The influence of biosolids treatment files on the mobility of metal trace elements

V. Maisonnave*, M. Montrejaud-Vignoles*, C. Bonnin, J.-C. Revel*** and C. Vignoles******

* Ecole Nationale Supérieure de Chimie de Toulouse, 118 route de Narbonne, 31077 Toulouse Cedex, France

** Anjou Recherche, Chemin de la Digue, BP 76, 78603 Maisons-Laffite Cedex, France

*** Ecole Nationale Supérieure d'Agronomie de Toulouse, Chemin de l'Agrobiopole, BP 107, 31326 Auzeville Tolosane, France

**** Générale des Eaux, 78 chemin des Sept Deniers, 31200 Toulouse Cedex, France

Abstract The production of sludge in France is estimated to be about 900,000 metric tons dry matter per year and 60% of this is recycled onto agricultural land. At present, the long term future of this procedure is open to question and among the different arguments being put forward are the levels of metallic trace elements and the risk of accumulation in soils.

This study presents the behaviour of metallic trace elements in sludges from three different treatment procedures: thickened liquid sludges, dewatered sludges and dried sludges. These biosolids are mixed with a clay soil and then placed in a temperature and humidity controlled glasshouse. Several containers are seeded with ryegrass and compared with controls. For the three harvests, covering all the amendments studied (including non-amended soil), the differences are not really representative. Absorption by the ryegrass is low in all cases. For the cadmium, the chromium, the nickel and the lead, the roots are 5 to 10 times more concentrated than the leaves. The majority of these elements stay absorbed in the roots, regardless of the amendment used.

The addition of the sludges has considerably reduced the uptake of water in ryegrass throughout its growth cycle. Quite apart from their fertilizing qualities, wastewater treatment plant sludges could offer important implications for irrigation.

Keywords Biosolids; fertilizers; metal trace elements; ryegrass; soil; transfers

Introduction

In France, the annual production of sludge from wastewater treatment plants is 900,000 tonnes dry matter and 60% of this is agriculturally recycled as a complement to farmers' manuring programs. This is because their organic matter, nitrogen and phosphorus content makes them good fertilizers as long as they do not also contain too many toxins such as trace elements. However, land application of wastewater treatment plant sludges causes concern and French regulations have been reinforced (decree of 8/12/97 and Statutory order of 8/1/98) halving maximum permitted levels of trace elements and dividing by four those for cadmium (10 mg/kg DM in 2004).

The object of the present study is to look at the transfer of metallic trace elements from wastewater treatment plant sludges as well as fertilizers, to the soil and plants in order to estimate the real risk. The study is based on an analysis of the behaviour of metallic trace elements when they are in soil-amendment mixtures in containers of vegetation, and of their bio-availability *vis à vis* the plants. A non-amended control has also been studied (Lindsay 1972, Jarvis *et al*. 1976, Mahler *et al*. 1978, Jarvis and Jones 1978,Cataldo *et al*. 1981).

In order to do this, vegetation containers have been set up using wastewater treatment plant sludges originating from three different treatment methods: thickened liquid sludges, dewatered sludges and dried sludges, mixed with soil and then sown with ryegrass. The

Water Science and Technology **Vol 44 No 2–3 pp 381–387** © 2001 IWA Publishing and the authors

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levels of metallic trace elements in the soil-sludge mixtures and in the plants and their roots, as well as the their growth, are compared to those of control containers with no added sludge or fertilizer.

Methodology

Sampling

A 150 kg sample of clay and humiferous soil, pH 6, was taken from plots of land in Poucharramet, France.

The sludges chosen for this experiment come from the Toulouse (France) wastewater treatment plant (550 000 EI). Samples are taken from three points on the treatment pathway.

- The thickened liquid sludge, 8%DM, is taken from the digester outlet: 10 two-litre samples are taken over 1 hour and these are then mixed and homogenized to give a representative sample.
- The dewatered sludge paste, 25%DM, is taken from the centrifuge outlet: Ten 1kg samples are taken over 1 hour and these are then mixed and homogenized. A quartering method is then used to obtain a representative sample of about 2 kg.
- The dried sludge granules, 90%DM, are taken from the heat drier outlet: Ten 500g samples are taken over 1 hour and these are then mixed and homogenized. A quartering method is then used to obtain a representative sample of about 1 kg.

