

Repetitive and Residual Effects of Sewage Sludge Application on Extractability and Plant Uptake of Cu, Zn, Pb and Cd

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Abstract: Land application of sewage sludge is potentially beneficial as an inexpensive nutrient source. However, the composition of municipal sludge varies widely with the source of the material, and heavy metal concentrations often exceed the levels found in the native soils. Thus, problem with the use of sewage sludge may exist from high plant available concentrations and subsequent uptake of heavy metals by plants. A four year field experiment was conducted to examine lettuce (*Lactuca sativa* L.) and spinach (*Spinacia oleracea* L.) uptake of heavy metals from a sludge amended soil (fine loamy, mixed thermic Typic haplarigid) and the effects on soil pollution. Sludge rates were 0, 22.5, and 45 t • ha⁻¹. To determine the effect of repetitive sludge applications, applications were repeated on a third of the plots in the second year and on a fourth of the plots in the third year. Thus, total sludge amounts applied were 0, 22.5, and 45 t • ha⁻¹ in plots that received one application, 0, 45, and 90 t • ha⁻¹ in plots with two applications, and 0, 90, and 135 t • ha⁻¹ in plots with three applications. EDTA-extractable Cu, Zn, Pb, and Cd concentrations in soil, and metal concentrations in plant shoots and roots were determined. Copper, Zn, and Pb EDTA-extractable concentrations in soil and metal concentrations in plant parts increased significantly with sludge dosage. Residual sludge application had a significant effect on EDTA-extractable and plant uptake of the metals. However, EDTA-extractable and uptake of the metals decreased with time approaching that in the control plots. The amount of decrease in extractable metals in soil and concentrations in plants were significantly effected by sludge rate. The EDTA was an effective indicator of plant-available metal concentration changes due to sludge applications. Application of sewage sludge increased lettuce and spinach yields significantly.

1 Introduction

Increasing volumes of sewage sludge, environmental constraints and costs associated with alternative disposal methods are factors that have led to an increased interest in utilizing or disposing sewage sludge as a fertilizer on agricultural land. (Alloway *et al.*, 1991). The application of sewage sludge to land provides significant benefit through the addition of organic matter, nitrogen, phosphorus and certain essential trace elements to the soil (Chang *et al.*, 1987). However, as sludges may also contain high concentrations of pollutants, depending on the source and treatment of the waste, utilization as a cheap fertilizer on croplands also rise the risk of soil pollution. In particular, the accumulation of heavy metals in soils and their potential transfer into food chains or ground and surface waters has raised considerable concern that has induced many countries to control agricultural sludge application by legal regulations. There is however, concern that heavy metals in soils amended with sludge may increase after termination of sludge application creating a so called "time bomb effect" (McBride, 1995). On the other hand it has also been suggested that heavy metals released due to the organic matter mineralization are maintained in chemical forms and are not readily bioavailable i.e. the "plateau effect" (Dowdy *et al.*, 1994). The objective of this study was to determine cumulative and residual sludge application soil extractability and plant uptake of heavy metals.

Table 1 Selected chemical properties of the applied sewage sludge

pH	6.4	
EC	10.2	dS/m
organic Matter	31.0	%
Phosphorus	3.0	%
Nitrogen	2.6	%
Sodium	380	mg/kg
Potassium	285	mg/kg
Calcium	846	mg/kg
Magnesium	330	mg/kg
Zinc	788	mg/kg
Copper	596	mg/kg
Lead	429	mg/kg
Cadmium	3.4	mg/kg
Iron	13 600	mg/kg
Nickel	47	mg/kg

2 Materials and methods

The experiment was conducted at the Isfahan University of Technology, Isfahan, Iran research station site (32.32 N. 51.23 E.). The average annual rainfall and temperature at the site are 140 mm and 14.5°C, respectively. The soil is fine-loamy, mixed, thermic Typic Haplargid.

Lettuce (*Lactuca sativa* L. cv. Great Lakes) and spinach (*Spinacia oleracea* L. cv. Virofly attica) were chosen as experimental crop plants because of high local importance and also because they are known as heavy metal accumulator plants. The sludge used in this experiment was a secondary, anaerobically digested municipal sewage sludge from the city of Isfahan. Chemical data on the composition of the sludge are given in Table 1.

