



Research article

Natural organic matter residue as a low cost adsorbent for aluminum

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ARTICLE INFO

Article history:

Received 16 November 2017

Received in revised form

9 March 2018

Accepted 11 March 2018

Keywords:

Aluminum

Humin

Remediation

Natural substances

Adsorbent

ABSTRACT

The contamination of aquatic and terrestrial environments by potentially toxic metals is highlighted by the possible impacts that their high availability can have on the environment. Thus, the development of alternative adsorbents that can be used in the remediation of contaminated areas is of great environmental interest. Humic, one of the fractions of natural organic matter, is a promising alternative in studies on the retention of different metals that are environmentally toxic. In this study, the influence of the organic and inorganic humic constituents that are involved in the retention of aluminum species was evaluated. After extraction and calcination to obtain the ashes (inorganic constituents), humic and ash samples were structurally characterized by Fourier transform infrared spectroscopy and scanning electron microscopy. Interaction studies between aluminum-humic and ash-humic were performed in the pH range of 4.0–8.0 and with various contact times. The results of the characterization of humic and ash showed different functional groups present in the structures of these materials. Based on the results of the interaction between humic-aluminum and ash-aluminum, it can be inferred that both the organic and inorganic components of humic are efficient at absorbing aluminum. However, the adsorption isotherms showed that humic and the ashes have different adsorption behaviors. Humic is the only fraction of natural organic matter with a significant inorganic constituent content; it is the fraction least used by researchers in this field and is often discarded as waste. In light of this, the results obtained in this work highlight the importance of humic as a natural adsorbent material. Humic may be promising for the removal of aluminum species in contaminated environments due to the presence of organic and inorganic constituents.

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1. Introduction

The excessive consumption of natural resources is among the main factors affecting air, water and soil quality (Pandey et al., 2014; Nejat et al., 2015). Inadequate waste disposal and the indiscriminate disposal of industrial and domestic effluents are the major sources of exposure of different pollutants to the environment (Kapusta and Sobczyk, 2015; Qing et al., 2015; Wang et al., 2015, 2016).

Among the major environmental pollutants, potentially toxic metals (PTM) stand out because their high mobilities and bio-availabilities increase the risks associated with these compounds

(Andreas and Zhang, 2014; Bolan et al., 2014; Fu and Wang, 2011). High levels of PTM in terrestrial and aquatic environments can lead to bioaccumulation in the tissues of different organisms (Bilalis et al., 2013; Kim et al., 2015). Metals such as lead (Pb), copper (Cu), cadmium (Cd), mercury (Hg), zinc (Zn), nickel (Ni), aluminum (Al), are the main metal species related to pollution and environmental toxicity (Chamier et al., 2015; Kapusta and Sobczyk, 2015; Zhang et al., 2016).

Al is one of the most commonly used metals worldwide; approximately 57 million tons is produced annually (Bray, 2015). Consequently, the high demand for Al has triggered environmental problems directly related to its production and consumption mechanisms (Kubová et al., 2005; Liu and Müller, 2013; Waters and Webster-Brown, 2013). The main environmental impacts related to the mobilization of aluminum species are due to its high concentration in the environment. At low concentrations, they do not

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present toxic effects, but changes in pH or the content of organic matter may lead to gradual increases in their concentrations and consequently to a significant increase in their toxicity (Jaishankar et al., 2014; Waters and Webster-Brown, 2013).

In aquatic and terrestrial environments, high concentrations of available aluminum species may present potential toxicological and cumulative effects in both plants and animals at different trophic levels, including influencing their growth and development (Authman, 2011; Bilalis et al., 2013; Oberholster et al., 2012). To evaluate the effects of high concentrations of aluminum on the environment, research has been carried out to investigate alternative forms of remediation for the areas affected by high concentrations of aluminum, reducing the mobility and availability of aluminum species (Lobo-Recio et al., 2013; Lodeiro et al., 2010).

The use of natural substances such as humin for absorbing organic and inorganic contaminants is efficient (Jacundino et al., 2015; Souza et al., 2016; Wang and Xing, 2005). The adsorption processes of humin may be favored because of its high resistance to microbial degradation, since strong interactions between the organic and inorganic soil matrix provide high stability and resistance to degradation (Almendros and Sanz, 1991; Rice, 2001; Zhang and Katayama, 2012).

Studies on humin will contribute to a better understanding of its use in the remediation of different environmental contaminants (Cerqueira et al., 2012; Cunha et al., 2012). Humin is the least studied fraction of natural organic matter because of its low solubility in aqueous solutions at all pH values. Soluble fractions are commonly commercialized as natural organic fertilizers and the humin fraction is often discarded as the residue of the soluble fractions (Peña-Méndez et al., 2005; Rice, 2001; Zhang et al., 2012).

