Chemosphere 65 (2006) 2456–2467

CHEMOSPHERE

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Study of the chemistry of an acid soil column and of the corresponding leachates after the addition of an anaerobic municipal sludge

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Received 23 September 2005; received in revised form 13 April 2006; accepted 15 April 2006
Available online 12 June 2006

Abstract

A column leaching study was carried out over a period of 77 days to determine the changes in the chemistry of an acid soil and of the corresponding leachates after the addition of an anaerobic sludge (equivalent to 69 Mg DW ha^{-1}). By the end of the experiment, the addition of the sludge to the soil had induced an increase in soil pH (from pH 3.6–4.0 to pH 4.1–4.8), in spite of the pronounced decrease in pH detected in the leachates by day 18 of the experiment. The decrease in pH (down to pH 3.3) occurred at the same time as leachate SO_4 and Fe peaked. Once the acidification attributed to sulphide oxidation ceased, the "liming effect" of the sludge became evident and counteracted further proton production – such as that associated with oxidation of NH_4 – at least for the duration of the study. Concentrations of Zn, Cd, Ni, and to a lesser extent, Pb in leachates displayed pulses at the beginning of the experiment (first 12 days), whereas the concentration of Cu followed a more irregular pattern; the concentrations of these metals never surpassed the European threshold values for drinking water. In contrast, concentrations of NO_3, Mn, and Cr in leachates had increased by the end of the experiment – in parallel with an increase in dissolved organic C (DOC) – and surpassed the European threshold for drinking water. Mineralisation of native soil organic C (SOC) was enhanced by the addition of this N-rich residue, and the organic C mass balance at the end of the experiment was negative. Nitrogen mass balance, although positive, exhibited a loss of 77% of the N added to the system. The results obtained indicate that application of this sewage sludge to a soil with a pH < 5, at the loading rate used here, and without liming (i.e., non fulfilment of the requirements of the present European Directive) may pose a risk in terms of groundwater contamination.

Keywords: Sewage sludge; Heavy metals; Leaching; Soil column

1. Introduction

The application of municipal wastewater sludge to agricultural soils is an attractive proposal for the sustainable management of this residue, as it involves recycling of the organic matter and nutrients present in the sludge, and provides a means of its disposal. The use of sludge as a soil amendment is, however, complicated by the low, but not insignificant, amounts of pollutants present in the sludge, with most concern to date being associated with the presence of heavy metals. The risks that these elements pose to ground water quality and ecosystem health depend on their availability and mobility through the soil profile. In turn, the availability and mobility of heavy metals depend on (i) chemical properties such as soil pH and redox potential, which affect chemical speciation and solubility (Lindsay, 1979), (ii) solute–solute interactions such as metal complexation with either organic or inorganic species (Vulkan et al., 2002), (iii) soil surface characteristics and metal–soil interactions, which affect sorption reactions (Sparks, 2005), and (iv) soil physical properties that affect water movement (Camobreco et al., 1996). Metal complexation reactions

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0045-6535/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.chemosphere.2006.04.044

and metal–soil interactions are, in turn, affected by pH and redox conditions, further affecting metal solubility (McLean and Bledsoe, 1992).

Ever-increasing amounts of sludges will probably be applied to agricultural land in the near future and, therefore, their application to soils must remain environmentally sustainable, especially in acid soils, where heavy metals are likely to be more mobile. In fact, acidic conditions often enhance the solubility of heavy metals in soils, which may be further acidified when a potential source of protons, such as an anaerobic sludge, is added to them. When treated under anaerobiosis, sludges generally reach methanogenic conditions, and under such conditions, sulphur is present in the form of metal sulphides. Oxidation of metal sulphides is a very acidic reaction (van Breemen, 1973), thus, addition of the residues to agricultural soils, which are generally well-aerated, may greatly affect the overall chemistry of the system.

