



Research article

Effects of clay minerals, hydroxides, and timing of dissolved organic matter addition on the competitive sorption of copper, nickel, and zinc: A column experiment



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ABSTRACT

Infiltration of heavy metal (HM) polluted wastewater can seriously compromise soil and groundwater quality. Interactions between mineral soil components (e.g. clay minerals) and dissolved organic matter (DOM) play a crucial role in determining HM mobility in soils. In this study, the influence of the timing of addition of DOM, i.e. concurrent with or prior to HMs, on HM mobility was explored in a set of continuous flow column experiments using well defined natural soil samples amended with goethite, birnessite and/or smectite. The soils were subjected to concurrent and sequential additions of solutions of DOM, and Cu, Ni and Zn. The resulting breakthrough curves were fitted with a modified dose-response model to obtain the adsorption capacity (q_0). Addition of DOM prior to HMs moderately enhanced q_0 of Cu (8–25%) compared to a control without DOM, except for the goethite amended soil that exhibited a 10% reduction due to the blocking of binding sites. Meanwhile, for both Zn and Ni sequential addition of DOM reduced q_0 by 1–36% for all tested soils due to preferential binding of Zn and Ni to mineral phases. In contrast, concurrent addition of DOM and HMs resulted in a strong increase of q_0 for all tested metals and all tested soil compositions compared to the control: 141–299% for Cu, 29–102% for Zn and 32–144% for Ni. Our study shows that when assessing the impact of soil pollution through HM containing wastewater it is crucial to take into account the presence of DOM.

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1. Introduction

Heavy metals (HMs) are considered potentially highly toxic pollutants and may pose a serious threat to environmental quality (e.g. Qin et al., 2006; Usman, 2008). Soil contamination with HMs may occur due to irrigation with contaminated water, the addition of fertilizers and metal based pesticides. Such problems are especially acute in arid developing countries such as Egypt where wastewater reuse could be a reasonable choice to mitigate the

shortage and scarcity of fresh water resources (Radwan and Salama, 2006; Alfarrar et al., 2011).

The fate and transport of HMs in soils and subsequently in groundwater aquifers are mainly controlled by the sorption capacity of soil constituents and aquifer materials (Alloway, 1995; McBride et al., 1997, 1999). Smectite and hydro-oxides minerals are known to exhibit high adsorption capacities for HMs owing to their high reactivity and large specific surface area (SSA) (Klaine et al., 2008; Hashim et al., 2011; Tang and Lo, 2013; Uddin, 2017). Several studies to date have addressed the sorption of HMs under equilibrium conditions onto different mineral surfaces (e.g., Serrano et al., 2005; Arias et al., 2006; Covelo et al., 2007; Refaey et al., 2014). However, to quantify the fate and distribution of HMs in different mineral surfaces, proper transport-related parameters of HMs in groundwater or wastewater treatment

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technology need to be evaluated (Chotpantarat et al., 2012). Column experiments have been conducted for this purpose but most of these studies were focused on a single metal (e.g., Gupta and Sharma, 2002; Liu et al., 2006; Miretzky et al., 2006; Chotpantarat et al., 2011). For instance, Abollino et al. (2008) reported that the adsorption capacity of smectite minerals for individual metal ions was 3.04, 3.61, and 3.63 mg/g for Cu, Zn and Ni, respectively. Only few studies addressed the competitive adsorption of Cu, Zn and Ni onto soil minerals under dynamic conditions. For instance, Seo et al. (2008) studied sorption of As, Cd, Cr, Cu, Hg, Pb and Zn onto sediments containing > 58% clay under dynamic conditions. This study reported that the sorption capacities of Cu and Zn were lower under competitive conditions (2.5 mg/g for Cu and 1.5 mg/g for Zn) than when sorption of individual metals was considered (13.4 mg/g for Cu and 9.3 mg/g for Zn). Likewise, Chauri et al. (2011) found that the adsorption capacity of Cu and Ni in competitive adsorption experiments were 0.033 and 0.024 mg/g, respectively.

In addition to competition between HMs, the adsorption behavior of metal ions from contaminated water is influenced by the presence of other components that may either impede or enhance adsorption and thus HM mobility. Of particular importance in the context of soil pollution with HM rich wastewater is the presence of dissolved organic matter (DOM). DOM is often found in considerable concentrations either in the wastewater itself (e.g., industrial and agricultural effluents) or in the soil (e.g. due to manuring). Such presence of DOM can exert a significant influence on the fate and transport of HMs in soil. Sorption of DOM to mineral surfaces is considered an important pathway for the retention and also the stabilization of OM (e.g., Kaiser and Guggenberger, 2000; Kalbitz et al., 2005; Mikutta et al., 2007). Therefore, the influence of DOM on HM mobility not only concerns the interaction of DOM with HMs, but also processes altering the mobility of the DOM itself in the soil. Understanding the mechanisms controlling the interactions of HMs with both mineral surfaces and DOM is therefore essential to get insight into transport and fate of metals in soils (Arshad et al., 2008; Cecchi et al., 2008). DOM was previously found to either hinder or promote HM adsorption to mineral surfaces in soils depending on the affinity of metal-ligand complexes for adsorbents (Kalbitz and Wennrich, 1998; Shuman et al., 2002; Jansen et al., 2003; Lackovic et al., 2004; Abollino et al., 2008; Refaey et al., 2014). However, most of the studies looking at the effects of DOM on the adsorption HMs by clay and hydroxide have been conducted in batch setups where DOM was in equilibrium with the HMs.

