Modelling the behavior of natural U and Ra in Forsmark, Sweden

LINDSAY KRALL[13*], GIADA TREZZI[2], JORDI GARCIA-ORELLANA[2], PER ANDERSSON[3], EVA-LENA TULLBORG[4], LUIS AUQUE[5] AND DON PORCELLI[6]

[1]SKB, Stockholm, Sweden (*Lindsay.Krall@skb.se)
[2]Universitat Autònoma de Barcelona, Spain
[3]Swedish Museum of Natural History, Stockholm, Sweden
[4]Terralogica AB, Gråbo, Sweden
[5]Universidad de Zaragoza, Spain
[6]Dept. of Earth Sciences, University of Oxford, Oxford, UK

It has been proposed that the fractured crystalline bedrock of Forsmark, Sweden host a spent nuclear fuel repoistory. Early site characterizations found elevated dissolved U in groundwater[1], prompting studies on the local origin and solubility of natural U. At Forsmark, hydrothermal fluid circulation (>1Ga) is connected to U oxidation and alteration of primary (U,Th)-minerals to U(VI)-silicates haiweeite and uranophane[2].

Water-rock interaction with respect to U has been investigated using U/Th-series disequilibrium and PHREEQC geochemical modelling. Eleven groundwater samples with 0.39-150 μg/L dissolved U were measured for ^{224,223}Ra ($\tau_{1/2}$<12d) and ^{226,228}Ra ($\tau_{1/2}$<1600y) using a Radium Delayed Coincidence Counter and gamma spectrometry.

Activity ratios (ARs) of U/Th-series nuclides offer insight into the rates of processes (alpha recoil, dissolution, sorption, advection etc.) that control their groundwater distributions upon interaction with a source, i.e. local minerals and the rock surface. Most Forsmark samples are correlated by ^{222}Rn/^{226}Ra and ^{223}Ra/^{235}U ARs, indicating relations between the above processes. The sample with the highest dissolved U (150 μg/L) has anomalously high ^{235}U activities, which may indicate that a distinct set of processes and/or rates have enhanced the U-series disequilibrium. The sample with second highest dissolved U (34 μg/L) has ^{226}Ra/^{238}U and ^{223}Ra/^{235}U ARs >2 orders of magnitude higher than the 150 μg/L U sample. Speciation-solubility calculations made using PHREEQC results have suggested that both samples are undersaturated with respect to uranophane and U speciation is dominated by low-sorbing $Ca_2UO_2(CO_3)_3^0$ and $CaUO_2(CO_3)_3^{2-}$. These ARs may provide further insight into e.g. respective rates and/or timing U dissolution and adsorption at Forsmark.

[1] Smellie *et al*. (2008) *SKB* R-08-84. [2] Krall *et al*. (2015) *Applied Geochemistry*.