



Speciation, bioaccessibility and potential risk of chromium in Amazon forest soils[☆]

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ABSTRACT

Even though the Amazon region is widely studied, there is still a gap regarding Cr exposure and its risk to human health. The objectives of this study were to 1) determine Cr concentrations in seven chemical fractions and 6 particle sizes in Amazon soils, 2) quantify hexavalent Cr (CrVI) concentrations using an alkaline extraction, 3) determine the oral and lung bioaccessible Cr, and 4) assess Cr exposure risks based on total and bioaccessible Cr in soils. The total Cr in both A (0–20 cm) and B (80–100 cm) horizons was high at 2346 and 1864 mg kg⁻¹. However, sequential extraction indicated that available Cr fraction was low compared to total Cr, with Cr in the residual fraction being the highest (74–76%). There was little difference in total Cr concentrations among particle sizes. Hexavalent Cr concentration was also low, averaging 0.72 and 2.05 mg kg⁻¹ in A and B horizon. In addition, both gastrointestinal (21–22 mg kg⁻¹) and lung (0.95–1.25 mg kg⁻¹) bioaccessible Cr were low (<1.2%). The low bioavailability of soil Cr and its uniform distribution in different particle sizes indicated that Cr was probably of geogenic origin. Exposure based on total Cr resulted in daily intake > the oral reference dose for children, but not when using CrVI or bioaccessible Cr. The data indicated that it is important to consider both Cr speciation and bioaccessibility when evaluating risk from Cr in Amazon soils.

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1. Introduction

Chromium (Cr) occurs naturally in soils and it is mostly from the parental material, averaging 54 mg kg⁻¹ worldwide (Kabata-Pendias, 2011). However, Cr concentrations in soils from ultramafic rocks can be as high as 60,000 mg kg⁻¹ (Alloway, 2013). Chromium can present as trivalent (CrIII) and hexavalent form (CrVI), with CrVI being of the most concern. Trivalent Cr is often present as Cr(OH)₃ and Cr₂O₃, which can be adsorbed onto soil surfaces or form complexes with organic matter, thus presenting low availability (da Silva et al., 2018b; Apte et al., 2006). Besides,

CrIII is an essential nutrient for humans and animals (Eastmond et al., 2008). Oxidation of CrIII to CrVI in soils can occur in the presence of O₂ and MnO₂ or due to human activity (Fendorf et al., 1992; Gress et al., 2015). Hexavalent Cr occurs as anionic species (CrO₄²⁻, HCrO₄⁻ and Cr₂O₇²⁻) and is poorly adsorbed onto soils, thereby presenting high availability (Kožuh et al., 2000). Due to its similar structure to phosphate and sulfate, CrVI is toxic and causes diseases including cancer (Costa, 2003).

Exposure to Cr causes nausea, fever, headache, and respiratory distress, with high exposure leading to serious health problem including lung cancer (Wilbur, 2000). Risk assessment is often based on total metal concentration (Cox et al., 2013). However, only a fraction of the metal is available to be absorbed by oral, respiratory or skin contact (Li et al., 2016). Various assays have been employed to evaluate metal bioavailability by simulating digestive or respiratory processes (Juhász et al., 2007; Huang et al., 2016). The simple bioaccessibility extraction test (SBET) and Gamble's solution protocols are the most employed in bioavailability studies for

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human risk assessment studies, which have been applied to soil and dust (De Miguel et al., 2012; Coufalík et al., 2016; Mendoza et al., 2017).

The Amazon region presents remarkable features including high biodiversity and forest density, extensive rivers, and upland and lowland soils. Ultisols and oxisols are the main soils in the region, which tend to have low metal concentration. However, some soils in the region from mafic and ultramafic rocks exhibit high metal concentrations (de Souza et al., 2017). During a highway construction in the Amazon forest, Brazil, symptoms such as fever, headache, body weakness and fainting were reported by local workers. Those symptoms can be associated with various sources, but probably including toxicity from heavy metals. Several studies reported Hg contamination in the Amazon basin due to its use in gold mining, thus it is the main metal studied in the region (Castilhos et al., 2015; Faial et al., 2015).

However, there is a gap regarding exposure to other metals including Cr and its risk to human health in the Amazon region. In fact, high Cr concentration (142 mg kg^{-1}) was reported in the Amazon basin area, being much greater than the background concentration of 44.8 mg kg^{-1} (dos Santos and Alleoni, 2013). Thus, the objectives of this study was to 1) determine Cr concentrations in seven chemical fractions and 6 particle sizes in Amazon soils, 2) quantify hexavalent Cr (CrVI) using an alkaline extraction, 3) determine the oral and lung bioaccessible Cr, and 4) assess Cr exposure risks based on total and bioaccessible Cr in Amazon soils.

2. Materials and methods

2.1. Soil characterization

In October of 2013, aleatory Oxisol soil samples were collected from a pristine area in the Amazon Forest, Brazil ($0^{\circ}20'21.35''\text{N}$; $66^{\circ}38'8.92''\text{W}$) (Fig. 1). Five samples of each representative horizon (A - 0–20 and B - 80–100 cm) were collected and homogenized. Soil samples were air-dried and sieved through a 2 mm sieve and

kept in closed container at room temperature.

