

Chromium Adsorption in Different Mineralogical Fractions from Subtropical Soils



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ABSTRACT

Safe application of chromium (Cr)-containing organic industrial wastes to soil requires considering the ability of the soil to adsorb Cr. In this study, the maximum Cr adsorption capacity was assessed for the bulk samples and their clay and iron-free clay fractions of four subtropical soils differing in mineralogy. To this end, the samples were supplied with Cr(III) nitrate solutions at pH 4.5 or 5.5. The results of Cr(III) adsorption fitted to a Freundlich equation and the adsorption capacity was positively correlated with soil organic matter and iron oxide contents. The clay fractions adsorbed more Cr per unit mass than the bulk soils and the iron-free clay fractions. The Cr(III) adsorption capacity increased with increasing soil pH due to more charges on adsorbing surfaces. Our results suggest that the soils rich in organic matter and iron oxides and having a pH above 4.5 are suitable for application of Cr(III)-loaded industrial wastes.

Key Words: adsorption capacity, environmental contamination, iron-free clay fractions, iron oxides, point of zero charge, specific surface area

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INTRODUCTION

Chromium (Cr) has been industrially used since its discovery in the 18th century. This metal was formerly used for its pigmentary properties; at present, it is also used in metal alloys and for electroplating, leather tanning, and wood preservation (Wilbur *et al.*, 2012). Common usage of Cr generates various wastes, which are usually disposed of by dumping in appropriate hazardous waste landfills or by addition to soil as a source of nutrients for microorganisms and plants (ABNT, 2004). Only Cr(III)-containing wastes can be used for the latter purpose owing to high mobility and toxicity of hexavalent chromium.

Application of Cr waste to agricultural soils has been scarcely explored (Ferreira *et al.*, 2003; Kray *et al.*, 2008). In fact, the oxidation state of Cr and hence its environmental contamination potential are influenced by soil physical, chemical, and mineralogical properties. For example, acid solutions (pH 2–4) con-

taining Mn(IV) oxides can easily oxidize Cr(III) to Cr(VI) (Dai *et al.*, 2009). However, López-Luna *et al.* (2009) and Trebien *et al.* (2011) found that Cr in some Mn oxide-rich soils was largely bound to organic components and its oxidation state was not affected by the presence of Mn as a result. Ferreira *et al.* (2003) used Cr-rich tannery sludge as fertilizer for soybean and corn and observed an increase in Cr content in soil but no effect on crop yield as compared to application of mineral fertilizers.

Chromium availability to plants and leaching to groundwater are influenced the Cr adsorption capacity in soil, which depends on the amount of Cr present and its oxidation state, as well as on soil physico-chemical properties (Sposito, 1984; McLean and Bledsoe, 1992; Brantley and Mellott, 2000; Ghosh *et al.*, 2013). Compared to sandy soils, clayey soils typically have more charges and specific surface area (SSA), and hence higher Cr adsorption capacity (Alcántara and Camargo, 2001; Lemke-de-Castro *et al.*, 2010; Cao *et al.*, 2011).

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Previous metal adsorption studies commonly deal with situations of environmental contamination, where some adsorbent materials are used to remove contaminants (Zhang *et al.*, 2010; Gupta *et al.*, 2013; Varga *et al.*, 2013). However, the scant available knowledge about the Cr(III) adsorption capacity of soils has so far precluded decision-making on the agricultural use of Cr-containing wastes. The primary purposes of this work were to assess the Cr adsorption capacity in the bulk samples and their clay and iron-free clay fractions of four subtropical soils and to relate it to their mineralogical and physical properties.

MATERIALS AND METHODS

Soil samples (0–20 cm depth) under native vegetation were collected in Rio Grande do Sul State, Brazil. The studied soils belonged to four subtropical soils differing in mineralogy: two Ultisols [Argissolo Vermelho distrófico arênico (PVd-1) and Argissolo Vermelho distrófico típico (PVd-2)], an Oxisol [Latosolo Vermelho distroférico típico (LVdf)], and an Inceptisol [Cambissolo Húmico aluminico típico (CHa)] (Soil Survey Staff, 2010; EMBRAPA, 2013). These soils were selected in terms of their widely ranging particle size distribution, organic matter content, and mineralogy, as well as these soil taxonomy classes representing 64% of Brazilian soils (Anjos *et al.*, 2012).

