Monitoring heavy metal concentrations in leachates of an acid forest soil during repeated applications of sewage sludge

G. Egiarte[1], M. Pinto[1], E. Ruíz-Romera[2], M. Camps Arbestain[1]*
[1]NEIKER, Berreaga, 1, 48160-Derio, Bizkaia, Basque Country, Spain.
[2]Departamento de Química e Ingeniería Ambiental, Escuela de Ingenieros, Alameda Urquijo, s/n. 48013-Bilbo, UPV, Basque Country, Spain.
**Corresponding author (gegiarte@neiker.net)*

Introduction

Metal transfer from sludge to soil and subsequently to groundwater represents one of the most critical long-term hazards related to the application of these wastes to soils. Some studies working with soils with a pH ranging from 4.5 to 8.1 held that this was practically insignificant as metals remained at the site of input (Dowdy et al., 1991; Sukkariyah et al., 2005). However, several field trials and column studies using soils with a pH ranging between 3.7 and 8.4 (Egiarte et al., 2006; Toribio and Romanyà, 2006) have evidenced the migration of metals with depth, highlighting the concern of groundwater contamination with these pollutants. Here in this study, the mobility of sewage sludge-derived heavy metals (Zn, Cd, Pb, Cr and Ni) in acid sandy soils under pine stands was monitored during 2.5 years after two consecutive applications of this waste at three different loading rates. Only those of Zn, Pb and Ni will be discussed here.

Materials and methods

The soil is a Dystric Cambisol (FAO-ISRIC-ISSS, 1998) developed from a mixture of marlstones and sandstones, in which the second dominates. The experimental design is a completely randomized design. Twelve (8.1 x 8.6 m) plots were established. Sludge was applied to 9 of the 12 plots at three different loading rates (2.4, 17 and 60 Mg ha^{-1}, DW sludge equivalent); there were three replicates per treatment and the remaining untreated plots were considered as controls. The soil has a pH ranging from 4.1 to 4.4, and an ECEC lower than 10 cmol(+) kg^{-1} below a depth of 4 cm. Soil texture was sandy clay for the A1 and Bw horizon, and sandy clay loam for the A2 horizon. The concentrations of the native Zn and Pb in the soil were higher in the A1 horizon (48.5 and 61.0 mg kg^{-1}, respectively) than in the deeper horizons. For Ni the weighted mean concentration for the whole soil was 6.5 mg kg^{-1}, respectively. Concentrations of Zn, Pb and Ni, in the municipal sludge used in 2001 were 8224, 96 and 169 mg kg^{-1}, respectively. The corresponding concentrations in the municipal sludge applied in 2002 were 8488, 155, and 167 mg kg^{-1}, respectively. Leachates were sampled at 25 and 50 cm depth, using Prenart Super Quartz soil water samplers 24-h after a significant rainy episode. Concentrations of Pb and Ni were measured with an atomic absorption spectrophotometry with GTA 100 graphite chamber, and those of Zn with an ICP-OES.

Results and discussion

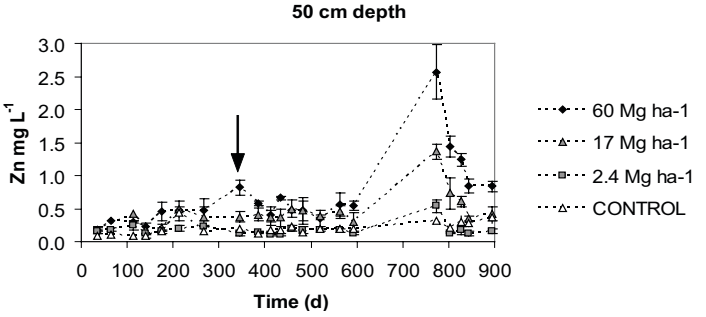
Zinc concentrations in the leachates

The concentrations of Zn in the leachates were the highest of all heavy metals considered in this study, with values ranging from 0.1 to 4.1 mg L^{-1} (at 25 cm depth data not shown) (Fig. 1). Overall, the general trend observed was that repeated applications of sludge at high doses resulted in high Zn concentrations in solution, whereas in the lowest dose (2.4 Mg ha^{-1}) treatment and the control, Zn concentrations remained between 0.1 and 0.2 mg L^{-1} for almost all sampling dates. Moreover, it should be noted that the concentrations of Zn in the leachates at 25 cm depth in the medium and high dose treatments (data not

shown) were higher than those corresponding to 50 cm depth (significant at $P < 0.05$ for the 60 Mg ha^{-1} treatment), which was mainly attributed to retention of Zn by the exchanges sites as water percolated through the profile.

Maximum values of Zn lost at 50 cm depth were obtained for the 60 Mg ha^{-1} treatment, represented the 0.7 % of the total Zn applied to these plots. The minimum amount of Zn lost corresponded to the 2.4 Mg ha^{-1} treatment, represented the 4.3 % of the total Zn applied to these plots. It should also be noted that the mean amount of Zn lost at 50 cm depth in the control plots was higher than the corresponding amount for the 2.4 Mg ha^{-1} treatment, indicating the existence of subsurface lateral flow contaminating the groundwater in the non amended plots.

