



Arsenic and selenium mobilisation from organic matter treated mine spoil with and without inorganic fertilisation

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ABSTRACT

Organic matter amendments are applied to contaminated soil to provide a better habitat for re-vegetation and remediation, and olive mill waste compost (OMWC) has been described as a promising material for this aim. We report here the results of an incubation experiment carried out in flooded conditions to study its influence in As and metal solubility in a trace elements contaminated soil. NPK fertilisation and especially organic amendment application resulted in increased As, Se and Cu concentrations in pore water. Independent of the amendment, dimethylarsenic acid (DMA) was the most abundant As species in solution. The application of OMWC increased pore water dissolved organic-carbon (DOC) concentrations, which may explain the observed mobilisation of As, Cu and Se; phosphate added in NPK could also be in part responsible of the mobilisation caused in As. Therefore, the application of soil amendments in mine soils may be particularly problematic in flooded systems.

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1. Introduction

Mining extractive industries are the major source of trace elements contamination in soils (Ernst, 2005). Derelict mine lands are present worldwide, and several of these mine-contaminated areas can be found in southern Spain (Clemente et al., 2005; Hudson-Edwards et al., 1999). Mine impacted zones can be frequently found under flooding conditions, especially when the mine wastes have been disposed in excavated pits or mine ponds forming highly compacted layers of spoil. In such environments trace elements may change their speciation, subsequently modifying their mobility and availability and, therefore, affecting the associated environmental risk (Borch et al., 2010).

Full restoration of mine ponds and pits will require re-vegetation and the establishment of a functional ecosystem, and in many cases (especially in Mediterranean regions) this may need organic and inorganic fertilisation (Mendez and Maier, 2008). In this sense, application of organic wastes to derelict mine soils contaminated with trace elements is a common practice in the restoration of such soils (Bernal et al., 2007). Mature (stabilised) organic materials often have given the best results for the immobilisation of inorganic

contaminants in the soil as they have a larger proportion of highly condensed organic matter, with low solubility in water and elevated microbiological stability (Bernal et al., 2007; Madejón et al., 2006). Olive mill waste compost is a promising material to be used as soil amendment (Albuquerque et al., 2011; de la Fuente et al., 2011; Pardo et al., 2011); it is cheap and abundant in Southern Spain, a region that produces one third of the world's olive oil (Albuquerque et al., 2004), and it may be a suitable material for the reclamation of mine soils in Spain (Burgos et al., 2010). The organic matter in the compost is stable, and it could also be a source of some plant nutrients (Albuquerque et al., 2005). However, its application to soil may also change soil biogeochemical environment and promote changes in trace elements solubility. For the restoration of these soils, inorganic fertilisation can be another (complementary) action to improve soil fertility. In addition, these applications may also cause biogeochemical changes in the soils.

Arsenic has attracted considerable attention in recent years, as its toxicity is of concern, and its speciation in soils is complex and very sensitive to redox conditions (Bhattacharya et al., 2007; Moreno-Jiménez et al., 2012). Under flooded conditions that can be temporarily found in derelict mine ponds even in low rainfall Mediterranean climate, As geochemistry can change dramatically, its solubility increasing under anaerobic conditions where As(III) predominates (Hartley and Dickinson, 2010). It may even be volatilised from rice paddy or mine waste soils in the presence of

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organic matter (Mestrot et al., 2009, 2011). In this context, the application of organic amendments could modify As speciation in soil and, therefore, influence its environmental risk.

Selenium, like As, is a metalloid present mostly as oxyanionic species in soil solution (Losi and Frankenberger, 1997), which additionally can be (bio)methylated and volatilised (Reamer and Zoller, 1980); therefore As and Se have been frequently studied together in soil research (Goh and Lim, 2004; Strawn et al., 2002). Notwithstanding, we need to better understand Se biogeochemistry and speciation in the soil–soil solution system, e.g. for biofortification of food purposes, although high Se can pose toxicity (Zhu et al., 2009).

