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SCIENCE DIRECT
Science of the Total Environment 363 (2006) 11–21
Science of the Total Environment
An International Journal for Scientific Research into the Environment and its Relationship with Humankind
www.elsevier.com/locate/scitotenv

Leaching of heavy metals (Cu, Ni and Zn) and organic matter after sewage sludge application to Mediterranean forest soils

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Received 25 January 2005; received in revised form 30 September 2005; accepted 4 October 2005
Available online 28 November 2005

Abstract

In Mediterranean dry and semiarid areas, soil organic matter is often depleted due to ancient and intensive human activity. Under these conditions the use of sewage sludge as a land reclamation technique may be a means to revert desertification processes and to enhance soil function and nutrient cycling. However, applications of heavy metal-contaminated sewage sludges can significantly increase potentially toxic metal concentrations in soils and metal transfer to freshwater and plants. The aims of this study are 1) to investigate the leaching of Cu, Zn and Ni from three contrasted Mediterranean forest soils (a basic loam, a basic clay, and an acid loam) treated with sewage sludges and 2) to explore the relationships between metal mobility and soil properties and with the leaching of organic matter. The selected soils were incubated in columns (5 replicates × 3 soils × 3 treatments). Treatments were (a) soil application of low metal content sewage sludge (LMS), (b) soil application of metal-enriched sewage sludge (MES), and (c) control. The sewage sludge application represented a dose of 6 kg dry weight m^{-2}. Soil columns were incubated at room temperature for 110 days and were irrigated weekly with deionised water to make a total of a 1130 mm. Leachates were collected and analysed for pH, EC, organic carbon Cu, Ni and Zn concentrations.

The concentration of metals and organic matter in the leachates depended on the soil characteristics and on the type of sewage sludge added to the soil. Basic soils with a high amount of clay showed the highest metal retention capacity, while acid soils with low clay content showed the lowest. Of the three metals studied, Ni exhibited the greatest mobility. Zn mobility was also rather high, particularly in the acid soil. Despite the fact that basic soils showed greater OM content than the acid soil, organic carbon in leachates after sludge addition was of the same order of magnitude in all studied soils. OM mobility may enhance the leaching of metals while the OM bound to soil particles may enhance the retention of metals. The rate of leaching per unit of metal in the soil, for Cu in all soils and for Zn in the basic soils, did not increase even in soils amended with metal-enriched sewage sludge. However, the total amount of Cu, Zn, and Ni leached through the 30 cm columns of the metal-enriched sewage sludge increased in comparison to the control soils. In spite of this fact, metal concentrations were in most cases well below limit for drinking water quality criteria of Spanish legislation. Thus, after one single addition of metal-enriched sewage sludge, for the studied Mediterranean soils, the risk of heavy metal pollution of groundwater appears to be low.

Keywords: Forest soil; Sewage sludge; Leaching; Organic carbon; Heavy metals; Copper; Nickel; Zinc

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doi:10.1016/j.scitotenv.2005.10.004

1. Introduction

The forest lands of the Mediterranean region have suffered from the impact of human activity for centuries. In this region, particularly in semiarid areas, many soils are badly degraded as a result of repeated forest fires, overgrazing, and inappropriate agricultural practices (López Bermudez and Albadalejo, 1990). On the other hand, the widespread abandonment of agricultural lands provides the possibility of land reclamation for soil protection and other forest purposes. Indeed, currently, in degraded areas, reforestation programmes are being implemented in an attempt to mitigate land degradation.

In dry and semiarid conditions Mediterranean forest soils often show low levels of organic matter at the soil surface that in some cases may favour the onset of land degradation processes. In degraded areas the use of high organic matter content residues may be considered as a means to enhance nutrient cycling and the general functioning of soils and may also help the establishment of new seedlings.

Sewage sludge contains organic matter and nutrients that have the potential to enhance forest productivity and several soil characteristics (Henry and Cole, 1997; Mosquera-Losada et al., 2001). However, sewage sludge addition always poses a risk to the environment resulting from nutrient imbalances and toxic element accumulation and leaching. Metal transfer from sewage sludges to soil and subsequently to groundwater and plants represents potential health and environmental risks (McBride et al., 1997; Bhogal et al., 2003). Heavy metals are often highly persistent in soil, with residence times as long as thousands of years (Alloway, 1990). Metals applied with sewage sludge may be retained in the soil as a result of their adsorption on hydrous oxides, clays, and organic matter; the formation of insoluble salts; or the presence of residual sewage sludge particles (Alloway and Jackson, 1991). Moreover, soil $CaCO_3$ has often been found to increase soil metal retention (Raikhy and Takkar, 1983); thus, in basic soils metal persistency is expected to be generally very high. A number of studies on acid soils have examined the leaching of heavy metals in sewage sludge amended-soil columns. While some studies have shown that the migration of heavy metals is negligible (Schiradoe et al., 1986; Dowdy et al., 1991; Camobreco et al., 1996), other studies have demonstrated that significant amounts of Zn, Cr, Cu and Cd (among others) were readily leached (Frenkel et al., 1997; Parakash et al., 1997; Karathanasis et al., 2005). It therefore appears that soil properties such as organic matter and soil pH may have a major effect on metal mobility. The addition of organic matter may also affect metal mobility, for instance, by increasing soluble organic matter andCu mobility may be enhanced, especially in sandy soils of high pH (McBride et al., 1999). Mediterranean forest soils often show relatively high pH and $CaCO_3$ content and a medium-to-low organic matter content at the soil surface. The addition of sewage sludge to these soils represents an increase of organic matter and metals that, combined with the soil characteristics, may show differential responses depending on the metal and on the studied soil. Thus, the aims of this study were to investigate the leaching of Cu, Ni, and Zn in three contrasted Mediterranean forest soils after the addition of sewage sludges, and to explore its relationships with soil properties and with the leaching of organic matter.

2. Materials and methods

2.1. Experimental design

Three contrasted Mediterranean forest soils were sampled in Eastern Spain to represent the typical soils occurring on the three major parent materials of the area. Soils were a Calcaric Regosol derived directly from marls and coluvions (basic loam), a Rendzic Leptosol derived from limestone (basic clay) and an Eutric Cambisol derived from loamy red sandstone (acid loam) (FAO-world reference base for soil resources, 1998). The top 30 cm layer of each soil was collected from three sites in the Region of Valencia

Table 1
Physical and chemical properties of the soils before sewage sludge application

Soil (parent material)	Sand	Silt	Clay	$CaCO_3$[a]	CEC	pH	OC[b]	Cu	Ni	Zn
	% (w/w)	% (w/w)	% (w/w)	% (w/w)	cmol kg^{-1}	H_2O	% (w/w)	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}
Basic clay (limestone)	23	34	43	15	10	8.4	2.4	12	29	52
Acid-loam (sandstone)	43	40	17	3.9	3.9	5.9	0.4	7	16	10
Basic-loam (marls)	40	40	20	51	3.8	8.4	2.8	1.6	6.3	13

[a] Active $CaCO_3$.
[b] Organic carbon.