



Viability of organic wastes and biochars as amendments for the remediation of heavy metal-contaminated soils



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HIGHLIGHTS

- The buffering capacity of six organic wastes and two biochars is evaluated.
- The target materials are tested for their capacity of sorbing heavy-metals.
- Sorption isotherms are fitted by the Freundlich and linear models.
- Metal sorption in the target materials was mainly affected by pH and DOC content.
- Two organic wastes and two biochars are proposed for remediation actions.

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ABSTRACT

Composts derived from municipal (MOW and MSW) and domestic wastes (DOM), wastes from the olive oil industry (OWH and OP), green waste (GW), and biochars (BF and BS) were investigated to test their viability for remediating metal-contaminated soils. In addition to common analyses, the characterisation included structural analyses (FTIR and ^{13}C NMR), determination of the acid neutralisation capacity (ANC) and the construction of sorption isotherms for target metals (Pb, Zn, Cd, Ni and Cu). MOW and GW had the highest ANC values (4280 and 7100 meq kg^{-1} , respectively), and MOW, GW, DOM, BF and BS exhibited the highest solid-liquid distribution coefficients (K_d) with maximum values in the 10^4 L kg^{-1} range. Sorption isotherms were fitted using linear and Freundlich models for better comparison of the sorption capacities of the materials. Based on their basic pH, high ANC and high sorption capacity, MOW, GW and biochars are the most promising materials.

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1. Introduction

Soil pollution by heavy metals is a worldwide problem due to their high toxicity, long-term persistence and their subsequent incorporation into the trophic chain, which can produce harmful environmental and health effects. Several remediation techniques have been investigated to reduce the environmental impact of metal-contaminated soils, such as phytoremediation, soil washing, stabilisation, solidification, electroremediation and excavation (Mulligan et al., 2001; Kumpiene et al., 2008). Among these techniques, remediation strategies that provide in situ immobilisation of contaminants due to the addition of materials to the contaminated soils exhibit great potential and can be applied to extended areas. In addition to diluting the concentration of the pollutant, these materials can reduce the mobility of heavy metals due to

various mechanisms, such as adsorption, precipitation or complexation, decreasing the (bio)availability of pollutants in soils. Agricultural, urban and industrial waste products can be used in soil remediation because these materials are typically inexpensive and available in large quantities (Lombi et al., 2002; Kumpiene et al., 2008). In addition to their positive effect on soil remediation, the use of such amendments also represents a satisfactory alternative for the beneficial reuse of wastes.

Organic materials of different origins, such as composts from municipal and food industry wastes and animal manures may be useful for remediation of soils contaminated with heavy metals (Clemente et al., 2007; Nwachukwu and Pulford, 2008). Biochars have also been studied as sorbents for inorganic and organic contaminants in water (Mohan et al., 2014) and some authors have been reported their use as amendments for soil remediation (Cao and Harris, 2010; Ahmad et al., 2014). The potential beneficial effect of these materials is not yet clear. Organic materials can decrease the availability of heavy metals by increasing the soil

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pH and the specific interaction of metals, because the reactive groups in the organic matter may complex and sorb the heavy metals present in the soil (Lee et al., 2004; Karlsson et al., 2007). Biochars may possess large surface areas, high porosities, alkaline pH and the ability to sorb dissolved organic matter (DOM) (Pietikäinen et al., 2000). However, some research results indicate that organic materials can also enhance metal mobility in soils due to a potential decrease in the soil pH, increase in the DOM in the resulting mixtures and the related increase in metal-DOM soluble complexes (Clemente et al., 2007; Khokhotva and Waara, 2010). Therefore, the addition of organic materials to contaminated soils causes either positive or negative effects on metal mobility depending on the characteristics of the material and of the receiving soil (e.g., soil pH, organic matter and clay contents). The use of compost originating from green, catering, paper and municipal solid waste significantly reduced the heavy metal concentration of Cu, Pb and Zn in a soil solution, but this effect was not observed for As (Farrell and Jones, 2010). The effect of other materials (e.g., green waste derived compost, spent mushroom compost, composted sewage sludge, coir compost) on the mobility of Cu, Pb and Zn depends on the amendment and soil combination (Van Herwijnen et al., 2007). Other studies on soil amended with biochar and green waste showed a significant reduction of Cd and Zn in pore water. However, the Cu and As concentrations increased (Beesley et al., 2010). Therefore, it is important to characterise the relevant physicochemical, structural and sorption characteristics of the materials that can play a role in the eventual metal mobility in the amended contaminated soils.