Soil conditioning with inorganic and organic amendments may disturb trace element mobility in the short-term (potentially provoking both positive and negative effects), but it has undoubtedly long-term benefits such as physical stabilisation, improvement of soil water holding capacity, drainage, fertility, etc., therefore getting better soil services to support ecosystem (plants) establishment and sustainability. The aim of this paper was to study the short-term changes in trace element solubility (As, Cd, Cu, Sb, Se and Zn) in an organic and/or inorganic amended contaminated mine soil under flooded conditions, focussing on the effects on As speciation (since it is very sensitive to these conditions). Selenium and As volatilisation was also measured as it is a part of their cycle that is frequently overlooked in soil studies.

2. Materials and methods

Soils were collected from Gosan-Cobre lake, the Rio Tinto mine tailing (Huelva, South Spain, 37° 44.079' N, 6° 35.447' W; Supplementary material Fig. SM1), a former copper mine that resulted in extensive contamination by mine spoils with high concentrations of As and other trace elements, such as Cu or Zn. The soil was air dried, disaggregated, sieved to 2 mm and homogenised. Soil inorganic and/or organic amendments were added to the soil, mirroring the necessary support that the mine impacted site may need for plant establishment and primary succession of a natural ecosystem. Olive mill waste compost (OMWC) was used as an organic fertiliser and soil amendment. It was prepared by mixing OMW and cow manure (11% on a dry weight basis), with a total composting time of 36 weeks (Cegarra et al., 2006). NPK fertilisation was used as the inorganic amendment to discern the effects of the nutrients from those of the organic matter. Soil (100 g) was mixed with the different inorganic (-/+ NPK) and/or organic amendments (-/+ C) resulting in the following treatments:

- 1: Soil (-C/-NPK).
- 2: Soil + NPK (applied solid at a rate of 85 mg KNO₃ and 37 mg (NH₄)₂HPO₄, approximately equivalent to 20N:8P:40K; Barrutia et al., 2009) (-C/+NPK).
- 3: Soil + 1% (w/w) OMWC (+C/-NPK).
- 4: Soil + 1% OMWC + NPK (+C/+NPK).
- 5: Soil + 5% OMWC (+C/-NPK).
- 6: Soil + 5% OMWC + NPK (+C/+NPK).

The mixtures were placed in closed vessels (reactors) in triplicate, water was added to double that of the soils water holding (field) capacity (WHC) and the reactors were placed in an incubation chamber in the dark and at 28 °C for 2 months. One Rhizon sampler (Eijkelkamp Agrisearch Equipment, The Netherlands) was inserted in each reactor and soil pore water was extracted after 7, 28 and 56 days of incubation. Pore water was conserved either in 1% HNO₃ for total elements concentration analysis or in 0.5 mM EDTA for speciation analysis (Mestrot et al., 2011). Redox potential in the reactors was monitored 3 times, after pore water sampling, by immersing the probe in the top layer of water in each reactor and then recording the value of Eh after 3 min equilibration. Arsenic in the headspace of the reactors was continuously sampled using traps as described by Mestrot et al. (2009) and processed accordingly: the air from the reactor was pumped out and passed through the trap (a column containing silver nitrate impregnated silica gel) during 4 weeks using a multichannel peristaltic pump running at 12 ml min⁻¹. The traps were then collected (at 4 and 8 weeks) and As was removed from the traps by digestion in concentrated nitric acid.

Soil pH and electrical conductivity (EC) were measured in a suspension 1:2.5 (w:v) soil:deionised water. Total-N and total organic-C (TOC) concentrations were measured in an automatic microanalyser (EuroVector, Milan, Italy). Available-P was analysed colourimetrically as molybdovanadophosphoric acid after extraction with 0.5 M NaHCO₃ (1:10, w:v). Soil (0.1 g) was digested using concentrated HNO₃:H₂O₂ (1:1) in block digesters at 100–140 °C for 4 h, the supernatant was diluted after

cooling to 10 ml, then an aliquot of 1 ml of this extract was diluted to 50 ml with milli-Q water and total elements concentrations determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent 7500c, Japan). Traps were digested in blocks as well, and trace elements were extracted using 1:2 HNO₃:H₂O₂ at 100–140 °C for 4 h, and diluting the supernatant to 10 ml. Soil soluble metal concentrations (0.1 M CaCl₂, 1:10 w/v) were determined by atomic absorption spectrometry (UNICAM 969 atomic absorption spectrometer). Soil available-As was determined in a 0.5 M NaHCO₃ extract (1:10 w/v) by hydride generation atomic fluorescence spectrometry (PS Analytical Millennium Excalibur, UK). All the analyses were done at least in duplicate and adjusted to values for oven-dried soil (105 °C for 24 h).