Soil characterization was performed following Donagema et al. (2011). Briefly, pH was determined in water using a 1:2.5 soil:solution ratio. The cation exchange capacity (CEC) was calculated by summing exchangeable ions including Ca, Mg, Na, K, H and Al. Organic carbon (OC) content was determined by the Walkley-Black method (Burt, 2004). Particle size distribution was determined using the pipette method with NaOH solution being the dispersing agent (Gee and Bauder, 1986). Sand fraction was separated by passing the soil through a 0.2 mm mesh sieve, while the silt and clay fractions were separated by sedimentation.

Concentrations of As, Cd, Co, Cr, Cu, Mn, Ni, Pb, Se and Zn were determined by digesting the soil using the USEPA Method 3050B following da Silva et al. (2018a). Briefly, 1 g of soil sample was suspended in 15 mL of 1:1 nitric acid:water solution and heated at 105°C for 6 h. After cooling, 1 mL of 30% H_2O_2 was added and digested for an additional 30 min before bringing samples to a 50 mL volume with double DI water. Metal concentrations were analyzed using inductively coupled plasma mass spectrometry (ICP-MS NexIon 300x, PerkinElmer Corp., Norwalk, CT).

2.2. Chemical and size fractionation of soil Cr

Soil Cr was separated into 7 fractions following sequential extraction method of Silveira et al. (2006). They included: (E1) soluble-exchangeable, (S2) surface adsorbed, (O3) organic matter, (M4) Mn oxides, (A5) poor crystalline Fe oxides, (C6) crystalline Fe oxides, and (R7) residual fractions. Briefly, 1 g of soil was extracted using: 15 mL 0.1 M CaCl_2 , 30 mL 1 M NaOAc at pH 5, 5 mL NaOCl at pH 8.5, 30 mL 0.05 M $\text{NH}_2\text{OH}/\text{HCl}$ at pH 2, 30 mL 0.2 M oxalic acid + 0.2 M NH_4 oxalate at pH 3, 40 mL 6 M HCl and HNO_3 -HCl digestion. After each extraction, samples were centrifuged at 2000 rpm for 10 min and then filtered using Q2 filter paper (Fisher). Chromium concentration was determined by ICP-MS.

Soil Cr was also separated into 6 particle sizes including <2000, 425–2000, 250–425, 150–250, 105–150 and < 105 μm . Chromium

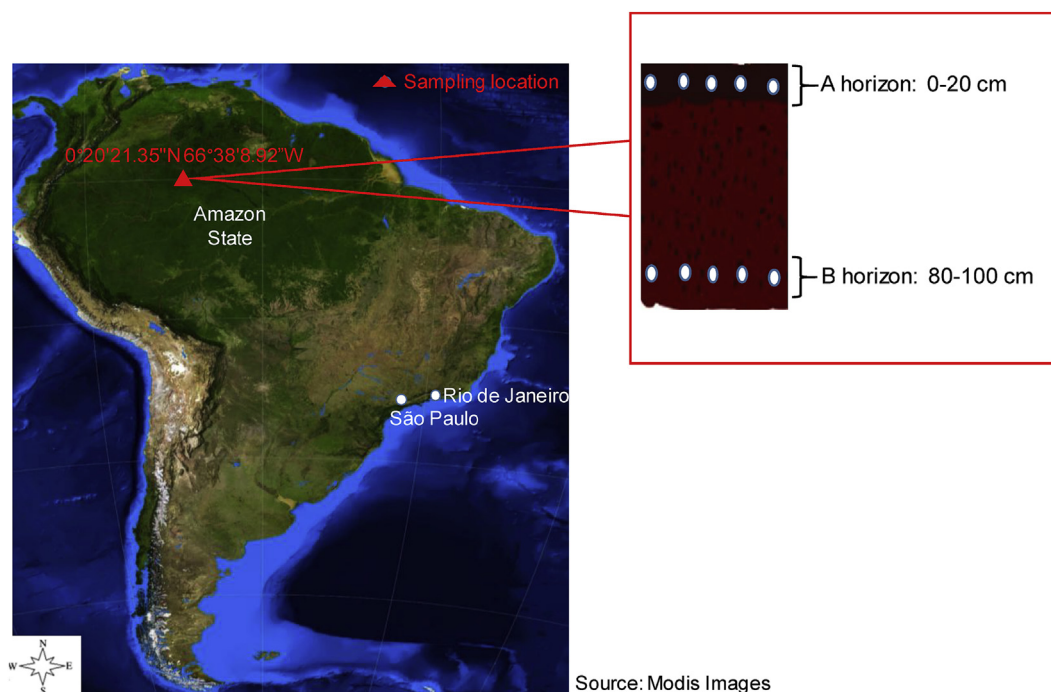


Fig. 1. Soils sampling location in the Amazon Rainforest, Brazil.

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