An experimental and field study on P, Si, As, Cr, V and Se binding to Fe- and Al-hydroxysulfates under oxic and anoxic conditions in acidic pit lakes: chemical *vs*. microbial controls

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Introduction and scopes

The chemical composition of acidic mine pit lakes usually include trace elements which may form either oxyanions or some other anionic species, depending on pH-Eh conditions and sulfate concentration. Some elements (e.g., P, Si) are biogeochemically important, whereas others (e.g., As, Cr, V, Se) may be highly toxic to aquatic ecosystems. In the pit lakes of the Iberian Pyrite Belt (SW Spain), the fate and transport of most of these trace elements has been shown to be closely associated to the formation and (meta)stability of low-crystallinity solid phases like schwertmannite and hydrobasaluminite [1]. The present work reports recent field and experimental observations on the mobility of these elements through the water column, redoxcline and the sediment/water interfase.

Results and discussion

Schwertmannite is the most abundant mineral product of microbial Fe^{II} oxidation in the studied pit lakes (pH 2.2-3.1) [1]. Field and experimental evidence exists to support that As, Cr and P are temporaly immobilized by sorption on µm- to nm-scale schwertmannite colloids. Experimental data (including titrations under oxic and anoxic conditions) coupled to geochemical modeling, indicate that the most important retention mechanism is that of sorption of arsenate, chromate and phosphate anions to the positively charged, highly reactive schwertmannite surfaces. The process seems to be reversible, and these elements can be again released to the aqueous phase during settling to the underlying, anoxic part by either (i) microbial reductive dissolution, and/or (ii) schwertmannite aging (conversion to jarosite and/or goethite).

The behaviour of Si, V and Se appears to be more closely linked to the precipitation of hydrobasaluminite or an analogous Al phase. An evident discrepancy exists between the sorption behaviour observed under field and experimental conditions. Such discrepancy could be accounted for by recent microscopic (SEM-EDS) findings, which suggest a strong microbial control on Al precipitation and the associated sorption of Si and other elements. The role of acidophilic microbes in the Fe-Al co-precipitation at pH~4.0 represents a geochemically singular aspect which deserves further investigation.

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The density of carbonate and silicate melts in the upper mantle

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Although carbonate melts are volumetrically minor phases in the mantle, they may control the mobility of C and its residence time in the mantle, ultimately contributing to the gobal carbon cycle [1]. Carbonate melts are also considered as effective metasomatic agents because of their wetting properties, high migration rate and characteristic trace element enrichment [2]. The density of carbonate liquids is thus an important parameter to model their percolation through the mantle and evaluate their behavior as metasomatic agents and carbon reservoirs, but available data remains scarce at relevant P-T conditions and melt compositions [3,4].

In this contribution we report *in situ* invetigations of the density of carbonate liquids in the Mg-Fe binary and Mg-Fe-Ca ternary systems at upper mantle conditions (2 GPa and 1900 K). Density was determined from the X-ray absorption contrast between the samples and a diamond capsule used to contain the sample at high pressure and temperature conditions. Experiments were perfomed using a panoramic Paris-Edinbugh press at ID27 beamline of the ESRF. Pressure and temperature were determined from the X-ray diffraction patterns of hBN and Pt using the double-isochore method. The measurements provide preliminary constrains on the equation of state of carbonate liquids representative for natural carbonatites, including melt compositions produced by the partial melting of carbonated peridotites [5]. The results are combined with own recent data for the density of silicate melts (rhyolites and phonolites) and literature data for mantle minerals to discuss buoyancy relations in the upper mantle and their evolutions with pressure to better quantify the extraction of C-bearing liquids from residual rocks during partial melting and the ascent of melts through the mantle. Ultimately, we will discuss the role of carbonate-silicate liquids as metasomatic agents and carbon reservoirs.

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