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The removal of lead and nickel from the composted municipal waste and sewage sludge using nanoscale zero-valent iron fixed on quartz

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ABSTRACT

Reducing the concentration of heavy metals including lead (Pb) and nickel (Ni) in organic contaminants such as municipal wastes and sewage sludge is of health and environmental importance. Nanoscale zero-valent iron (NZVI) particles can effectively remove heavy metals from contaminated aqueous and solid media. It was accordingly hypothesized that it is possible to recycle and detoxify organic waste materials containing heavy metals using NZVI and NZVI fixed on quartz (QNZVI). The objective was to investigate the effects of NZVI type, concentration (2% and 5%) and contact time on the removal of Pb and Ni from raw compost, compost fermented with beet molasses, and leachate using a factorial design. The results indicated the significant reduction of DTPA-Pb and DTPA-Ni concentration, in all the organic compounds treated with NZVI and QNZVI ($P = 0.01$), compared with control. Increased concentration of NZVI in all treatments, increased the rate of DTPA-Pb and DTPA-Ni ($P = 0.01$) at 113.1% and 180% for Pb (NZVI at 2% and 5%), and at 16.3% and 23.3% for Ni, irrespective of the NZVI type. The reducing trend of extractable Pb and Ni in all the organic compounds was the same, quick reduction at the beginning, followed by a negligible rate. The highest reduction rates for Pb (at one hour) and Ni (at 672 h) were equal to 72.93% and 23.27%, respectively. NZVI at 2% was more efficient than NZVI at 5%. There were not any significant differences between NZVI and QNZVI on the removal of Pb and Ni from the organic contaminants. It is possible to immobilize and reduce the concentration of heavy metals such as Pb and Ni in organic contaminants using NZVI, which is affected by NZVI properties, concentration, and contact time, as well as by organic contaminant type.

1. Introduction

The rate of organic matter in arid and semi arid areas, is little, for example, in Iran in most arable fields, it is less than 1% (Six et al., 1998). Accordingly, treating such fields with organic compounds, improves the properties of soils such as soil water retention, pH, nutrient content, soil microbial activities, etc. Using biosolid wastes can be a suitable method for both supplying the arable fields with the essential organic matter, and for recycling such compounds (Karathanasis et al., 2005).

Research work has indicated the increased use of solid waste, for the production of compost, in agricultural fields. The use of compost can favor the agricultural production by the following. 1) Increasing the essential micronutrients, 2) decreasing pH, 3) reducing soil erosion, 4) enhancing soil microbial activity and population, 5) reducing the use of chemicals such as fertilizer, pesticides and insecticides, 6) improving soil properties, 7) reducing the amount of solid waste, and 8) enhancing the environmental quality (Smith et al., 1998; Zebarth et al., 1999; Knowles et al., 2011; Park et al., 2011).

However, there are disadvantages with the use of solid waste, (especially in a long time period) including: 1) the loss of soil fertility, and 2) adverse effects on plant and human health (pathogenic and heavy metal presence) (del Carmen Florido et al., 2011). Accordingly, it is of significance to use methods, which can increase the healthiness of such solid wastes and result in their proper recycling.

The organic amendments are able to absorb heavy metals, and hence can be used for the process of bioremediation (Beesley et al., 2014). However, due to the increased concentrations of heavy metals, the use of such organic amendments in the agricultural fields can adversely affect the environment (Filgueiras et al., 2002). The behavior of heavy metals in the soil is determined by their chemical properties and their mobility rather than their total concentration (Bradl, 2004). According to the WHO, the most important heavy metals affecting the health of plant, microbes and human are arsenic (As), cadmium (Cd), mercury (Hg) and lead (Pb). It is also worth mentioning that Pb can be deposited from the atmosphere, especially in the contaminated areas,

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and similar to Cd, nickel (Ni) is also found in the composition of chemical fertilizers (Lee et al., 2005; Liu et al., 2007).

It has been indicated that the use of nano methods may be an efficient method for the removal of heavy metals from pollutants including organic compounds (municipal waste and sewage sludge). Iron nanoparticles including nanoscale zero-valent iron (NZVI) are not expensive, nontoxic, and insoluble, and due to their really fine size, and great surface area, and reaction rate can efficiently and strongly fix different contaminants, especially in situ (Li et al., 2011; Xu et al., 2012; Nasiri et al., 2013; Azari and Bostani, 2017).

The use of NZVI can be a cost effective method, and friendly to the environment (Liu et al., 2008). The nano products containing nanozeolites, and nano iron oxides, as well as phosphate and sulfate nono particles can be effectively used for immobilizing heavy metals in biosolid wastes (Zhang et al., 2010; Feng et al., 2014). Different parameters may influence the bioremediation potential of NZVI including their type, concentrations, and contact time as well as the properties of the pollutants (Azari and Bostani, 2017).

The experiments on soil and water samples have indicated the higher efficiency of NZVI on the removal of metal cations such as zinc, cadmium, copper, nickel, lead and silver, compared with the traditional methods (Cao and Zhang, 2006; Li and Zhang, 2007). It has also been indicated that it is possible to enhance the mobility of NZVI using different compounds (Jiemvarangkul et al., 2011).

The stability of NZVI particles is an important point affecting their activity and their use for environmental remediation. Accordingly, we hypothesized that if the NZVI particles are fixed on quartz, their stability and hence their removal efficiency may increase. Research work has indicated that it is possible to enhance the removal efficiency of NZVI if such particles are fixed on different products (Hsieh et al., 2010; Hwang et al., 2014).

