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Towards sustainable remediation of contaminated soil by using diasporic bauxite: Laboratory experiments on soil from the sulfide mining village of Stratoni, Greece

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ABSTRACT

Various mineral-based amendments can be used in raw or modified form for inducing immobilization of inorganic contaminants in soil with different modes of molecular-scale sequestration. In this study, diasporic bauxite in raw form as well as after heat treatment at different temperatures (105 °C, 350 °C and 450 °C) was tested for its effectiveness in reducing the mobility of potentially harmful elements (PHEs) in contaminated soil. A pot experiment was set up where raw and calcined bauxite was mixed at different proportions with contaminated soil from the sulfide ore mining village of Stratoni, north Greece. Total concentrations in soil have averages of 1000 mg/kg Pb, 712 mg/kg Zn, 6 mg/kg Cd, 2900 mg/kg Mn and 296 mg/kg As. The effectiveness of bauxite amendment was evaluated by comparing leachable PHE concentrations of treated and untreated soil after a four week period of repeated cycles of wetting and mixing. Both raw and calcined bauxite in a mixing proportion of 7% reduced leachable concentrations of the elements in comparison to the original soil. Samples that were mixed with activated bauxite (105 °C, 350 °C) and raw bauxite showed similar results. The corresponding reduction of water leachable fraction of PHEs was determined as high as 75% for Zn, 66% for Mn, 55% for Cd, 51% for Pb, and 41% for As. Extractable concentrations by the Toxicity Characteristic Leaching Procedure (TCLP) remained relatively low for Pb (60% reduction), Zn (30% reduction) and As (28% reduction). The maximum efficiency was observed by using treated bauxite at the temperature of 450 °C with negligible water leachable concentrations after treatment. This difference in retention efficiency of elements between the heat treated bauxite samples is attributed to phase transitions that occur in Al-oxyhydroxides and Fe-oxides/oxyhydroxides of bauxite when heated at higher temperatures.

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1. Introduction

Sustainable remediation of contaminated land, in contrast to traditional practices such as excavation and removal of impacted soil, requires the adoption of more efficient strategies that conserve resources and protect air, water, and soil quality through reduced emissions and other waste burdens (Hodson, 2010). The immobilization of inorganic - non degradable contaminants in soil has been suggested as a sustainable remediation method aiming in breaking the pathway between the source and the receptor in the widely used 'source- pathway-receptor' risk assessment approach. Within this framework, various mineral-based amendments can be used in raw or modified form for inducing immobilization of inorganic contaminants in-situ, with different modes of molecular-scale sequestration within the soil matrix (e.g., Garau et al., 2007; van Herwijnen et al., 2007; O'Day and Vlassopoulos, 2010; Zotiadis et al., 2012; Almaroai et al., 2014).

However, questions concerning the sustainability of soil treatment can only be addressed by long-term studies which are generally scarce, e.g. Mench et al. (2006).

Among the different natural materials that have been proposed as effective stabilization agents for metal(oids) in soil, oxides and their precursors have been extensively studied. Komárek et al. (2013) have critically reviewed literature data on mechanisms involved in the immobilization process by using iron, manganese and aluminum oxides as well as the influence of such amendments on plants and microbial population. Their conclusions underline that there is considerable elemental specificity with respect to the immobilization efficiency depending not only on the type of amendment, but also on the physicochemical characteristics of the soil. Also, they have identified research data gaps on the long term stability of the newly formed oxides in the treated soils and raised concerns on potential unwanted concentration increases of trace elements contained in some stabilizing amendments such as red mud and steel shot.

Bauxite, the primary Al ore, consists of a mixture of minerals including gibbsite (Al(OH)₃), boehmite (γ-AlO(OH)) and diasporite (α-

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AlO(OH)), mixed with goethite (FeO(OH)) and haematite (Fe₂O₃), the clay mineral kaolinite and small amounts of anatase (TiO₂) and ilmenite (FeTiO₃). It is the principal raw material in the production of alumina with a series of additional industrial uses that have been developed over the years. Studies on the potential use of bauxite for environmental applications are focused to water treatment and decontamination. Specifically, the use of bauxite has been proposed for the effective removal of anionic species such as phosphates (Altundoan and Tmen, 2002), fluoride (Sajidu et al., 2012; Lavecchia et al., 2012), Cr(VI) (Erdem et al., 2004; Baral et al., 2007) and As (Fares et al., 2009) from contaminated water and aqueous solutions. Calcined bauxite has been proven to be effective in the removal of both As(V) (Mohapatra et al., 2008) and As(III) (Bhakat et al., 2007) from water.

Concerning the treatment of contaminated soil, some studies have demonstrated the effective use of red mud, the industrial residue produced in large quantities during the extraction of alumina from bauxite, for metal immobilization (Lombi et al., 2002; Gray et al., 2006). Specifically, by employing laboratory or field scale experiments it has been shown that red mud results in a redistribution of heavy metals from soluble and exchangeable pools to the Fe, Al and Mn oxide fractions. Once heavy metals are specifically sorbed onto these oxide surfaces, potentially they may become irreversibly fixed as a result of several mechanisms, including migration of metal into micropores on the surface of the oxides, solid state diffusion of ions into the crystal lattice of the oxides, or perhaps occlusion of metals (Gray et al., 2006 and references therein). Red mud specific chemisorption and metal diffusion into the lattice of Fe and Al oxides combined with the effective raise in soil pH after amendment has been suggested as probable mechanisms responsible for the fixation of metals (Lombi et al., 2002). The effectiveness of synthetic Al(OH)₃ and FeOOH for decreasing the water-extractable fraction of As in contaminated soil from mining activities has also been demonstrated in lab-scale experiments (García-Sánchez et al., 2002). However, to the best of our knowledge, bauxite ore has never been tested as a mineral based amendment for immobilization of metal(oid)s in contaminated soil despite its favorable mineralogical-geochemical characteristics and relatively low cost.