These samples are then distributed among the vegetation containers.

Apart from the above samples, a lawn fertilizer characterized by agronomic values for N, P, and K of 20, 8 and 5 respectively, and a nutrient solution supplying the plant with the necessary mineral elements without recourse to absorption from the soil, have been chosen to amend the control pots.

Setting-up the vegetation containers

Soil and amendments mixtures. The French ministerial order of 8 January 1998 fixes the maximum quantity authorized for land application of sludges at 30t DM/ha/10 yr, and it is this dose that has been chosen, added all at the same time, to make the mixtures of soil and the wastewater treatment plant sludges. For the dose of fertilizer, agricultural practice is 500 kg/ha/yr and so this was used for making the soil-lawn fertilizer mixtures. These amendments were repeated after the second ryegrass harvest.

Eight pots were made for each amendment: 4 pots were sown with ryegrass to study the plant's growth, and 4 pots had no seeds and were used to study sludge mineralization. Table 1 shows all the vegetation containers set up.

In order to mimic the real growing conditions, the containers filled with the soil-amendment mixtures have been placed in a phytotron (a hermetically sealed enclosure with regulated temperature and humidity) for 15 days with nothing else being added, in an effort to recreate the maturation period of a field in winter.

The water contained in the sludges and transferred to the mixtures evaporates

Table 1 Cultivation pots in phytotron

	Control pots	Mixture pots	With rye-grass	Without rye-grass
Soil only	8 pots		4 pots	4 pots
Soil + nutritive solution	4 pots		4 pots	
Soil + fertlizer	8 pots		4 pots	4 pots
Soil + liquid sludge		8 pots	4 pots	4 pots
Soil + dewatered sludge		8 pots	4 pots	4 pots
Soil + dried sludge		8 pots	4 pots	4 pots

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completely. At the end of this maturation, all the containers are about the same weight and represent the initial growth medium. They are then emptied, the lump broken up and homogenized to simulate ploughing in the spring. Then the soil-amendment mixtures are put back into the containers and replaced in the phytotron: they are now ready for sowing.

Sowing. The plant used is the tetraploid variety "Exalta" of Italian ryegrass (*Lolium multiflorum*), chosen for its rapid and abundant growth and for its capacity to absorb metallic trace elements easily. In addition, a large number of individual plants can be grown (about 500) giving representative samples. In order to produce sufficient quantities of plant matter for subsequent analyses, the sowing density for the containers has been set at 630 kg per hectare, which is about 500 seeds per container. The latter are watered to obtain optimal soil humidity conditions for maximum plant growth. Watering is carried out every other day in the germination phase and then every day just before harvesting, always to two thirds the field capacity.

Physico-chemical analyses

The amendments were dried for 24 h at 105°C and then ground up using a mechanical, agate pestle and mortar. The metallic trace elements contained in the two wastewater treatment plant sludges and the lawn fertilizer were put into solution using aqua regia and microwave mineralization in accordance with the norm Standard Methods 3030 F, K.

Sampling of the soil and soil-amendment mixtures was carried out before the first sowing, after the second harvest, before the second sowing and after harvesting of the second amendment. Figure 1 gives a visual presentation of the different sampling stages, which were made by taking core samples in each container and then mixing four of these samples from the same type of container. The metallic trace elements contained in the soil-amendment mixtures were put into solution by fusion at 1050°C and acid recovery of the calcinated residue in accordance with the Standard Methods norm 3030 J.

The plants were harvested 1 and then 2 months after the initial sowing, and a third harvest was made 1 month after the second sowing. The roots were pulled up at the end of the experiments. Figure 1 gives a visual presentation of the different harvests.

These 3 ryegrass harvests, plus the roots, were dried for 24 h at 105°C and then ground up using a mechanical, agate pestle and mortar. Next they were calcinated for 16h at selected temperatures, maximum 500°C. The resultant ash was then acid recovered (nitric acid). All the solutions thus obtained were then assessed using Induced Coupled Plasma Mass Spectrophotometry (SM 3120B) to obtain values for Cd, Cr, Cu, Ni, Pb, Zn.