In 1996, air dried sewage sludge was applied at rates of 0 (control), 22.5, and 45 t • ha⁻¹ to plots of 4 m × 6 m size and mixed into the topsoil (20 cm depth). The treatments were replicated three times with 4m alleys between the replications. One week after sludge application lettuce and spinach were planted in 30 cm rows. The plants were irrigated weekly or as needed using the well water at the site. The irrigation water had an electrical conductivity of 1.2 dS • m⁻¹. Lettuce and spinach were harvested 90 and 70 days after planting, respectively. In the second year (1997) each plot was divided into two subplots (4 m × 4 m and 4 m × 2 m), and the sludge was applied to the larger subplot, using the same dosage as in the first year. After a week again, lettuce and spinach were planted. In the third year (1998), the same applications were once more repeated on the half each of these larger subplots and lettuce and spinach were planted in the same way as before. Thus, total sludge dosages were 22.5 and 45 t • ha⁻¹ for one year application, 45 and 90 t • ha⁻¹ for two years applications, and 67.5 and 135 t • ha⁻¹ for three years sludge applications.

In 1999 (fourth year) due to water shortage at the site because of a severe drought, we did not plant any crops. However, soil samples were collected similarly as in previous years to determine the effect of time on metal extractability. Although plant roots were also collected, at harvest plant yields were determined as fresh weight of aboveground biomass. Aboveground and root samples dried at 65 °C for 48 h and digested in a solution of 70% HNO₃ + concentrated HCl and 30% H₂O₂ for analysis of total Zn, Cu, Pb, and Cd by atomic absorption spectrometry (AAS). Soil samples were collected from 0 cm—20 cm depth, air dried, sieved (2 mm), and analyzed for EDTA extractable Zn, Cu, Pb, and Cd using AAS (Fujii and Cory 1986). All statistical analysis was performed using SAS version 6.10 for personal computers (SAS, 1993).

3 Results and discussion

3.1 Extractable metals

All sludge applications significantly increased soil EDTA extractable concentrations of Cu, Zn, and Pb. Extractable Cd concentration increased significantly only after a single $45 \text{ t} \cdot \text{ha}^{-1}$ sludge application (Fig.1a and Fig.2a) or multiple 22.5 and $45 \text{ t} \cdot \text{ha}^{-1}$ applications, but not after a single $22.5 \text{ t} \cdot \text{ha}^{-1}$ application. Extractable metal concentrations were highest in the first year of sludge application. In general, EDTA extractable metal concentrations thereafter decreased from year to year, approaching the concentrations of the control plots. The calcareous soils in central Iran are mostly low in plant available Cu and Zn. These results show that one application of $22.5 \text{ t} \cdot \text{ha}^{-1}$ sewage sludge can increase the EDTA concentrations of these metals for at least four years (Fig.1a and Fig.2a).

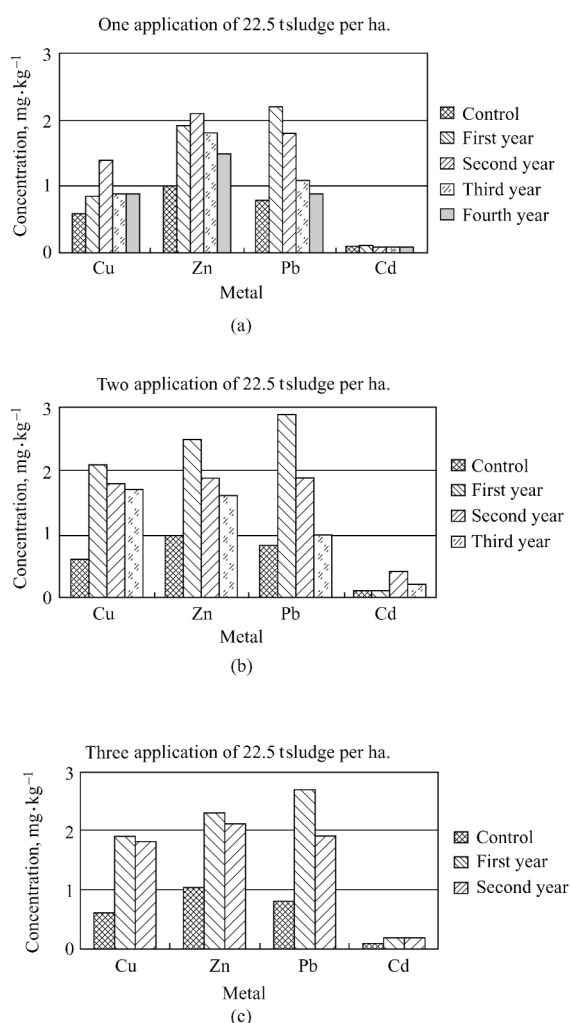


Fig.1 EDTA extractable metals in soil after application of $45 \text{ t} \cdot \text{ha}^{-1}$ sludge in (a) 1996, (b) 1996 and 1997, and (c) 1996, 1997 and 1998

