The influence of co-contaminant complexing agents on radionuclide environmental behaviour

MIRANDA J. KEITH-ROACH[1,2]*, ESTELA REINOSO-MASET[1], COLIN C. MAY[1], LINDSAY YOUNG[1] AND PAUL J. WORSFOLD[1]

[1]Biogeochemistry Research Centre, University of Plymouth, Plymouth, PL4 8AA, UK
[2]Kemakta Konsult, S-11293 Stockholm, Sweden
(*correspondence: mkeith-roach@plymouth.ac.uk)

Organic complexing agents that are co-disposed in radioactive waste or formed *in situ* may enhance the migration of radionuclides at contaminated sites. However, the mechanisms of the interactions are poorly understood. We have therefore undertaken an extensive program of research to probe the speciation of radionuclide-organic co-contaminant complexes, competitive interactions with common metal ions and the mechanisms by which the complexing agents impact radionuclide sorption and transport. This work has utilised electrospray ionisation mass spectrometry for the characterisation of aqueous speciation, kinetic batch sorption experiments using a relevant, characterised terrestrial sand (Drigg sand) and dynamic column experiments coupled with the k1D transport code. Thorium (IV), U (VI), Sr (II) and Cs (I) were selected as model radionuclides of different oxidation states and EDTA, NTA, picolinic acid and isosaccharinic acid were included as key complexing agents.

The complexing agents generally influenced radionuclide behaviour in the expected order of Th>U (VI)>Sr, with no discernable effect on Cs, and EDTA generally exerted the greatest influence of the complexing agents. However, the results highlighted the complexity of these interactions. In several cases, the speciation of the complexes was more diverse than suggested by the existing speciation databases. Also, the kinetics of exchange between radionuclide complexes and common metal ions varied from occurring virtually instantaneously to over several months, and could involve precipitated metal phases.

Batch experiments demonstrated that the concentration of the radionuclide strongly influences the effect that a complexing agent can have, in terms of both the kinetics of sorption and the equilibrium position. Finally, transport experiments identified that the formation of ThEDTA complexes enhances Th transport, but that at higher Th concentrations, EDTA-mediated colloidal Th transport occurs. The key data from these studies will be presented and discussed in terms of the environmental importance of the complexing agents.

Geochemical study and U/Th dating of the Akköy fissure ridge travertine (SW-Turkey): Paleoclimatic and paleoseismic interpretations

S. KELE[1]*, M. ÖZKUL[2], A. GÖKGÖZ[2], C.-C. SHEN[3], I. FÓRIZS[1], M.O. BAYKARA[2] AND M.C. ALÇIÇEK[2]

[1]Hungarian Academy of Sci., Institute for Geochemical Research, Budaörsi út 45, H-1112 Budapest, Hungary (*correspondence: keles@geochem.hu)
[2]Pamukkale Univ., Dep. of Geol. Eng., TR-20070 Denizli, Turkey
[3]National Taiwan University, Taipei, Taiwan

Fissure ridge travertines are elongated, wedge-like nonmarine carbonate deposits formed due to CO_2 degassing from carbonate rich thermal spring waters. They are well-known indicators of past and present seismic activity, including Quaternary and prehistoric major earthquake events, as generally deposited from springs discharging in co-seismic extensional fissures along major active faults [1, 2]. The precipitation of travertines is strongly influenced by the water supply controlled mainly by climate, thus the question must be raised, whether the tectonic and/or climatic processes has the major role on their deposition.

To answer the question, we performed detailed U-series dating and stable isotope and trace elemental study of travertines collected along a vertical section, through the bedded layers of the Akköy, Karakaya Hill fissure ridge (Denizli Basin, SW-Turkey). The U-series age data range from 45±108 ka to 18±0.3 ka. The more or less continuous travertine deposition at the Akköy fissure ridge during cold-dry and warm-wet climatic events indicates that the travertine deposition was controlled strongly by tectonic processes, i.e. the area of the Denizli Basin was seismically active during the period mentioned above. The trace element concentrations of the bedded travertine deposits show temporal change in the geochemistry of the travertine depositing thermal water.

[1] Hancock *et al*. (1999) *J. Struct. Geol*. **21**, 903–916.
[2] Uysal *et al*. (2009) *Chem. Geol*. **265**, 442–454.