

Biogeochemical behaviour of Pu in a contaminated soil from Aldermaston, UK

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Understanding the biogeochemical behaviour of actinides in the environment is essential for the long-term stewardship of radionuclide-contaminated land. Plutonium is of particular concern due its high radiotoxicity, long half-life and complex chemistry. Here, we investigate the biogeochemistry of Pu in a contaminated soil as microbial processes have the potential to mobilise Pu via the reduction of Pu (IV) to the potentially more mobile Pu (III). A series of microcosm experiments were designed to monitor changes in Pu solubility in an anaerobic environment stimulated via the addition of 10 mM glucose to Pu contaminated soils. A substantial shift in the 16S rRNA gene profile was observed between days 0 and 44 most notably with an increase in *Clostridia*, known glucose fermenters that have been reported to facilitate the reduction of Pu (IV) to Pu (III) [1]. A minor increase in Pu solubility was observed at day 44, decreasing to initial levels by day 118. The negligible change in Pu solubility, despite the onset of reducing conditions and formation of Fe (II), would suggest the Pu is highly refractory and unsusceptible to the influence of the surrounding geochemistry. To further examine the geochemistry of the Pu, a series of sequential extractions were performed. Around 75% of the Pu from the Aldermaston site was found to be associated with the highly refractory, residual fraction of the soils. This value greatly exceeds the fraction of residual Pu at other nuclear sites reported in the literature after similar extraction protocols (34% at Sellafield and 8% at Dounreay [2]) and further suggests the Aldermaston Pu is highly resistant to changing geochemical conditions. Further work is currently being undertaken to investigate the impact of citric acid producing fungi on Pu solubility as citric acid has the potential to mobilize Pu. This information is important for understanding both the long-term mobility of Pu in the environment and for developing remediation options for Pu-contaminated soils.

[1] Francis *et al.* (2008) *Environ. Sci. Technol* **42**, 2355–2360.

[2] Kamosa (2002) *J. Radioanal. Nucl. Chem* **252**, 121–128.

What stays in the slab and what returns to the surface? A geochemical mass balance model perspective

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We have developed the Arc Basalt Simulator (ABS), a quantitative forward model to calculate the mass balance of slab dehydration and melting, and slab fluid/melt-fluxed mantle melting, in order to quantitatively evaluate magma genesis beneath arcs. ABS models can reproduce magma compositions in many arcs.

The model suggests that the slab-derived component at volcanic fronts (VF) is mostly generated by dehydration, but successful models for most VF and all rear arc (RA) magmas also require the slab to melt. The compositions of slab fluids and melts are controlled primarily by the breakdown of amphibole and lawsonite beneath the VF and by the breakdown of phengite beneath the RA in addition to residual eclogite mineral phases including garnet, clinopyroxene, and quartz.

In the model, about 78-98% of relatively fluid-immobile elements including Nd and Hf in the arc lavas come from mantle peridotite. However, most liquid-mobile elements come from the slab. Modeled residual peridotite compositions are similar to those in some supra-subduction zone ophiolites and mantle xenoliths, providing constraints on reactions in the mantle wedge.

Altered oceanic crust (AOC) and sediment in the residual slab are modified by the subtraction of melt- and fluid-mobile elements. Unmodified AOC potentially becomes the EM I mantle component after 1 Ga, whereas melted AOC can have extremely fractionated U-Pb and become the HIMU source after 1-2 Ga. Element re-distribution beneath arcs can form the recycled materials that have been detected in ocean island basalts.