The aim of this study is to evaluate the suitability of eight materials (i.e., green waste, two materials derived from the olive oil industry, domestic compost, two municipal solid waste composts and two biochars) for use as amendments for the remediation of metal contaminated soils. In this context, the pH buffering capacity and metal (Cd, Cu, Ni, Pb and Zn) sorption capacity of the materials, as well as additional structural and physicochemical parameters, were investigated to determine the most suitable materials.

2. Materials and methods

2.1. Materials

Six organic and two biochar materials were examined. They were compost from municipal organic waste (MOW), compost from municipal solid waste (MSW), green waste (GW), compost derived from food leftovers (DOM), olive wet husk (OWH), olive pomace (OP), biochar derived from tree barks (BF), and biochar derived from vine shoots (BS).

More detailed information about the materials, as well as the methods used for their physicochemical and structural characterisation is given in the [Supplementary material](#).

2.2. pH titration test

The buffering capacity of the materials was determined by a pH titration test (CEN/TS, 2006). First, the initial pH of each sample was measured in a 1:100 sample/deionised water ratio. Next, consecutive volumes of 200 μL of 1 M HNO_3 or 1 M NaOH were added, and the suspension was stirred. The resulting pH of the suspension was measured 20 min after each addition of the acid or base. The acid and base additions were repeated until the pH range of 2–12 was covered. This test also allowed for the determination of the acid neutralisation capacity (ANC) of the materials, which is defined as the quantity of acid or base (meq kg^{-1}) required to shift the initial pH of the material to a pH of 4. Therefore, for those materials with an initial pH higher than 4, the ANC permits

prediction of its buffering capacity with respect to external acidic stresses.

2.3. Sorption isotherms

Batch sorption tests were performed in polypropylene centrifuge tubes. The material samples (2 g) were pre-equilibrated with 50 mL of a 0.01 M CaCl_2 solution for 16 h using end-over-end shaking. After a pre-equilibration time, different volumes of the metal solution (i.e., Pb, Zn, Cd, Ni and Cu) were added in order to work with ten initial concentrations within the 0.05 to 10 meq L^{-1} range. Finally, the resulting suspensions were end-over-end shaken for 24 h, as recommended by international procedures and previous works (OECD, 2000; Sastre et al., 2006).

The samples were centrifuged, and the obtained supernatants were filtered through a 0.45 μm filter. Major and trace elements were determined in the solutions using a Perkin Elmer Model OPTIMA 3200RL ICP-OES. The detection limits of the inductively coupled plasma optical emission spectrometer are shown in the [Supplementary material](#).

The metal sorbed concentrations were calculated from the quantification of the metal concentration in the initial solution (C_i) and after equilibrium (C_{eq}) using the following equation:

$$C_{sorb} = \frac{(C_i - C_{eq})V}{m} \quad (1)$$

where C_{sorb} is the sorbed amount of metal (meq kg^{-1}), m is the mass of material (kg) and V is the volume of contact solution (L).

The solid–liquid distribution coefficient, K_d (L kg^{-1}), was calculated for each initial concentration as the ratio between the amount of target metal sorbed onto the material, C_{sorb} (meq kg^{-1}), and the amount of metal at equilibrium in the aqueous phase, C_{eq} (meq L^{-1}), according to the following equation:

$$K_d = \frac{C_{sorb}}{C_{eq}} \quad (2)$$

Metal sorption yields (S) for the materials were calculated as follows:

$$S(\%) = \frac{(C_i - C_{eq})}{C_i} \times 100 \quad (3)$$

2.4. Data treatment and fitting of sorption isotherms

The experimental sorption isotherms were fitted to various sorption models (i.e., Freundlich and linear equations).

The Freundlich model is represented by the following equation:

$$C_{sorb} = K_f C_{eq}^N \quad (4)$$

where the K_f and N parameters are obtained from the C_{sorb} vs. C_{eq} fitting. The K_f parameter describes the partitioning of the solute between the solid phase and the liquid phase, and N is related to the heterogeneity of sorption sites and is indicative of the non-linearity of the sorption isotherm. Values of $N < 1$ indicate a high affinity of the soil for the metal at low concentration, whereas values of $N > 1$, for metals, might be explained as the sorption being inhibited by a competing reaction within the solution (e.g., complexation with a ligand). If $N = 1$, the Freundlich equation approaches a linear model, and K_f becomes equal to the K_d value, which remains constant for the whole concentration range.

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