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Optimizing the combined application of amendments to allow plant growth in a multielement-contaminated soil

A. Sevilla-Perea [a, *], M.C. Romero-Puertas [b], M.D. Mingorance [a, 1]

[a] *Instituto Andaluz de Ciencias de la Tierra (UGR-CSIC), Avda. de las Palmeras 4, 18100 Armilla, Granada, Spain*
[b] *Estación Experimental del Zaidín (CSIC), C/ Profesor Albareda 1, 18008 Granada, Spain*

HIGHLIGHTS

- The system behaviour was successfully modelled by Response Surface Methodology.
- Compost and bottom ash addition to a mine soil decreased metal availability.
- Arsenic extractability was sharply increased by high doses of bottom ash.
- Adding the soil with compost (6.8%) and bottom ash (3.1%) improved vegetation growth.

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ABSTRACT

This study was aimed to 1) properly understand the dynamics of toxic elements (Al, Fe, Mn, Cu, Pb, Zn and As) in a sulphide-mine soil after combined application of compost from urban sewage sludge (SVC) and bottom ashes from biomass combustion (BA) and to 2) optimize the combination of both amendments for vegetation growth. Soil was amended following a D-optimal design and the mixtures (15 in total) were incubated during 30 d. At the end of the incubation, the effects of amendments on the assessed variables as well as the process modelling were evaluated by Response Surface Methodology (RSM). The process modelling confirmed that quadratic models were adequate to explain the behaviour of the assessed variables ($R^2 \geq 0.94$ and $Q^2 \geq 0.75$). Both amendments significantly increased pH and electrical conductivity, while reduced metal extractability. A different behaviour of As respect to metals was observed and high doses of BA sharply increased its extractability. The optimization process indicated that adequate conditions for vegetation growth would be reached adding the soil with 6.8% of SVC and 3.1% of BA (dry weight). After amendments application the germination and root elongation of three energy crops were significantly increased while lipid peroxidation was decreased. Therefore, the combined application of SVC and BA to a contaminated soil could improve soil conditions and might be expected to have an advantage during plant growth. Moreover, the RSM could be a powerful technique for the assessment of combined amendment effects on soil properties and their effective application in multielement-contaminated soils.

1. Introduction

The large volume of wastes in mine tailings is one of the major environmental concerns in abandoned mines. Sulphide-mine waste mainly consist of pyritic material that oxidize and are characterized by very low pH, high electrical conductivity (EC), enhanced solubility of metals and As and low nutrient availability (Forján et al., 2014; Santos et al., 2014). The toxicity in tailings depends mostly on the easily extractable portion of the total concentration of elements due to their mobility toward water and plants (Anjos et al., 2012; Root et al., 2015). The toxic conditions strongly constrain the vegetation establishment and cause pollution problems in air, soils, aquifers and food chain (Anjos et al., 2012; Houben et al., 2012). These concerns have promoted the emergence of cost-effective alternatives for soil reclamation. Among them, significant efforts have been made to assess the effectiveness of some wastes to remediate contaminated soil as

* Corresponding author.
E-mail address: ana.sevilla@iact.ugr-csic.es (A. Sevilla-Perea).
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some of them can improve the characteristics of the soils, increasing the pH and accelerating the natural processes which decrease the mobility and bioavailability of toxic elements (Kumpiene et al., 2008; Houben et al., 2012; Forján et al., 2014).

Sewage sludge is an inevitable by-product of wastewater treatment which can be composted prior to its soil application in order to prevent harmful effects on soil, animals and water (Haynes et al., 2009). Compost application increases soil fertility and decreases mobility/availability of toxic elements because it buffers soil pH and adsorbs or complexes metals with reactive groups of organic matter (Kumpiene et al., 2008). By contrast, some reports have shown that compost could increase the metals and As mobility mainly due to interactions with dissolved organic carbon (González et al., 2012; Beesley et al., 2014).

Bottom ash (BA) is a waste material continuously discarded from thermal power plants and made from agglomerated ash particles which are too large to be carried in the flue gases (Meawad et al., 2010). Approximately, the 44% of BA is recycled for road aggregate or structural fill (Meawad et al., 2010) but alternative uses need to be investigated for unused BA (Mukhtar et al., 2008). Bottom ash is predominantly alkaline and has proven to be an important source of nutrients (Mukhtar et al., 2008) and to reduce extractable metal contents after soil application (Lee et al., 2011). By contrast, due to the high alkalinity, it could have a reverse effect on metals and As stability (Kumpiene et al., 2008). Ashes could be used as an additive to fertilizer, and their addition to organic wastes improves amendment quality and liming potential, and may thus reduce the amount of compost required to raise the pH to suitable levels for plant growth (Bougnom et al., 2010). However, limited research has been conducted about combined application of BA and compost on contaminated soil, a fact that requires more attention (Mukhtar et al., 2008).

