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Influence of lead in the sorption of arsenate by municipal solid waste composts: metal(loid) retention, desorption and phytotoxicity



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HIGHLIGHTS

• MSW-composts were able to sorb As(V) at acidic pH only when Pb(II) was present.

• Pb(II) and As(V) were simultaneously sorbed by the MSW-composts investigated.

 \bullet Sequential extraction indicated different interactions between As(V) and Pb(II).

• As(V) phytotoxicity was buffered by MSW-composts when Pb(II) was present.

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ABSTRACT

The ability of two municipal solid waste composts (MSW-C) to sorb As(V) in the presence of Pb(II) and in acidic conditions was investigated. Sorption isotherms and kinetics showed that both MSW-C were able to sorb As(V) in a similar way (~0.24 mmol g⁻¹ MSW-C), but only when Pb(II) was present (0.45 mmol L⁻¹). The concomitant sorption of Pb(II) by both MSW-C (~0.40 mmol g⁻¹) suggested that the metal cation was likely acting as bridging element between the negatively charged functional groups of composts and As(V). SEM-EDX analysis of the MSW-C + Pb(II) + As(V) systems supported the association between Pb(II) and As(V), while sequential extraction procedures and organic acids treatment showed that As(V) was strongly retained by MSW-C + Pb(II) and suggested the presence of different interaction types between As(V) and Pb(II). Plant growth experiments highlighted the key role of Pb(II) in the reduction of As(V)-phytotoxicity for triticale plants (×*Triticosecale* Wittm.) in the presence of MSW-C.

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1. Introduction

Arsenic (As) and lead (Pb) are toxic elements to humans and hazardous to the environment. They often coexist at high concentrations in contaminated soils and groundwaters due to industrial and agricultural practices such as mining, ore processing, metallurgy, and insecticide application. For instance, the critical presence of Pb and As in many North American orchards was attributed to the extensive application of lead arsenate as insecticide from the late 1800s until the 1960s (Liang et al., 2014; Lim and McBride, 2015; McBride et al., 2015; Moon and Dermatas, 2007; Wang and Mulligan, 2009).

Remediation of As and Pb co-contaminated soils by chemical stabilization may be difficult lead being commonly present as divalent cation [Pb(II)], and arsenic as arsenate oxyanion, arsenious acid and/or arsenite in the 4.0–9.5 pH range. Consequently, remediation techniques based on the addition of selected amendments are not expected to be fully effective at reducing the solubility and bioavailability of both elements (Fleming et al., 2013; Liang et al., 2014; Mele et al., 2015). Depending on the soil pH and the point of zero charge of the amendment, this latter will show a quite variable affinity towards cationic and/or negatively charged contaminants.

However, the addition of organic matter (OM) such as compost, to degraded soils polluted with As and Pb, besides representing a sustainable way to enhance the organic matter content of such soils, could be an effective strategy for decreasing the mobility of both contaminants and improving the physico-chemical and biological properties of the soils (e.g. Beesley et al., 2014; Gadepalle et al., 2008; Hartley et al., 2010; Moreno-Jiménez et al., 2013; Tandy et al., 2009). In



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principle, compost added to polluted soils could affect Pb and As mobility through different mechanisms and interaction types. In particular, the organic matter of compost could reduce the concentration of easily bioavailable Pb in soil due to the formation of insoluble Pb-organic matter complexes. The functional groups of the organic matter involved in such immobilization process include carboxyl, phenolic, quinone, sulfhydryl, and hydroxyl groups, the majority of which are negatively charged at neutral pH and can easily interact with Pb cations (Paradelo and Barral, 2012; Wang and Mulligan, 2009). Likewise, a decrease of bioavailable arsenic in soils amended with organic matter could be expected as reported by some researchers (Cao et al., 2003; Gadepalle et al., 2008). As pointed out by these authors, arsenate can be adsorbed onto high molecular weight (insoluble) organic matter reaching a maximum adsorption around pH 5.0. In this case, and similarly to phosphate, the formation of stable mono and di-esters bonds involving the hydroxyl functional groups of OM and the arsenate [As(V)] anion was likely the most relevant immobilizing mechanism (Wenzel, 2013). It was also recently suggested that polyvalent metal cations such as Al(III) and Fe(II/III) can act as bridges between As(V) and insoluble OM, further decreasing the mobility of As(V) in OM amended soils (Fleming et al., 2013; Ko et al., 2007; Sharma et al., 2010, 2011; Udovic and McBride, 2012). Given that a similar role can be played by Pb(II), the addition of compost to As(V) and Pb (II) co-contaminated soils could contribute to reduce the bioavailability of both elements. However, the structure of these ternary complexes and the mechanisms of their formation and stability in soil still remain obscure (Sharma et al., 2010).

On the other hand, many experimental results showed that compost addition to As and Pb co-contaminated soils may increase the mobility and leaching of the metalloid (Fleming et al., 2013; Hartley et al., 2010; Moreno-Jiménez et al., 2013; Tandy et al., 2009; Udovic and McBride, 2012). Dissolved organic carbon (DOC) and inorganic compounds of compost (e.g. phosphate and/ or sulfate) may increase As mobilization by competing for retention sites on the soil surfaces (Beesley et al., 2014; Lin et al., 2008; Wang and Mulligan, 2009). Moreover, the formation of soluble As-DOC or Pb-DOC complexes in such amended soils was also highlighted by several researchers (Fleming et al., 2013; Lin et al., 2008; Liu et al., 2011).