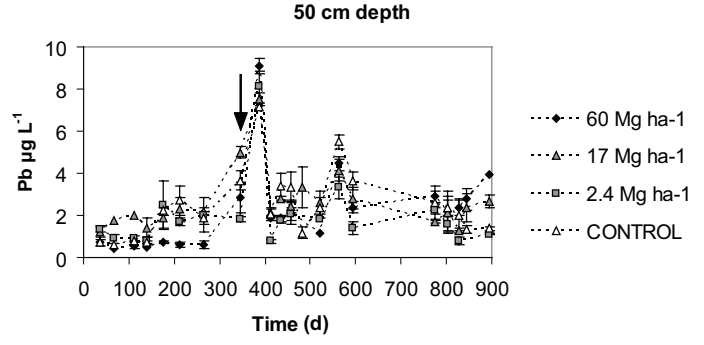
Fig.1. Mean concentrations of Zn at 50 cm depth. Bars represent standard errors of the means (n = 3) The arrow indicates the time of application of the second sludge



Lead concentrations in the leachates

Concentrations of Pb in the leachates were very low for all treatments and control plots, with values ranging between 0.3 and 9.1 $\mu g\ L^{-1}$ (Fig. 2).

Fig. 2. Mean concentrations of Pb at 50 cm depth. Bars represent standard errors of the means (n = 3) The arrow indicates the time of application of the second sludge



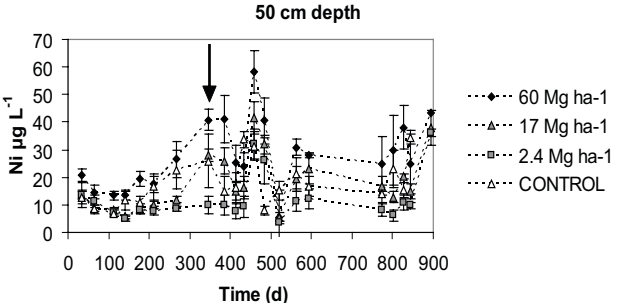
The lack of significant differences ($P < 0.05$) in concentrations of Pb for the different treatments was common throughout the experiment and thus the dose of sludge had no effect on Pb leachability. The results obtained were as expected and consistent with previous results (Gavalda et al., 2005), given the low solubility of this element. The lack of significant differences ($P < 0.05$) between the two depths studied suggests that Pb was not retained by exchange sites, probably owing to the presence of other dominant cations that counterbalanced the charges of these surfaces.

On the other hand, it should be noted that the concentrations of Pb in the leachates at 50 cm depth throughout the experiment were well below the limit for drinking water quality established by the Spanish legislation (RD 140/2003), of 25 μg L^{-1}. The amounts of Pb leached at 50 cm depth represented the 0.2 %, 0.7 %, and 2.6 % of the total Pb added to the soil with 60, 17, and 2.4 Mg ha^{-1} doses of sludge, respectively. Thus, the general trend was that only very small fractions of the Pb incorporated to the soil by the addition of sludge were lost as water percolated through the soil profile.

Nickel concentrations in the leachates

Concentrations of Ni in the leachates corresponding to all treatments and throughout the experiment ranged between 3.4 and 58.3 μg L^{-1} (Fig. 3). High losses of Ni through the soil profile encountered in the plots treated with the highest dose of sludge were expected, given the low retention of this element at exchange sites and the high mobility of this element in soils. Nonetheless, the higher concentrations of Ni in the leachates from the 60 Mg ha^{-1} plots collected at 25 cm depth (data not shown) compared with those collected at 50 cm depth (Fig. 5B) suggest the existence of sorption reactions as water percolates through the profile.

Fig. 3. Mean concentrations of Pb at 50 cm depth. Bars represent standard errors of the means (n = 3). The arrow indicates the time of application of the second sludge.



Conclusions

The dose of sludge was important for Zn and Ni, in that repeated applications of sludge at a rate of 60 Mg ha^{-1} resulted in significantly ($P < 0.05$) higher concentrations of these elements in the leachates than observed for the rest of treatments. Concentrations of Pb were always very low, in accordance with the low solubility of this element.

References

Dowdy, R.H., Latterell, J.J., Hinesly, T.D., Grossman, R.B. and Sullivan, D.L., 1991. Trace metal movement in an aeric ochraqualf following 14 years of annual sludge applications. J. Environ. Qual. 20, 119-123.

Egiarte, G., Camps Arbestain, M., Ruíz-Romera, E., and Pinto, M., 2006. Study of the chemistry of an acid soil column and of the corresponding leachates after the addition of an anaerobic municipal sludge. Chemosphere 65, 2456-2467.

FAO-ISRIC-ISSS, 1998. World Reference Base for Soil Resources. http://www.fao.org/docrep/W8594E/W8594E00.htm

Gavalda, D., Scheiner, J.D., Revel, J.C., Merlina, G.,Kaemmerer, M., Pinelli, E. and Guiresse, M., 2005. Agronomic and environmental impacts of a single application of heat-dried sludge on an Alfisol. Sci Total Environ 343, 97-109.

R.D. 140/2003 por el que se establecen los criterios sanitarios de la calidad del agua de consumo humano. B.O.E. no. 45 de 21/2/03.

Sukkariyah, B.F., Evanylo, G., Zelazny, L., and Chaney, R.L., 2005. Recovery and distribution of biosolidsderived trace metals in a clay loam soil. J. Environ. Qual. 34, 1843-1850.

Toribio, M. and Romanyà, J., 2006. Leaching of heavy metals (Cu, Ni, and Zn) and organic matter after sewage sludge application to Mediterranean forest soils. Sci Total Environ. 363, 11-21.