Repetitive sludge applications of 22.5 and $45 \text{ t} \cdot \text{ha}^{-1}$ (two or three years applications in row) increased EDTA extractable metal concentrations in the soil significantly more than only single applications (Fig.1b,c and Fig. 2 b,c). Again similarly as in the single application EDTA extractable metal concentrations dropped in the years following the first sludge application back towards control levels. The

results in this study clearly show that in general extractable metal concentrations were highest first year after the initial sludge application but decreased thereafter with time. However, the amount of decrease in EDTA extractable metals was dependent on sludge application load. Similar results were reported by other investigators (Logan *et al.*, 1997; Chang *et al.*, 1987; Cory *et al.*, 1987; Brown, *et al.*, 1998). High temperatures and optimum soil moisture due to the frequent irrigation are the main factors favoring fast mineralization of the organic matter in the sludge and release of metals into soil. We assume that high cation exchange capacity and high pH of this soil were the main causes of reducing the metal extractability with time (Chaney and Ryan, 1993).

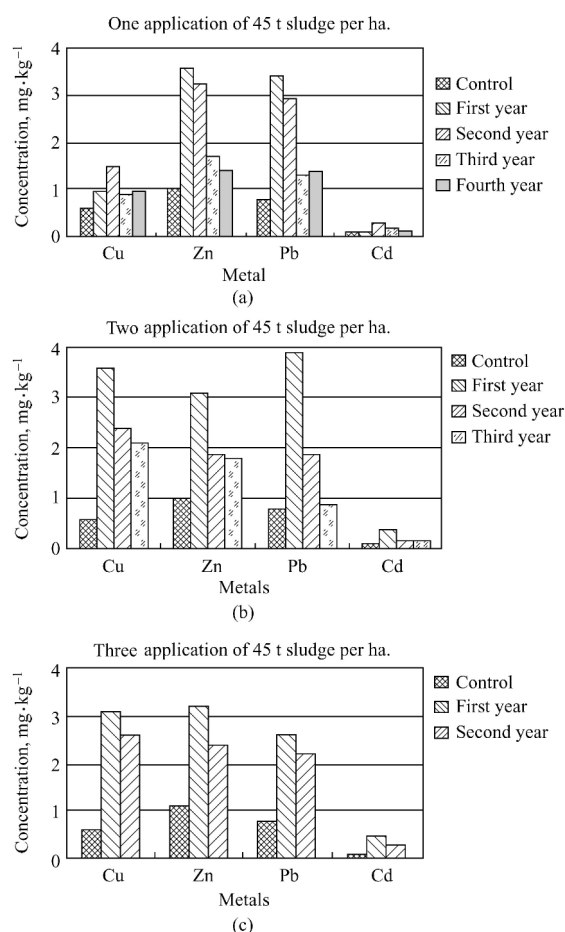


Fig.2 EDTA extractable metals in soil after application of 22.5 t/ha sludge in (a)1996, (b) 1996 and 1997, and (c) 1996, 1997, and 1998

3.2 Plant tissue concentrations

Plant concentrations of the heavy metals increased significantly with sludge applications except for Cd (Fig.3 and 4). The increase in concentrations was more pronounced with higher sludge application dosage (Fig.3b and 4b). In both lettuce and spinach the concentrations decreased in the years after the last sludge application. In general, following the same trend as we saw for EDTA extractable metals in the soil. As we expected the rate of decrease appeared to be slower for the 45 t · ha⁻¹ sludge application load. However, after three years, the metal concentrations generally were still significantly higher than the control levels. Logan *et al.* (1997) found that heavy metal concentrations in corn and lettuce decreased within 2 years after sludge application. Similar results were reported by Juste and Mench (1992) with regard to Cd concentration in corn that decreased to background. The review by Chang *et al.* (1987)

suggests that no significant changes in plant concentrations are likely to occur after the first 2 to 3 years of sludge application.

