Coupling of arsenic mobility to microbial sulfate reduction in subsurface environments

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Arsenic mobilisation is an important concern in many subsurface soil, sediment and groundwater systems. In such systems, microbial sulfate reduction often dominants anaerobic electron flow, thereby leading to the production of aqueous sulfide and the formation of iron sulfide species. However, the effects of microbial sulfate reduction on arsenic mobility in subsurface environments is poorly constrained.

This presentation describes some of our recent discoveries illustrating how arsenic mobility is coupled to subsurface sulfur biogeochemistry, especially to the production of sulfide via microbial sulfate reduction.

Our approach has been to conduct a series of integrated studies on this topic, including (i) abiotic mono-mineralogical batch experiments[1], (ii) advective-flow column experiments[2] and (iii) anoxic incubation experiments with complex arsenic-contaminated floodplain soil.

The results show that microbial sulfate reduction can have contradictory consequences for arsenic; either substantially retarding or greatly enhancing arsenic mobility. These vastly divergent outcomes depend on multiple factors, including hydrological flow conditions, the presence or absence of elevated porewater sulfide concentrations, aqueous arsenic concentrations and reaction time.

The fact that sulfate reduction can both retard or enhance arsenic mobility, depending on environmental conditions, represents a serious challenge for managing arsenic-contaminated systems. In particular, it implies that great caution must be exercised when aiming to exploit natural sulfur biogeochemistry as a strategy for mitigating arsenic mobility in subsurface environments.

[1] Burton *et al.*, *Environ. Sci. Technol.* (2013), **47**, 2221-2229. [2] Burton *et al.*, *Chem. Geol.* (2013), **343**, 12-24.

Unradiogenic lead in the mantle source of Mid-Ocean Ridge Basalts

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The isotope composition of lead (Pb) in the silicate Earth appears to be far too radiogenic for evolution from chondritic (primitive solar system) material over 4.57 billion years, the so-called 'Pb paradox'. Loss of Pb to the core, storage in the lower continental crust, or arrival in a late veneer, have all been proposed as mechanisms to account for this imbalance, but each remains problematic. Recently is has been suggested that the upper mantle itself, could serve as a complementary reservoir of unradiogenic Pb, sourced in either sulphide [1] or silicate minerals [2], but it remains unclear why such material is not sampled by oceanic basalts.

This study presents high-precision double-spike Pb isotope data for the consituent phases of Mid-Ocean Ridge Basalts (glass, plagioclase and sulphide) from a single ridge segment from the FAMOUS region (36°50'N) on the Mid-Atlantic ridge. Separated phases from individual samples show a remarkable variation in Pb isotope composition, greater than that seen for all basalts previously analysed from this ridge segment. These variations cannot be explained by assimilation of seawater altered oceanic crust or by contamination from the Azores hotspot, both of which carry a radiogenic Pb isotope signature. Rather they indicate mixing between an early extremely unradiogenic melt, from which plagioclase crystallised, sourced by mantle material showing long-term depletion of U, and a later more radiogenic melt sampled by the glass. Elemental data suggest that the mantle source of this early melt was small-degree, water-rich with a garnet-like signature. Therefore, at least, some of the Pb in the Earth's mantle that is sampled by MORB is unradiogenic and complements the radiogenic composition seen in many oceanic basalts and continental crust. It remains to be seen whether variations in ^{207}Pb-^{206}Pb seen in other ridge segments reflect a similar contribution from an unradiogenic source.

[1] K.W. Burton et al. (2012) Unradiogenic lead in Earth's upper mantle. Nature Geoscience **5**, 570-573. [2] J. M. Warren, S. B. Shirey (2012) Lead and osmium isotopic constraints on the oceanic mantle from single abyssal peridotite sulfides. Earth. Planet. Sci. Letts. **59–360**, 279–293.

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