Due to their little rate of oxidation-reduction, NZVI can be used as reductants, which are cheap, effective and environmentally friendly (equation 1).

$$Fe^0 \rightarrow Fe^{2+} + 2e^- E^0 = -0{\cdot}441 \quad (1)$$

Fe (II) can be then oxidized (by loosing an electron) to Fe (III).

Most of the research work on the effects of NZVI fixing the heavy metals has been done in aquatic environments, but data on the important issue of removing heavy metals from organic compounds using NZVI, is little. It is because detoxification and recycling such products are of environmental and economical significance as they can also be used as organic amendments in the agricultural fields. Accordingly, this research work was conducted to investigate the effects of NZVI on the removal of heavy metals from organic compounds. Due to the following important details this research work was proposed and conducted. 1) With respect to the toxicity of organic compounds by heavy metals there are concerns on the use of composted solid waste and sewage sludge in the agricultural fields, and 2) the use of nano particles as the new generation of heavy metal extractants. It was accordingly hypothesized that it is possible to recycle and detoxify organic waste materials containing heavy metals using NZVI and NZVI fixed on quartz (QNZVI). The objective was to investigate the effects of NZVI type, concentrations and contact time on the removal of Pb and Ni from organic contaminates including composted waste materials (raw, fermented with beet molasses, and leachate).

2. Materials and methods

2.1. Experimental design

The experiment was a factorial in three replicates with the following treatments: 1) three different organic compounds including raw compost, compost fermented with beet molasses, and leachate, 2) two nanoscale zero-valent iron (NZVI) compounds including NZVI and NZVI fixed on quartz (QNZVI), 3) NZVI and QNZVI concentrations of 2 (N1) and 5% (N2), and 4) the NZVI and QNZVI contact time of 1, 4, 16, 24, 48, 168, 336, 672, and 1344 h with the organic compounds. The reasons for the selection of such NZVI and QNZVI concentrations were according to the following: 1) finding the most efficient concentrations, 2) economical reasons, to make it possible to try it at the pilot and then large scale, 2) avoiding toxic effects, if any, 3) to try the greater concentrations, if essential.

2.2. NZVI and QNZVI synthesis

The NZVI and QNZVI were synthesized using the method of sodium borohydride according to the following details (Wang and Zhang, 1997; Zhang, 2003). It has been indicated that due to its simplicity and chemical homogeneity, the use of sodium borohydride (NaBH4), as a chemical reductant, is a useful method, (Sun et al., 2008; Li et al., 2009) for the synthesis of NZVI, and is accordingly widely used, compared with the other methods, (Hwang et al., 2011, 2014). First, 75 ml of methanol was poured into a 250-ml volumetric flask and was brought up to volume using distilled water. Five grams of iron sulfate was poured into the volumetric flask containing methanol, and using a stirrer was well mixed. The acidity of the solution was adjusted using a 2-N sodium hydroxide solution from 3.5 to 6–7. The solution was mixed with sodium borohydride (two grams of sodium borohydride were poured into a 25-ml volumetric flask and the solution was brought up to volume), which was mixed with the solution gradually, and stirred for 40 min. The solution was centrifuged for 15 min at 5000*g* per minute. The supernatant was poured away and the solid phase was filtrated using methanol and Whatman filter paper #42. The solid particles were finally dried, weighed and used to treat the organic waste materials, in a closed room using nitrogen gas. The synthesis of QNZVI was also done according to above details, however, just before adding iron sulfate to the solution of water and methanol, one gram of quartz sand (washed with chloridric acid and dried at 110 °C using oven) was used. The SEM (scanning electron microscoy) method was used to indicate the physical properties of NZVI (Kim et al., 2012) particles.

2.3. Experimental procedure

The samples were collected from the Recycling Center of Tehran, Iran, and after air drying and passing thorough a 0.86 μ sieve, their physical and chemical properties were determined using the standard methods (Miransari et al., 2008). The effects of NZVI type, concentration, contact time, and the organic compounds were investigated by adding 0.2 and 0.5 g NZVI to 10 g (2% and 5% w/w) of each organic compound in three replicates. The treated samples were incubated in centrifuge tubes for 1, 4, 16, 24, 48, 168, 336, 672 and 1344 h, at 25–30 °C. The tubes were sealed using parafilm, and to avoid anaerobic conditions a few holes were made in each parafilm. It must be mentioned that to avoid the quick oxidation of NZVI (with air), NZVI weighing and adding was conducted in a closed room in the presence of N_2 gas. During the incubation period the samples were regularly mixed. Finally, Pb and Ni contents were extracted (Lindsay and Norvoll, 1978) and the atomic absorption spectrometer (Model: Analytic Jena Contra AA300) was used to determine their concentrations.

2.3.1. Statistical analysis

The experiment was a four-way factorial, in three replicates, in which the effects of organic waste, NZVI type, NZVI concentration and NZVI contact time as well as their interactions were tested on the removal of Pb and Ni from the organic contaminants. Data were subjected to analysis of variance using SAS and the single effects of the treatments as well as their two-, three- and four-way interactions were examined on the removal of Pb and Ni from the waste compounds. Using SAS the sum of squares (S.S.) and the related mean squares (S.S./d.f.) were calculated, and according to the calculated F values by SAS, which are