The aim of this study is to assess the ability of red bauxite to reduce mobility and availability of a range of metal(oid)s in soil contaminated by sulfide mining and processing. In order to better understand the mechanism responsible for metal fixation bauxite was used in raw form as well as after thermal activation over a range of temperatures. We report on the changes in the mobility of Pb, Zn, Cd, As and Mn as affected by different mixing proportions of thermally activated bauxite and by mixing with bauxite calcined at different temperatures.

2. Materials and methods

2.1. Bauxite amendment and contaminated soil

The adsorbent material chosen for the present study was Greek diasporic bauxite. S&B Industrial Minerals S.A. (currently IMERYS S.A.) kindly provided the samples originating from baghouse dust collectors of the bauxite facilities at Itea, central Greece. In such facilities the major sources of dust emissions are the dryers and emissions are controlled with fabric filters; baghouse dust removal efficiencies of 99% are achievable. Dedusting of the fabric filters produces relatively fine grained bauxite material which is sorted in two fractions: a fine grained (<50 µm) and a coarser grained (50 µm–1 mm). Preliminary testing of both grain sizes for the treatment of contaminated soil verified that the fine grained material was more effective for PHEs fixation and was subsequently used for the experiments of the present study without any further grinding. For heat activation, 100 g of bauxite samples (<50 µm) placed in porcelain dish were heated in a muffle furnace at 105, 350, 450 and 800 °C for 4 h. After the heating period, the samples were cooled in a desiccator, and preserved in closed vessels containing silica gel during the experiments.

Contaminated soil was collected near the residential area of Stratoni mining village, at Chalkidiki, north Greece. A detailed discussion on the soil contamination and potential health concerns in this particular area is presented elsewhere (Argyraki, 2014). It is sufficient to say here that the area is the only example of active mining and processing of Pb–Zn sulfide ore in Greece. Mining operations and ore processing to produce sphalerite and galena concentrates at the Stratoni flotation plant over the last 60 years have resulted to surface soil contamination with Pb, Zn, Cd and As. A composite soil sample of 30 kg was collected from the 0–20 cm depth by mixing 5 sub-samples collected over an area of 2500 m², air dried and sieved to <2 mm. The soil was thoroughly homogenized by manual mixing and was used to fill 1-kg polypropylene pots.

2.2. Soil treatment and analytical procedures

Six treatments were prepared by mixing calcined (350 °C) bauxite at different proportions (0%–control, 1%, 2%, 4%, 5% and 7%). Three additional treatments were prepared by mixing raw and heat treated (105 and 450 °C) bauxite at a proportion of 7%. All treatments were mixed thoroughly and watered with deionized water. Homogenization of treated soil was repeated on a weekly basis over a one-month period (4 weeks), in parallel with soil rewetting to maintain near-saturation soil conditions. Soil pH was monitored weekly by taking potentiometric measurements after mixing soil with distilled water at a ratio of 1:2.5. A final set of samples was collected at the end of the 4-week period to study the final geochemical characteristics of the treated soil against the control. Three subsamples (100 g) of each soil treatment sample were shaken by rotation in polyethylene bottles with 1000 ml of distilled water for a period of 24 h according to the standard leaching test EN 12457-2 (2002). The solids were separated by vacuum filtration through a 0.45 µm cellulose filter. Concentrations of Pb, Zn, Cd, As and Mn in the collected solutions were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). An additional set of treated soil samples was subjected to the Toxicity Characteristic Leaching Procedure (TCLP) (USEPA, 1992) in order to examine the effect of induced acidity on the treated soil. Concentrations of the same elements as above were measured by ICP-MS. In addition to the pot experiment, 50 g of raw and calcined bauxite (350 °C) underwent four wetting cycles during the same 4-weeks time period in order to investigate any mineralogical and structural changes due to re-hydration.

The total elemental content of bauxite was determined by X ray fluorescence (XRF) while the mean concentrations in contaminated soil were measured by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) after total dissolution of five replicates of the collected composite field sample by using a mixture of strong acids. Mineralogical analysis of all materials used has been performed by powder X-ray diffraction (P-XRD) using a Siemens D-500 diffractometer with Cu–Kα radiation, and scans from 5° to 65° 2θ with a scan rate of 2°/min; step interval = 0.02; voltage 40 = kV; current = 30 mA. Scanning electron microscopy (SEM) and energy dispersive spectra (EDS) analysis were carried out on resin-impregnated/carbon-coated samples

Table 1
Chemical and mineralogical composition of raw bauxite. (LOI: loss of ignition at 1000 °C).

Chemical composition		Mineralogical composition	
Parameter	w/w (%)	Mineral phase	w/w (%)
Al ₂ O ₃	47.56	Boehmite/diaspore	21.1
Fe ₂ O ₃	17.85	Diaspore	36.8
SiO ₂	9.28	Kaolinite	14.7
CaO	4.44	Goethite	17.8
TiO ₂	3.17	Hematite	3.4
S	0.45	Anatase/rutile	2.7
Mn	400 (mg/kg)	Calcite	5.4
Zn	89 (mg/kg)		
LOI	15.50		

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