Coupled use of Sr and Ra isotopes to assess Ra mobility and water-rock interaction in sandstone aquifers

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Sr and Ra isotopes trace water-rock interaction of divalent cations; however, $^{87}Sr/^{86}Sr$ is influenced by weathering and ion exchange whereas Ra isotopes record the balance between α-recoil and Ra sinks. These differences may allow water-rock interaction to be diagnosed more accurately than with one isotope system alone. This study investigates the anoxic, Cambrian Mt. Simon (MTS) and Jordan (JDN) sandstone aquifers, which occur across the upper midwestern USA and are utilized in areas such as the Twin Cities area in Minnesota. Use of these water resources is complicated by elevated naturally-occurring Ra.

Ra and Sr isotope ratios vary systematically between MTS and JDN, *e.g.* $^{228}Ra/^{226}Ra$ (median 1.2 and 0.5, respectively) and $^{87}Sr/^{86}Sr$ (median 0.709 and 0.711, respectively). In most of MTS, $^{228}Ra/^{226}Ra \geq 1$ is consistent with other sandstone aquifers [1], and Ra is probably generated by α-recoil and relatively inefficient adsorption. However, $^{228}Ra/^{226}Ra \leq 1$ in JDN and major ion chemistry imply a carbonate-like source that yet contradicts the radiogenic $^{87}Sr/^{86}Sr$ relative to Cambrian seawater. The Sr data exclude dissolution of primary carbonate, but do not exclude interaction with secondary carbonate. Enlarged fractures and pores [2] and secondary calcite [3] are present, at least locally, in JDN. In JDN, overall lower $^{224}Ra/^{228}Ra$ (generally <1) may also indicate that some Ra is released by dissolution and/or transported through enlarged pores. Geographic trends of $^{228}Ra/^{226}Ra$ in MTS [4] and JDN may coincide with geological and/or geochemical controls on Ra. Correlations of $^{87}Sr/^{86}Sr$ and ^{226}Ra (Spearman ρ= -0.70 and 0.44 in MTS and JDN, respectively) imply that Sr isotopes provide additional constraints on the differences between MTS and JND, and on Ra mobility in fresh waters in sandstone aquifers.

[1] Vengosh *et al.* (in press) *Environ. Sci. Technol.* doi: 10.1021/es802969r. [2] Runkel *et al.* (2006) *Sediment. Geol.* **184**, 281-304. [3] Runkel *et al*. (2003) Minn. Geol. Surv. Rep. Inv. 61. [4] Lively *et al.* (1992) Minn. Geol. Surv. Info. Circ. 36.

Coupled hydrogeological and geochemical modelling at watershed scale under sensitive tropical climate

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The small Mule Hole watershed (4.3km²) is located in the sub-humid zone of the steep climatic gradient of the Karnataka plateau, South India (Braun *et al*., 09 [1]). This transition zone is particularly sensitive to climate variations. The watershed is monitored since 2003 (ORE-BVET). The soil cover, developed on gneiss is composed of permeable red soils on the slopes and swelling clay rich black soils in the valley which contain significant amount of pedogenic carbonates. The stream is ephemeral and flows only in response to strong rainy events.

In order to investigate the weathering processes and the tranfer of chemical elements in the watershed, we use a lumped hydrogeological model coupled with a geochemical model WITCH (Goddéris *et al*., 06 [2]). This coupling enables to simulate scenarii of weathering under varying rainfall regimes. We explore the impact of a warmer global climate, as increased monsoon precipitations are expected.

The results of modelling under current rainfall are compared with field data on a three year measurment period. They show that the stream is mostly made of overland flow, so weathering flux mostly go downward to the groundwater. In the red soils, silicate weathering occurs all along the profile. In the black soils, silicate weathering only occurs within the topsoil horizon. In the underlying horizon, only pedogenic carbonates are broken down. Overall, 60% of the dissolved Ca during the rainy period re-precipitate during the dry period as carbonates (38%) or Ca-smectite (22%); the remaining 40% of the dissolved Ca is transferred toward the groundwater.

Modelling with twice the current rainfall indicates that carbonates can still precipitate in black soils during dry period as long as a Ca source is present. The CO_2 consumption by silicate weathering is expected to increase about 50% in red soils and about 200% in black soils.

[1] Braun *et al*./ *Geochimica et Cosmochimica Acta* **73** (2009) 935–961. [2] Goddéris *et al*./ *Geochimica et Cosmochimica Acta* **70** (2006) 1128-1147.