However, in a recent study employing batch experiments to investigate the competitive adsorption of Cu, Zn and Ni on several mineral phases, we discovered that the timing of the addition of DOM to soils, i.e. concurrent with or sequential to HM addition, most likely plays a significant role in regulating the mobility of HMs in soils (Refaey et al., 2014). While providing this valuable, novel insight, our previous batch study only yielded a snapshot at particular liquid to solid ratios and is unsuitable for capturing the dynamics of a realistic soil system where flow kinetics should be taken into account (Maszkowska et al., 2013). To study these dynamics a column approach is needed to enable time-dependent monitoring of contaminant leaching from soil and waste materials under more realistic environmental conditions (Maszkowska et al., 2013). As also recently acknowledged by other scientists, this knowledge gap has yet to be addressed (e.g. Chotpantarat et al., 2012; Anna et al., 2015). Addressing this knowledge gap is urgently needed in particular in the context of designing appropriate strategies for the remediation of HMs contamination in soils (Chotpantarat et al., 2012).

Following from the foregoing, the objectives of the present study were to build on the results of our recent batch adsorption

study (Refaey et al., 2014) in order to (1) unravel the effect of the timing of the addition of DOM on the competitive adsorption of Cu, Ni and Zn onto different soil compositions in a kinetic system, (2) quantify the fate and transport of metals in different mineral surfaces and (3) gain insights into leaching behavior under actual environmental conditions. For this a column approach was used in order to accommodate the dynamic characteristics of metals interaction with DOM and soil minerals.

2. Materials and methods

2.1. Sampling area and soil selection

Soils were sampled in Southern Limburg, The Netherlands at 50° 53' 37.61" N lat; 5° 53' 34.56" E long. The samples were collected from the C horizons according to their variations in soil composition (e.g. grain size, color, and organic material) after removing the A horizons on the top (up to 45 cm depth). The A horizon has a silty loam texture and is characterized by angular blocks, more sticky, brown to dark grey, and clay-rich. The A horizon has a sharp boundary to the B horizon. The B horizon (up to 75 cm depth) is yellowish brown, soft, silty, burrows traces, less blocky, less permeable, and contains brownish rich materials. The B horizon changes gradually downward to the C horizon. The C horizon is rich in Fe-oxide, has a silty loam texture, and low clay fraction content (28%).

The soil was selected based on a previous application in a different study focusing on soil-water interactions. Selection criteria were that the C horizon is of uniform grain size texture (silty texture), lacks native HMs, and is poor in native OM content. Samples of the C horizon were therefore used in the current column approach. The C horizon was extensively characterized before its application in the present study (see 2.2).

2.2. Physico-chemical and mineralogical characteristics of the studied soil

Field water content was determined by drying soil samples at 105 °C for 24 h. The soil pH_{H2O} was also measured (1:2.5 ratio). The cation exchange capacity (CEC) of soils was measured using the method of Hendershot and Duquette (1986). Major cations (Ca²⁺, Mg²⁺, K⁺, and Na⁺) and major anions (Cl⁻ and SO₄²⁻) were measured using inductively coupled plasma optical emission spectrometry ICP-OES (PerkinElmer-Optima 3000XL) and San⁺⁺ Automated Wet Chemistry Analyzer-Continuous Flow Analyzer (Skalar), respectively. Total carbon (TC) content was determined with a C/N analyzer (Elementar Vario EL). Total content of pedogenic (hydr)oxides was estimated as dithionite-citrate-bicarbonate extractable iron (Fed) (AAS, PerkinElmer) using the method of Mehra and Jackson (1960). Mn-oxide and active (oxalate extractable) Fe- and Al-(hydr)oxide (Fe_o and Al_o) contents were measured using the method of Searle and Daly (1977). For mineralogical identification, X-ray diffraction analysis was performed using a Philips (now PANalytical) PW 1830 instrument, with a Philips PW 3710 control unit (Cu Ka radiation with wavelength 1.54056 Å produced at 50 mA and 40 kV) to identify the clay minerals present in the clay fraction (Brindley and Brown, 1980; Refaey et al., 2015).

2.3. Column experiments

2.3.1. HMs solution, DOM solution, and soil preparation

Mixed solutions of chloride salts of Cu, Ni, and Zn (~25 mg L⁻¹) were used. DOM was prepared by aqueous extraction from soil with added natural manure following the method described by Refaey et al. (2014). The same source of DOM was used in our previous

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