Experimental constraints on the development of Os isotopic heterogeneity in the Earth's mantle

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Owing to differences in compatibility between Re and Os, the decay of ^{187}Re to ^{187}Os is an exceptional tracer of melting in the Earth's upper mantle. There is a wide consensus that the mantle displays Os isotopic heterogeneity at the grain scale, which is exemplified by large variations of ^{187}Os/^{188}Os in oceanic basalts. The decoupling of the Re-Os isotopic system during partial melting of the mantle, is thought to be associated with alloy/sulfide equilibrium at high temperature. Alloys remain at the source and develop non-radiogenic Os isotopic ratios, as they have very low Re/Os. On the other hand, sulfides have high Re/Os and develop more radiogenic ^{187}Os/^{188}Os over time. However, not much is understood on the timing and conditions at which the de-coupling of the Re-Os isotope system takes place. Alloy model ages calculated using Os isotopes can be as old as 4.1Ga. The relative long-lived nature of these alloys implies that these phases are extraordinarily resilient to isotopic re-equilibration despite being hosted by mantle lithologies. Sulfides on the other hand, are broadly younger than alloys and are vulnerable to low-T metasomatic re-equilibration.

In order to provide constraints on the high-T stability of Os-rich alloys, we have carried out piston-cylinder experiments where variable proportions of Re, Os and Ir were equilibrated with FeS at temperatures ranging between ~1800 and ~2000K and constant pressure (15 kbar). Experiments were carried out in closed graphite capsules, with oxygen fugacity buffered near the C-CO redox buffer. Results show that Re, Os and Ir form an almost complete solid solution in the alloy at temperatures exceeding 1900K. At T lower than 1900K, a miscibility gap develops in the Re-Os-Ir-Fe-S and run products consist of a Re-rich sulfide melt, an OsIrFe alloy and a discrete Re alloy phase. We also show, that depending on the initial proportions of Re, Os and Ir in the charge, alloy-sulfide pairs will develop different ^{187}Os/^{188}Os over time.

Our results preclude equilibration of Os-rich alloys in the mantle at temperatures exceeding 1900K, as alloys would be richer in Re, which is not the case for natural alloy compositions. Furthermore, we show that alloys have to be isolated from mantle sulfides for up to several billion years, with implications to our understanding of mantle convection.

The fate of sorbed contaminants during the biogeochemical cycling of iron in natural environments

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Bacteriogenic iron oxides (BIOS) naturally occur in a wide range of environments, including wetlands, hydrothermal sea vents and hot springs. BIOS are essentially composed of neutrophilic iron-oxidizing bacteria and poorly ordered iron oxides, such as ferrihydrite. Given their high surface reactivity and surface area, they have been shown to be efficient sorbents of aqueous contaminants. The present study investigates the redox stability of naturally occurring BIOS and the fate of their sorb contaminants (As and Sr). Results indicate that BIOS samples (composed of ferrihydrite and smaller amounts of lepidocrocite and goethite) from a wetland area and gold mine tailings undergo rapid microbial reduction in the presence of a well known iron-reducing bacterium (i.e., *Schewanella putrefaciens*CN32). In fact, the reduction rates observed for the various BIOS samples far exceed those of synthetic iron oxides (ferrihydrite). The results also show that the presence of sorbed Sr and As (present as outer-sphere and inner-sphere complexes, respectively) stabilizes BIOS during microbial reduction by blocking reactive sites onto the iron oxides. Finally, the fate of Sr and As during reduction mirrors that of Fe(II), suggesting that all sorbed contaminants are likely sorbed onto the iron oxides.