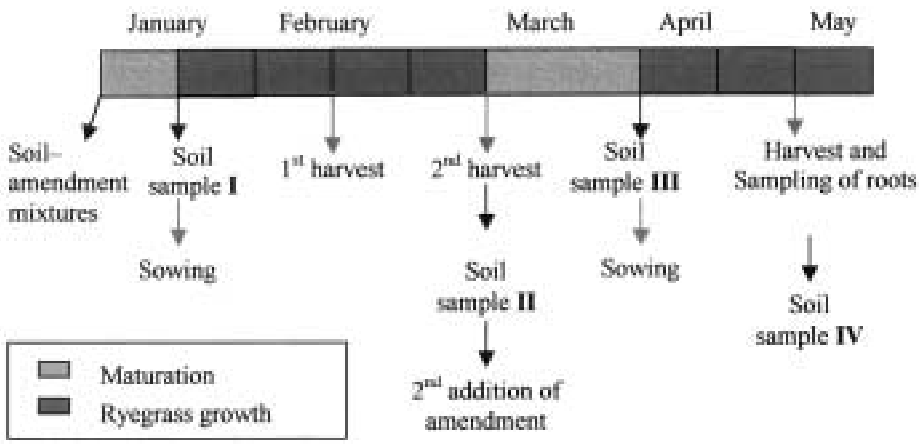

Figure 1 Sampling of soil-amendment mixtures and ryegrass harvests

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Results

Amounts of metallic trace elements in sludges and fertilizers

Figure 2 shows the levels of Cd, Cr, Cu, Ni, Pb and Zn in the 3 Toulouse biosolids and in the lawn fertilizer.

Distribution of metallic trace element levels in the soil-amendment mixtures

Figure 3 shows the levels of cadmium, chromium, copper, nickel, lead and zinc in the soil and mixture samples taken at the beginning of the experiment.

Levels of metallic trace elements in the plants and the roots

Figure 4 (a–e) shows the levels of cadmium, chromium, copper, nickel, lead and zinc measured for the three ryegrass harvests and roots which were grown on a soil amended or otherwise with sludges and fertilizers. The levels of lead and chromium are similar as is their evolution. All values are in mg/kg DM.

Water efficiency

Figure 5 shows the water efficiency over the three harvests. This data allows a comparison to be made between the water requirements of growing ryegrass relative to the different amendments. They correspond to the mass of water necessary for the plant to produce 1 g of dry matter.

