

Arsenate adsorption at the sediment–water interface: sorption experiments and modelling

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Abstract Arsenate adsorption was studied in three clastic sediments, as a function of solution pH (4.0–9.0) and arsenate concentration. Using known mineral values, protolytic constants obtained from the literature and K_{ads} values (obtained by fitting experimental adsorption data with empirical adsorption model), the constant capacitance surface complexation model was used to explain the adsorption behavior. The experimental and modelling approaches indicate that arsenate adsorption increases with increased pH, exhibiting a maximum adsorption value before decreasing at higher pH. Per unit mass, sample S₃ (smectite–quartz/muscovite–illite sample) adsorbs more arsenate in the pH range 5–8.5, with 98% of sites occupied at pH 6. S₁ and S₂ have less adsorption capacity with maxima adsorption in the pH ranges of 6–8.5 and 4–6, respectively. The calculation of saturation indices by PHREEQC at different pH reveals that the solution was undersaturated with respect to aluminum arsenate ($\text{AlAsO}_4\cdot 2\text{H}_2\text{O}$), scorodite ($\text{FeAsO}_4\cdot 2\text{H}_2\text{O}$), brucite and silica, and supersaturated with respect to gibbsite, kaolinite, illite and montmorillonite (for S₃ sample). Increased arsenate concentration (in isotherm experiments) may not produce new solid phases, such as $\text{AlAsO}_4\cdot 2\text{H}_2\text{O}$ and/or $\text{FeAsO}_4\cdot 2\text{H}_2\text{O}$.

Keywords Sediment–water interface · Adsorption · Surface complexation model · Arsenate · Clays

Introduction

Aquifers used to supply drinking water in many countries worldwide continue to be enriched with As, causing serious health problems related to its known toxicity (Kapaj et al. 2006; Hopenhayn 2006; Singh et al. 2007). The presence of As in soils, sediments and water is attributed to natural sources, such as the weathering of minerals with high As contents, and to human activities (use of arsenical fertilizers, pesticides, industrial and mining activities) (Smedley and Kinniburgh 2002). Although anthropogenic sources have contributed to the increase of As concentration in groundwater, in global terms the natural weathering of As-bearing rocks (Guillot and Charlet 2007) is the dominant mechanism of As enrichment in drinking water reservoirs, e.g., Bangladesh, India, Vietnam, Argentina, Chile, Mexico and Brazil (Smedley and Kinniburgh 2002).

The geochemical control of As retention in sediments is important to health risk assessments and remediation strategies, because its toxicity, mobility and bioavailability are functions of its state and local chemical environment (Beaulieu and Savage 2005). Furthermore, As dynamics in soils is largely controlled by the properties (including pH, redox potential and other ions competing for sorption sites) that influence its adsorption onto mineral and colloid surfaces, such as iron, aluminum and manganese (oxy)hydroxides, and clay minerals (Beaulieu and Savage 2005). In connection with this, previous research has shown that arsenate adsorption is related to Al and Fe (oxy)hydroxides, and to the clay content of sediments (Jiang et al. 2005). Therefore, its adsorption capacity

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