
8.1-16.0	Very severe pollution	
>16.0	Excessive pollution	

contamination" as the content interval within which any measured value of metals has not or will not have immediate negative effects on plant growth and development or on other environmental components. The term "soil pollution" was defined as the content interval within which any measured value induces negative effects on some or all the environmental components. This is because high metal concentrations exceed the capacity of the soil to fix chemical elements (via colloidal adsorption and complexation), and hence induce negative effect on soil functions (Lacatusu, 1998). According to this methods of classification, values of C/p higher than 1 indicates "pollution", whereas those lower than 1 indicates potential contamination (Table 2). Using this classification, the sludge-amended soils were 'moderately contaminated' with regard to Co and Pb, "severely contaminated" with regard to Zn and Cu, and "slightly polluted" with regard to Ni. The soils are "moderately polluted" in terms of Cd and Cr (Table 2).

Plants use Zn and Cu as essential micronutrients; hence, the presence of these two metals at concentrations measured in the sludge-amended soils is not a point of concern. On the contrary, the presence of these elements may be beneficial for soils that have been cropped for many years, resulting in micronutrient deficiency. However, if the concentrations of these metals were higher than their maximum allowable limits, they would have potentially harmful effects on soil, plants and animals (Qiao et al., 2003). On the other hand, Pb and Co do not have any nutrient value, and, above a threshold concentration, can be highly toxic to the plants and the environment as a whole. Hence, presence of these metals even in "moderate contamination" range may be of concern. Of particular concern is the presence of Ni, Cd, and Cr in the "pollution" range. Not only are these metals toxic to plants, they tend to remain in the soil for long periods of time (McGrath, 1987). However, the evaluation of total metal concentrations in soils is only useful as a global index of contamination; it provides little or no indication of their bioavailability, mobility and reactivity (Walter et al., 2002). Moreover, the C/p index of Lacatusu (1998) was developed without considering the important physico-chemical characteristics of soils such as pH, OM and clay content, which tremendously impacts the uptake of metals by plants.

Recycling of nutrients contained in sewage sludge satisfies the sustainability principle. On the other hand, sewage sludge may contain high levels of certain toxic metals so that its use as a fertilizer may pose a risk to both the environment and human health, thereby contradicting the principle of environmental protection. The benefits and risks must be weighed against one another prior to land application of sewage sludge. While the short-term solution is to use only the sewage sludge of the highest quality, the long-term solution is to develop systems and techniques that satisfy criteria for both sustainability (nutrient recycling) and precaution (environmental protection). Results obtained from this study dealing with long-term, unplanned, indiscriminate usage of sewage sludge in agricultural fields indicate the potentially harmful effects of such practice on the environment in terms of soil metal enrichment.

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