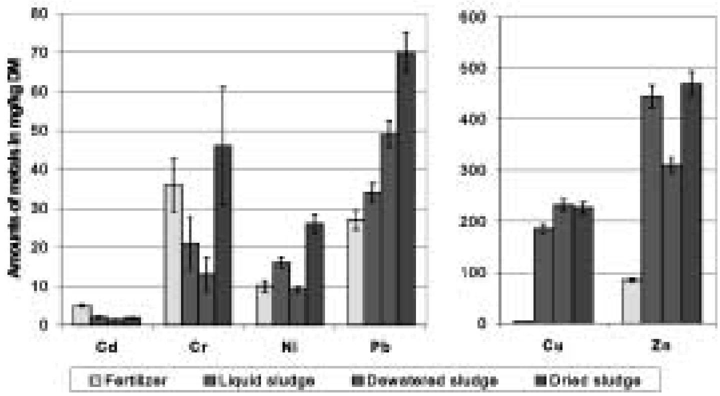

Figure 2 Amounts of metal trace elements in the amendments

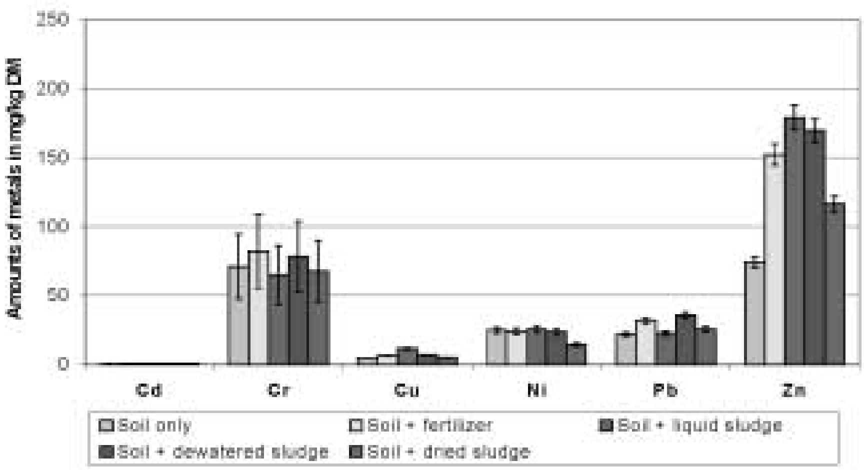

Figure 3 Amounts of metal trace elements in the amendments-soil mixtures

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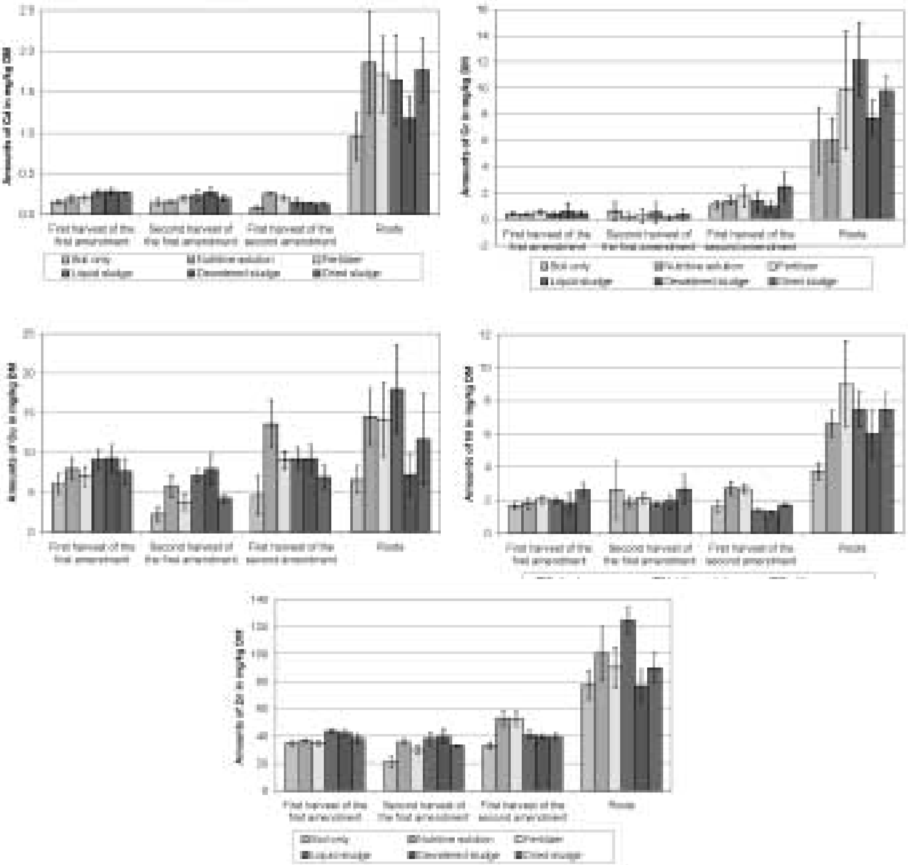

Figure 4 Amounts of (a) Cd, (b) Cr, (c) Cu, (d) Ni and (e) Zn in the 3 harvests and in the roots

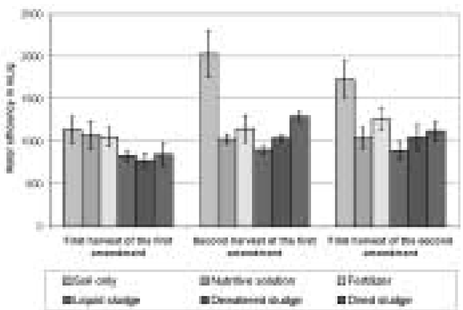

Figure 5 Water efficiency over the three harvests

Discussion

Metallic trace element concentrations in the soil and the mixtures

The addition of sludge when making the mixtures with the Poucharramet soil has been carried out in accordance with French regulations: 30 metric tons DM/ha over 10 years, which corresponds to about 20 g of dried sludge for 2 kg of soil. This is a relatively large dilution, i.e about 100 times. Because of this, the levels of metallic trace elements such as Cd, Cr, Cu, Ni, and Pb found in the soil-sludge mixtures, differ very little from those found in non-amended soil and the same is true for the fertilizer. For copper, there is a small increase with liquid sludges and this difference is more pronounced for zinc: the addition of

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sludge and likewise fertilizer, produces a distinct increase in zinc in the soil. As the soil is poor in these oligo-elements at the outset, this result is of interest.