Taken together, these data indicate the lack of a general consensus on the influence of added compost on Pb and As mobility in soil. This is understandable since a number of variables such as soil properties, compost composition, nature and concentration of pollutants can influence the compost-driven (im)mobilization of the two pollutants (Sharma et al., 2011; Udovic and McBride, 2012).

This implies the need to deepen the knowledge on the interactions between As, Pb and compost and, in this regard, it should be mentioned that the studies addressing this issue are rare if not missing at all. For instance, Wang et al. (2015) investigated the simultaneous sorption of As and Pb by materials containing both organic and inorganic phases (i.e. Mn oxide-biochar composites). However, given the mixed nature of the composites, the contribution of the organic phase in the sorption process remained unclear.

Therefore, the aim of this study was i) to evaluate the sorption capacity of two municipal solid waste composts (MSW-C) with respect to As(V) and Pb(II) at acidic pH; ii) to assess the possible interactions involved in the sorption processes and iii) to investigate the role of two low molecular weight organic acids (LMWOAc) commonly present in soils, i.e. citric and malic acids, in the release of Pb(II) and As(V) sorbed by MSW-C. Finally, the influence of the MSW-C with respect to As(V) and Pb(II) toxicity for triticale plants (*×Triticosecale* Wittm.) was addressed.

2. Materials and methods

2.1. Municipal solid waste composts (MSW-C)

Two MSW-C, i.e. MSW-C1 and MSW-C2, were obtained from the Secit S.p.A. facility plant of the Consorzio ZIR, Chilivani-Ozieri (Sassari, Italy) and from Verde Vita S.r.l. (Porto Torres, Sassari, Italy) respectively.

The composts were dried at 25 °C, then ground and sieved. Particles smaller than 2 mm in diameter were used for chemical analyses which were performed according to Chefetz et al. (1996) and Castaldi et al. (2008) (Table 1). The elemental composition (CHN) of composts was determined using an elemental analyzer (Leco CHN 628) with Oat meal Leco part n° 502-276 as calibration sample. The method reported by Ciavatta et al. (1990) was used to determine the content of humic substances.

The total concentration of selected trace metals was determined on dried composts (105 °C) using a flame atomic absorption spectrometer (Perkin Elmer Analyst 600) after microwave (Milestone MLS 1200) digestion of the samples with aqua regia (HNO₃/HCl, 1:3 ratio). For sorption/desorption trials, compost samples were brought to pH 4.5 with either 0.01 or 0.1 M HNO₃ solutions. The chemical characteristics of the composts at pH 4.5 are reported in Table 1. Each determination was conducted on triplicate MSW-C samples and mean values \pm standard deviations are reported.

2.2. Sorption isotherms of As(V) on MSW-C in the presence of Pb(II) at pH 4.5

Since preliminary experiments revealed that MSW-C1 and -C2 did not adsorb As(V) in a wide pH range (4.5–9.0), simultaneous sorption isotherms of As(V) and Pb(II) were carried out to evaluate the possible influence of Pb(II) on As(V) sorption by MSW-C. At the pH selected for this trial, i.e. 4.5, and at the highest metal(loid) concentrations tested (i.e. 0.45 mM), more than 99% of Pb(II) and As(V) added to aqueous solutions was present in solution thus ruling out the formation of Pb(II) and As(V) precipitates (as also predicted by the Medusa Software). MSW-C1 and -C2 samples brought at pH 4.5 with HNO₃ solutions (0.01–0.1 M), were artificially polluted with solutions containing increasing concentrations of As(V), and constant concentrations of Pb(II) [0.45 mM Pb(NO₃)₂]. Sodium arsenate dibasic heptahydrate (Na₂HAsO₄ 7H₂O) and lead nitrate [Pb $(NO_3)_2$ were used as sources of As(V) and Pb(II) respectively for sorption isotherms and sorption kinetic studies. Chemicals were of analytical grade and used without further purification. Polyethylene bottles containing 0.1 g of MSW-C1 or -C2 were filled with 100 mL solution of As(V) and Pb(II) at pH 4.5. Nine different As (V) concentrations were used [from 0.04 to 0.45 mM].

The final mixtures were shaken for 24 h at constant temperature (25 ± 1 °C). Afterwards, the samples were centrifuged at 8000 rpm for 15 min and filtered using filter paper 0.2 µm. As(V) in the supernatant was measured by ion chromatography using an IonPac AS9-HC Analytical Column equipped with an Ion-Pac AG9-HC Guard Column, 4 mm. Sodium carbonate (11 mM) was employed as the eluent at a flow rate of 1.0 mL min⁻¹. The sample loop was 10 µL. The concentration of Pb(II) in the supernatant was determined with a flame atomic absorption spectrometer (Perkin Elmer Analyst 600). Each point of the sorption isotherms was carried out on three independent samples and mean values ± standard deviations are reported.

2.3. Sorption kinetics of As(V) and Pb(II) on MSW-C at pH 4.5

Simultaneous sorption kinetics of As(V) and Pb(II) by MSW-C were carried out. For kinetic studies, polyethylene bottles con-

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