Arsenic speciation in pore water was determined by anion exchange High Performance Liquid Chromatography ICP-MS (HPLC, Hamilton PRP-X-100, Switzerland). Also, simultaneous coupling of Electro Spray Ionization–Mass Spectrometry (Orbitrap Discovery, in FT mode) with ICP-MS (Element2, in low resolution mode) after separation using HPLC (Accela) was used to ensure that the predominant species in pore water was DMA (all the equipments from Thermo Fisher, Bremen, Germany). Thioarsenic species can elute at the same retention time as DMA after anion exchange separation (Hansen et al., 2004), hence the importance to precisely characterise DMA by ESI–MS and to monitor its exact mass.

A certified reference material (NCS ZC 73007, agricultural Chinese soil, 18 mg As kg⁻¹, 0.25 mg Cd kg⁻¹, 32 mg Cu kg⁻¹, 1.7 mg Sb kg⁻¹, 0.51 mg Se kg⁻¹ and 100 mg Zn kg⁻¹) was also digested and found to obtain recoveries of 92% for As, 88% for Cd, 76% for Cu, 103% for Sb, 87% for Se and 101% for Zn, with a coefficient of variance <8% for all of them.

Statistical analyses (ANOVA and Multiple Linear Regression) were carried out using the software SPSS version 19.0 (SPSS Inc.). Differences between means were determined using Tukey's test, and normality of the data was checked by the Kolmogorov–Smirnov's test.

3. Results and discussion

3.1. General changes in the chemical composition of soil and pore water

Olive mill waste compost application significantly increased soil pH, EC, TOC, TN and extractable Cu concentrations (Table 1), especially when applied at the higher dose. The increase of pH, TOC and Cu extractability in an acid mine soil after OWMC application was found in other experiments using this material (Burgos et al., 2010; Pardo et al., 2011). Previous studies have reported a decrease of total trace elements concentrations in polluted soils when they are amended with high rates of organic materials (i.e. higher than 10% w:w of compost, which may dilute the contamination; Karami et al., 2011). In our study, the treatments did not cause any dilution effect on trace element total concentrations (results not shown) and no changes in soil mineralogy can be expected. Inorganic fertiliser application increased, as predictable, P and As extractable concentrations in the soil, and no relevant effects of the amendments were found for Cd and Se extractability while that of Zn was reduced when treated with the higher compost dose and inorganic fertiliser (Table 1).

Olive mill waste compost changed the composition of the pore water in relation to both dissolved organic-C (DOC) and trace elements concentrations (Fig. 1, $p < 0.001$). Regarding trace elements, the increased DOC concentrations that resulted after compost addition may be responsible for the increase of Se and Cu concentrations in pore water. Mobilisation of Cu in soils has been also previously attributed to its interaction with DOC, through the complexation with organic molecules, potentially soluble and thus mobile in the soil (Bernal et al., 2007; Kunhikrishnan et al., 2011). Eich-Greatorex et al. (2010) reported an increase of Se availability in soils with high organic matter, which may be due to the competition for soil binding sites by DOC or by labile P compounds (Eich-Greatorex et al., 2007; Masset et al., 2000). Meanwhile, Zn and Cd concentration slightly decreased, possibly as a consequence of the combination of (i) increase in pore water pH caused by the organic amendment and (ii) the sorption on stable organic matter in compost (Park et al., 2011). NPK also induced a slight mobilisation of Cu and Se (Table 2, $p < 0.05$). In the case of Se, this mobilisation may be partly explained by the application of P, able to compete

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