No major differences have been observed between the three sludges except for the zinc where transfer depends upon the amount of dry matter in the sludge.

Transfer of trace elements to the plant

The cadmium is not absorbed by the above-ground parts of the ryegrass, it is fixed only in the roots. There are no significant differences relative to the sludge treatment method and the behaviour of the metallic trace elements is similar in the sludges, the fertilizer and the non-amended soils.

The absorption of chromium is low and identical for the three harvests, regardless of the fertilizer or sludge under consideration. Liquid sludge provides the most chromium to the roots where it peaks, and therefore this element is not bio-available to the whole plant but only the roots. After the second amendment, the chromium concentrations in the plant increase, but in a similar fashion on non-amended soil and soils amended with sludges or fertilizers. Chromium is therefore sparingly bio-available and only in the long term.

The amount of copper added to the soil by the sludges is greater than that added by fertilizers; however the amounts absorbed by the plants are greater with the fertilizers. The form of copper contained in the fertilizers is therefore more bio-available than that found in the different types of sludge studied.

Nickel is mainly absorbed by the roots for all the sludges and fertilizers studied, although more so with the fertilizers.

The lead is almost unavailable to the above ground parts of the ryegrass. It is found for the most part in the roots.There are no significant differences between the types of sludge for this element.

Zinc is absorbed regularly over the three harvests and in a similar fashion for the various amendments. At the end of the experiment, the roots have the highest levels. The soil-dried sludge mixture from Toulouse seems to behave like a non-amended soil medium.

In all cases and regardless of the metal studied or the type of sludge, transfer of metals to the above ground parts of the plant and to the roots are similar for sludges and fertilizer.

Water efficiency

The water efficiency distribution is inversely proportional to the production of plant matter: the least productive containers of growing ryegrass consume the most water. The addition of sludges to a soil permits a reduction in plant water uptake: almost three times more water is needed on a non-amended soil to produce the same amount of plant matter as on a soil-sludge mixture.

Conclusions

This study has enabled observations to be made concerning the transfer of metallic trace elements from wastewater treatment plant sludges to the soil and the rye-grass plant, using vegetation pots placed in a phytotron. Because the soil-sludge dilution is relatively high, the amounts of metallic trace elements in the soil-sludge mixtures are little different from those found in untreated soil.

The differences are relatively small for all the amendments studied. Because the soil-sludge mixtures only contain a low concentration of metallic trace elements, absorption by the rye-grass is low in all vegetation pots. Several main conclusions would seem to be suggested by this study. They concern sludges that have been mixed with soil in vegetation pots:

- metallic trace elements in liquid sludge are more bio-available, doubtless because they are more mobile in the liquid phase;

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- metallic trace element pollution is less in dewatered sludge and when mixed with soil it behaves like untreated soil. It is therefore the best for land application especially if it can be made hygienic;
- dried sludge contains more metallic elements at the outset due to the heat treatment which, by destroying the organic structures, concentrates the metals and makes them more bio-available; in addition its agronomic value is lower;
- metallic trace element levels are lower at the outset in the fertilizers but higher afterwards in the plants: their bio-availability is therefore greater.

The addition of sludges to a soil allows a reduction in water consumption for ryegrass: quite apart from their fertilizing qualities, wastewater treatment plant sludges can offer interesting perspectives as regards irrigation and economize large quantities of water.

In the conditions of the experiment (vegetation pots, application doses of 30t DM of sludge and rye grass cultivation), transfer of trace elements to the soil is therefore almost non-existent and very low to the leaves of ryegrass. Only the roots absorb larger quantities of the metals compared to controls of untreated soil. These metallic trace elements seem to be blocked in the roots and do not travel to the leaves. In conclusion, when the authorized land application doses are respected, the accumulation of metallic trace elements in the soils or plants is not significant.

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