The addition of organic residues to land usually contributes to an increase in the organic C content of soils, which is important in terms of C sequestration and as a means of recovering the depleted organic matter content of agricultural soils. However, rapid C mineralization without a net increase in SOC content has been reported to be associated with the use of anaerobic sludges (Adani and Tambone, 2005; Egiarte et al., 2005). Kirchmann and Bernal (1997) ranked the C stabilization efficiencies of the various waste treatments, measured in terms of decomposition of waste C, as follows: aerobically-treated and composted > non-decomposed > anaerobically-treated. The greater decomposition of organic C from anaerobic residues than from the other treated wastes that takes place once the waste is applied to soils, is mainly due to the presence of more short-chain C organic acids in the former, which rapidly decompose under the oxidizing soil conditions (Kirchmann and Lundvall, 1993).

Column studies have frequently been used to provide information about element release and transport in soil, chemistry of soil and leachates, and to carry out kinetics and mass balance studies (Camobreco et al., 1996; Grolimund et al., 1996; Temminghoff et al., 1997; Voegelin et al., 2003; Qureshi et al., 2004). They may, therefore constitute an adequate tool for the achievement of the objectives of the present study, which were to examine: (i) the changes that take place in the chemistry of an acid soil and in the leachates after the addition of an anaerobic sludge, (ii) the downward movement, distribution, and mass balance of organic C, N and heavy metals through the soil after the addition of sludge, and (iii) the relationship between the above two goals.

2. Materials and methods

2.1. Soil

The experiment was carried out using a soil with no prior history of sludge application. The soil is a Dystric Cambisol (FAO-ISRIC-ISSS, 1998) developed from a mixture of marlstones and sandstones, in which the former dominates. The horizons used for the column study were the following: A1 (0–4 cm), A2 (4–22 cm), and Bw (22–48 cm), the characteristics of which are reported in Table 1. Soil was sampled by horizons, air-dried at 30 °C until constant weight, then ground and sieved (2 mm mesh), prior to preparing the soil column. The soil has a pH lower than 4, a cation exchange capacity (CEC) lower than 15 cmol(+) kg^{-1} below a depth of 4 cm, and a base saturation (BS) lower than 17% (Table 1), like many soils located in high leaching environments. Organic C content in the A1 horizon was 93.8 g kg^{-1}, whereas that of the A2 horizon was about 4 times lower (23.5 g kg^{-1}). 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, Cu, and Pb in the soil were higher in the A1 horizon (48.5, 13.4, and 61.0 mg kg^{-1}, respectively) than in the deeper horizons (Table 2). For Cd, Ni, and Cr, the effect of metal concentration in the uppermost soil horizon was not observed (Table 2), and the weighted mean concentrations for the whole soil were 5.8, 0.8, and 33.5 mg kg^{-1}, respectively.

2.2. Sewage sludge

We used an anaerobic municipal sewage sludge from the Durango wastewater treatment plant (Bizkaia, Spain). The sludge underwent preliminary treatment by centrifugation before being applied to the soil column. Chemical characteristics of the sludge applied to the plots are shown in Table 3. Organic C in the municipal sludge was 205 g kg^{-1}, total N 42.4 g kg^{-1}, P Olsen 689 mg kg^{-1}, C/N ratio 4.8, and pH 7.7. Concentrations of Zn, Cd, Ni, Cu, Pb, and Cr in the municipal sludge were 10924, 6, 208, 456, 151, and 762 mg kg^{-1}, respectively (Table 3).

2.3. Column study

The PVC column used in the present study was of inner diameter (ID) 10.6 cm and length 60 cm. The column was hand-packed with small amounts of soil and in accordance with the profile horizons. The soil column rested on a 2 cm column of coarse fragments taken from the soil parent material. This column was placed on a nylon net (25 μm mesh), which rested on a Whatman 41 filter to retain the solid material in the column. A support base with several drainage holes of 1.5 mm ID was placed at the bottom of the column. A funnel, filled with 5 mm diameter fiberglass beads, was attached to the base to conduct the leachates, which were collected in 250 ml Erlenmeyer flasks. The municipal sludge of dry weight (DW) 23% was applied on top of the column at a loading rate of 69 Mg ha^{-1} (DW equivalent). Thus, 60.5 g of sludge (DW equivalent) were added to the soil column, and the total amounts of Zn, Cd, Ni, Cu, Pb, and Cr applied to the soil column were 661, 0.3, 13, 28, 9, and 46 mg, respectively. At these doses,