Experimental study of REE behavour during apatite dissolution in presence of iron and organic matter

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The absolute concentration and distribution of Rare Earth Elements (REE) in many natural aquatic systems is controlled by 1) the dissolution/precipitation rates and 2) the solubility of phosphate minerals (c.f. Poitrasson *et al*., 2004), which themselves may be influenced by the presence of iron colloids and aqueous organic acids. Taking account of the results of Kohler *et al*. (2005) the effect of iron colloids and dissolved organic acids on the behaviour of REE in natural fluids were determined by the dissolution of natural apatite ($Ca_5(PO_4)_3F$) in the presence of a variety of selected aqueous fluids. To best replicate natural system, experiments were performed in closed-system reactors at 25°C and pH 4 and 6. After a short initial period due to the presence of REE in the initial apatite, the reactive solutions become supersaturated with respect to rhabdophane ($REE(PO_4)\cdot 6H_2O$), which buffers total REE concentration and alters the REE spectra. Complimentary synthetic lanthanum rhabdophane ($La(PO_4)\cdot 6H_2O$) dissolution/precipitation experiments were used to determine the steady-state rates of this reaction and to determine aid in the interpretation of the apatite dissolution experiments.

Results to be presented will be used to assess the role of rhabdophane precipitation and the presence of iron colloids and dissolved organic acids during the chemical evolution of surface and near-surface waters. Further analysis of these results is used to access the applicability of aqueous REE speciation models to predict the composition of REE in natural waters.

References

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Hydrochemical characteristics of surface water in two boreal granitoidic settings, Eastern Sweden

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The Baltic Sea and its terrestrial surroundings consist of a unique environment due to the brackish water and still active land uplift (up to 8-9 mm/year). Consequently, the near-coastal catchment areas are characterised by young and unweathered soils. The overall aim of this study was the increase the understanding of the chemical dynamics that control water chemistry in small natural catchments in this region, with focus on spatial and temporal trends. The work was done within the Swedish nuclear waste programme and two different areas were studied: Forsmark which has a carbonate-rich till and Laxemar with a carbonate-poor till. One of these will be selected as a deep repository for spent nuclear fuel.

Surface water (lake and stream water) were collected and analysed for major elements, nutrients and trace elements (U and REEs). The water sampling period lasted for nearly four years and continuous flow measurement were carried out over the last two years. Repetitive seasonal cycles in surface water chemistry have been observed each year along with seasonal cycles in stream discharge. The major findings were: (1) the concentrations of elements derived from rock weathering increased in stream water during flow, (2) sporadic intrusions of brackish water from the Baltic Sea resulted in strong increase in salinity (Na, Cl^-, Br^-, SO_4^{2-}), (3) a huge increase in biological activity during summer months resulted in decreased concentrations of NH_4^+, NO_3^- and Si and increased pH and concentrations of chlorophyll a, O_2, DON, POC, PON and POP, (4) the U concentrations were high at both areas and a possible source is reduced U-minerals in the overburden due to the young and unweathered soil and (5) the REE concentrations were low and characterised either by an increase in relative concentrations throughout the lanthanide series (Forsmark) or flat patterns (Simpevarp), and were by speciation modelling predicted to be carried as organic complexes.