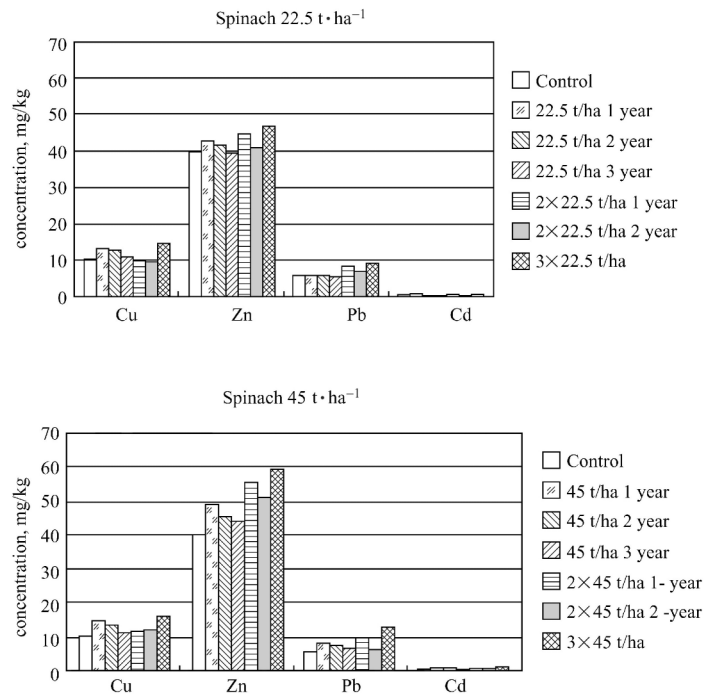


Fig.3 Metal concentration in spinach after application of sludge in different years

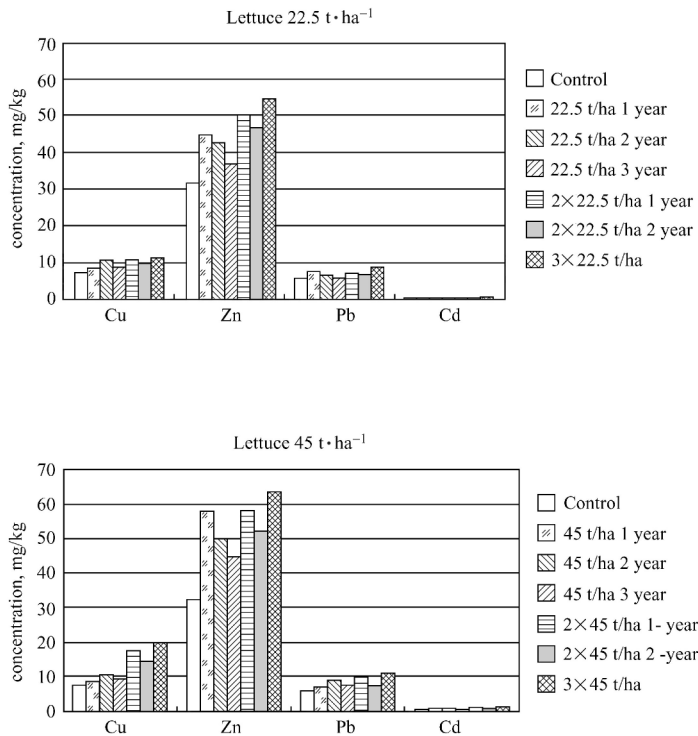


Fig.4 Metal concentration in lettuce after application of sludge in different years

Similar to the increases in EDTA extractable soil metal concentrations, application of sludge in two consecutive years also increased the metal concentration in both plants significantly more than only single application. Again the plant metal concentrations decreased with time (Fig.3,4). Cadmium concentrations in the plant showed some increasing trend only after application of $45 \text{ t} \cdot \text{ha}^{-1}$ sludge two years in row. However, the Cd concentration were much lower than the phytotoxic levels of $5 \text{ mg} \cdot \text{kg}^{-1}$ for sensitive crops (Kirchmann, 1994).

Metal concentrations in lettuce and spinach roots were not significantly different from shoot concentrations for any of the metals therefore, the results are not presented.

Metal concentrations in lettuce and spinach were not significantly different. The highest Cu, Zn, Pb, and Cd concentrations were found in plants grown on plots that had received two $45 \text{ t} \cdot \text{ha}^{-1}$ applications in consecutive years. However, the concentration of all the metals were below phytotoxic concentrations of 20, 200, 10, and $10 \text{ mg} \cdot \text{kg}^{-1}$ for Cu, Zn, Pb, and Cd respectively (Kabata and Pendias, 1992).

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