The selection of suitable substrates is a critical step for revegetation practices as interactions among the soil and amendments is complex and is not quite understood. In some cases, an amendment immobilises one pollutant and at the same time, increases the mobility of another (González et al., 2012; Beesley et al., 2014). In other cases, adding two amendments together is less effective relative to individual application (González et al., 2012). Therefore, the design of combined amendments requires more attention to ensure an environmental friendly application.

From a practical viewpoint, the Response Surface Methodology (RSM) is an empirical modelling tool useful for analysing and optimizing the response of multivariate systems (Myers and Montgomery, 2002). RSM provides a significant amount of information from a small number of experiments and describes the interaction of independent factors and their effects on the responses (Sevilla-Perea et al., 2014). This method have been recently used by some researchers for optimization of several reclamation processes on degraded soil such as carbon mineralization and nutrient release after organic amendment applications (Sevilla-Perea et al., 2014, 2015), immobilization of toxic elements (Koo et al., 2011; Naseri et al., 2014) and phytoremediation (Feng et al., 2009). But despite its high potential, the use of the RSM to optimize the combined application of amendments for soil reclamation has been insufficiently investigated.

This study was aimed at using the RSM as a tool to provide insights into the system behaviour of a multielement-contaminated soil after compost and BA application helping to (1) modelling, the combined effects of amendments on agrochemical properties (pH and electrical conductivity) and toxic element availability and (2) predicting the most adequate combination of amendments to reduce soil toxicity and confer optimal agrochemical properties to allow successful re-vegetation.

2. Materials and methods

2.1. Contaminated soil and amendments

The soil (15 kg) was collected in a tailing dump of Riotinto mining district (province of Huelva, SW Spain) using a shovel and filling three sacks of 5 kg. Once in the laboratory, the soil was spread (3–5 cm high) on plastic trays and dried for one week at environmental conditions. Riotinto mining district is located in the Iberian Pyrite Belt whose ore deposit includes one of the largest deposits of pyrite (FeS_2) and other metallic and polymetallic sulphides as chalcopyrite ($CuFeS_2$), sphalerite (ZnS), galena (PbS) and arsenopyrite (FeAsS) (Chopin and Alloway, 2007). The results of the exploitation (nowadays closed) are visible through the bare areas and several waste dumps spread in the zone containing hazardous elements, untreated sulfides and host rocks. The region has a semiarid climate with 570 mm rainfall and soil temperatures 14–27 °C.

The compost of sewage sludge and plant remains (SVC) was supplied by a local company. It was produced by composting a urban sewage sludge and agricultural by-products (50:50, w:w), particularly branches and twigs pruned from olive trees. The mixture was kept in stacks approximately 3–4 m high during 6–10 months of maturation.

The BA was provided by a local biomass power plant which uses, as fuel, a mixture of olive mill waste (alperujo) and energy crops (40:60, w:w).

The three materials were air dried, sieved (2 mm) and characterized. Electrical conductivity (EC) and pH were measured in aqueous suspension 1:2.5 (w:v); organic C by a modified Walkey and Black method (Mingorance et al., 2007); cation exchange capacity was determined with the Meier and Kahr (1999) method; soil exchangeable acidity (H^+ and Al^{3+}) was extracted with 1.0 M KCl and measured by titration with NaOH 0.1 M according to Pansu and Gautheyrou (2006); extractable PO_4^{3-} was determined according to the ammonium molybdate-ascorbic acid method. Soil and amendments were finely ground (50 μm) and microwave digested (MLS-1200 Milestone) with aqua regia (3051A; USEPA, 1998) and total element concentrations were analysed by ICP-OES (iCAP 6500 Duo, Thermo Fisher Scientific,WI, USA). A standard reference materials was employed (Montana Soil, NIST 2111) to verify instrument accuracy. The results obtained for the standard reference materials show a recovery range of 94–103%. All analyses were performed in triplicate and all results were calculated on a dry weight basis.

2.2. Experimental design

The RSM consists of a group of mathematical and statistical techniques based on the fit of empirical models to the experimental data obtained from experimental design. Before applying RSM, a design of experiment is required to select the points where the response should be evaluated (Sevilla-Perea et al., 2014). For this study, a design of experiment determined by the D-optimality criterion described elsewhere was chosen. The D-optimality criterion enables a more efficient construction of a quadratic model (Myers and Montgomery, 2002) selecting a number of design points from a larger set of candidate points. From a statistical point of view, a D-optimal design leads to response surface models for which the maximum variance of the predicted responses is minimized. With the D-optimal design several variables are tested simultaneously with a minimum number of trials (Myers and Montgomery, 2002).

A D-optimal design was built assuming: 1) the sum of the three components (soil + SVC + BA) represents 100%; 2) the doses of each amendment were adjusted from 0 to 30% and 3) the sum of