The fine fraction (< 2 mm) of the soils was analysed for pH in a 1:1 (weight:volume) soil:water suspension, organic matter (OM), clay content, and cation exchange capacity (CEC) at pH 7.0 (Tedesco *et al.*, 1995). Soil samples were digested with concentrated HNO₃ to dissolve all elements potentially becoming “environmentally available” (method 3050B; USEPA, 1996) and Cr in solution was determined by an atomic absorption spectrophotometer (AAS) (A-Analyst 200, Perkin Elmer, USA). Iron in pedogenic iron oxides (Fe_d) was extracted with dithionite-citrate-bicarbonate mixture at 80 °C (Mehra and Jackson, 1960) and that in the low-crystallinity iron oxides (basically ferrihydrite) and organic complexes (Fe_{ox}) was extracted with 0.2 mol L⁻¹ ammonium oxalate at pH 3.0 in the dark (Schwertmann, 1964). Fe_d and Fe_{ox} were also determined in the clay fraction. Dissolved Fe was quantified by AAS in all cases.

Soil samples were dispersed with 1 mol L⁻¹ NaOH and the clay fraction was collected following Stoke’s law. Soil CHa was pretreated with hydrogen peroxide to oxidize organic matter and facilitate dispersion. The clay fraction was flocculated with 0.5 mol L⁻¹ HCl and salts in the resulting sediment were removed by immer-

sion in ultrapure water with a dialysis bag. A portion of the clay fraction was freeze-dried for mineralogical analysis and another portion treated with dithionite-citrate-bicarbonate mixture at 80 °C (Mehra and Jackson, 1960) to obtain the iron-free clay fraction.

The SSA of the bulk and clay fraction samples was determined with the Brunauer, Emmett and Teller method, using N₂ as adsorbate on a Micrometrics ASAP 2010 instrument (Micrometrics, USA). The goethite (Gt) and hematite (Hm) in the clay fraction were determined by diffuse reflectance spectroscopy (DRS). Spectra in the wavelength range of 380–800 nm were acquired in 0.5-nm steps with a Cary 5000 spectrometer (Varian, USA) and smoothed every 30 points to calculate the reflectance (*R*), by using the Kubelka-Munk function of $f(K-M) = (1 - R)^2/2R$. The amplitude of the peaks for Gt at 420–450 nm and Hm at 530–570 nm in the second-derivative spectra was used to estimate the Gt/(Gt + Hm) ratio (Scheinost *et al.*, 1998). Powder X-ray diffraction (XRD) patterns were obtained at 0.05° 2θ intervals over the range 3–80° 2θ, using a counting time of 0.2 s on a Bruker D8 Advance instrument (Bruker, Germany).

Solutions containing 0, 5, 10, 15, 30, 40, 60, 80, and 120 mg L⁻¹ Cr(III) as nitrate with a pH 4.5 or 5.5 were added to the bulk soils, clay fractions, and iron-free clay fractions at a soil:solution ratio of 1:10 (weight:volume) for determination of Cr adsorption capacity. The resulting suspensions were shaken horizontally for 2 h, stored at 25 °C for 20 h, and centrifuged before measuring the Cr concentration in the supernatant by AAS. The Cr adsorption capacity was calculated from the Freundlich equation:

$$q = KC^{1/N} \quad (1)$$

where *q* is the amount of metal adsorbed (mg kg⁻¹) at an equilibrium concentration of adsorbate (*C*) (mg L⁻¹) and *K* and *N* are the coefficients related to the adsorption capacity and adsorption affinity.

Statistical analyses

Statistical analyses were performed with the software SAS for Windows. The data exhibiting significant differences at *P* < 0.05 by *F*-test were subjected to Tukey’s test at the same probability level to separate means. The data were also evaluated *via* Pearson’s correlation analysis.

RESULTS AND DISCUSSION

Soil properties

The pH of the soil samples ranged from 4.6 to 5.4,

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