Hayes et al 2017, assumed that humin is the most abundant operationally defined class of organic substances in the terrestrial environment, and the major contributor to the sequestration of OC in soil.

The main objective of this study was to evaluate the use of humin as a low-cost adsorbent for the remediation of aluminum species. For this, the parameters related to the adsorptive capacity of humin for aluminum, as well as the influence of its organic and inorganic constituents on the adsorption processes, were studied.

2. Materials and methods

2.1. Collection of soil samples

Soil samples with high organic matter contents were collected in the district of Taquaral, in the city of Ribeirão Preto - SP, Brazil. The samples were air-dried to constant mass and passed through 2.0 mm sieves (Goveia et al., 2013; Li et al., 2015). The total organic matter content found in the soil of the present study was 29%.

2.2. Humin extraction

The initial extraction from soil samples was carried out using a solution of 0.1 mol L⁻¹ sodium hydroxide (NaOH) in a ratio of 1:10 between the soil and the extraction solvent under an inert N₂ atmosphere with mechanical agitation for 4 h. After extraction, the material was allowed to stand for approximately 24 h (Cerqueira et al., 2012; Zaccone et al., 2009). The precipitate was washed with distilled water to remove residual NaOH. After this procedure, the samples were dried at 60 °C until a constant mass was reached (Jesus et al., 2011; Souza et al., 2016; Wang and Xing, 2005).

2.3. Obtaining the inorganic constituents of humin (ash)

The ash was obtained by calcination in a porcelain crucible at

750 °C for 4 h in a muffle furnace (Griffith and Schnitzer, 1975).

2.4. Characterization

2.4.1. Fourier transform infrared spectroscopy (FTIR)

The adsorption spectra of the humin and ash samples were obtained using KBr pellets (1:100 sample/KBr (m/m)), and the spectra were acquired on a Nicolet IR 200 FT-IR spectrometer (Thermo Scientific) in the region of 400–4000 cm⁻¹ in transmission mode with 4 cm⁻¹ resolution and 32 scans (adapted from Tivet et al., 2013).

2.4.2. pH at the zero-point charge (pH_{ZPC})

To determine the pH_{ZPC} of the humin and ash surfaces, 150 mg of humin or ash was added to 50 mL of 0.1 mol L⁻¹ NaCl, and the pH values of the solutions were adjusted (2.0, 4.0, 6.0, 8.0, 10.0 and 12.0) by the addition of 0.1 mol L⁻¹ HCl or NaOH. After 24 h, the final pH of each solution was measured. The difference between the initial and final pH values was plotted, and the point of intersection between the initial and final pH values could be observed (Jia et al., 2002; Vijayakumar et al., 2012).

2.5. Study on humin/ash interactions with aluminum species

Studies were carried out on the humin and ash samples to evaluate the influence of the organic and inorganic components of humin on the interaction with aluminum species.

2.5.1. Aluminum speciation with visual MINTEQ

The determination of the aluminum species in solution was evaluated in Visual MINTEQ 3.1 software, considering different concentrations and pH values. The software allows the analysis of the solubility, speciation, complexation and sorption reactions in solid or aqueous phases (Çelen et al., 2007; Chamier et al., 2015; Gustafsson, 2016).

2.5.2. Determination of aluminum by microwave plasma atomic emission spectrometry

The aluminum concentration in solution was determined using an MP-AES 4200 microwave plasma atomic emission spectrometer (Agilent Technologies). The calibration curves for the determination of aluminum in solution were prepared from a standard solution of aluminum (1000 mg L⁻¹). The pH values of the solutions were adjusted by the addition of 0.1 mol L⁻¹ NaOH or HNO₃ depending on the pH of the solution being studied. The curves were prepared in the range of 0.5–10.0 mg L⁻¹ for samples of low Al concentration and 10.0 to 50.0 mg L⁻¹ for high-concentration samples. The readings were performed on three replicates at a wavelength of 396.152 nm (Goncalves et al., 2016; Kamala et al., 2014).

2.5.3. Study of the influence of pH on the adsorption of aluminum/humin and aluminum/ash

The influence of pH on the aluminum/humin interactions was evaluated at different pH values (4.0, 5.0, 6.0, 7.0 and 8.0) and after different contact times (5, 10, 15, 30, 60, 120, 180, 300, 600, 1440, 2880 and 4320 min). Humin/ash samples (100 mg) were combined with 50 mL of 10 mg L⁻¹ aluminum solution and subjected to mechanical agitation. After the contact period, the aluminum/humin and aluminum/ash solutions were filtered through 0.45 µm cellulose filters. The concentrations of aluminum in the filtrate were quantified by microwave plasma atomic emission spectrometry (MP-AES).

The influence of pH on aluminum adsorption by humin and ash was calculated from the mass of the adsorbed metal per mass unit

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