

Effect of climate change-driven sea water intrusion on the mobilisation of Tc(VI) from reduced sub-surface sediments

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Nuclear fuel cycle operations have resulted in a significant legacy of contaminated land that requires long term management or remediation. Future sea level rise predictions suggest that nuclear facilities situated in coastal locations will become threatened in terms of seawater intrusion. Increased pore water salinity coupled with any influx of oxygenated seawater may impact on the stability of radionuclides associated with sub-surface sediments. Of particular concern is the high yield, redox sensitive fission product ⁹⁹Tc which, under reducing conditions, accumulates in sediments as hydrous Tc(IV)n.H₂O phases.

In this work Tc (1 μM) was reduced and sorbed with sediments under anaerobic conditions. Although nitrate has been reported to inhibit Tc reduction, in this study extensive sorption of Tc (87%) was observed during nitrate reduction. Tc reduction increased to 98% with the onset of Fe reduction.

Mobilisation of Tc(IV) from initially nitrate- and iron-reducing sediments into groundwater and seawater was then studied using batch and column experiments. Batch results showed that Tc was oxidised prior to mobilisation in groundwater and seawater. Limited release into degassed seawater suggested that the formation of soluble Tc(IV) carbonate or colloidal species was not significant. Release of Tc from the initially nitrate- and iron-reducing sediments into groundwater was broadly similar, with 36-48% mobilised after 90 days. In contrast, the initial redox status of the sediments had a marked effect on Tc mobilisation into seawater. Significantly less Tc was released from sediments with ingrowth of Fe(II) (17 ± 2%) compared with the nitrate-reducing sediments (45 ± 7%). These results suggest that Fe(II) phases are able to hinder Tc reoxidation and release into seawater. In column experiments a near instantaneous pulse (4-7%) of Tc was mobilised, followed by a slower sustained release that continued for the duration of the month long experiments, resulting in > 90% Tc release.

Reduced sediments may therefore act as a secondary source of Tc to marine environments during sea level rise-driven intrusion. Mobilisation occurs primarily via reoxidation whilst changes in pore water salinity alone appear unlikely to increase the rate and extent of Tc release.

Mercury and enstatite chondrite origins by equilibrium condensation from chondritic-IDP enriched vapor

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Mercury's core is at least 60% of its mass, compared to Earth's ~32%. The origin of Mercury's anomalous core and low FeO surface mineralogy [1] are outstanding questions in planetary science. If Mercury accreted primarily from a local annulus of precursor solids [2], then Mercury's composition may result from chemical controls on equilibrium partitioning of Fe and Si between those solids and coexisting vapor. High temperatures and enrichment in solid condensates or 'dust', relative to H-rich vapor, are likely conditions near the midplane of the inner solar protoplanetary disk. FeO-rich silicate liquids similar to the liquids quenched in ferromagnesian chondrules are thermodynamically stable in oxygen-rich systems that are highly enriched in a dust of CI-chondrite composition [3]. However, the solids surviving into the orbit of Mercury's accretion zone were probably more similar to FeO-poor, anhydrous, ice-free and organic-rich chondritic (interplanetary) dust particles (C-IDPs) [4]. Chemical systems enriched in C-IDP dust can produce condensates with atomic Fe/Si 50% above chondritic [5] for conservative estimates of C-IDP composition and enrichments of up to 1000x. These Fe/Si ratios approach that estimated for bulk Mercury. Stable minerals are FeO-poor, and include CaS and MgS. These reduced species are also found in enstatite chondrites. Disk gradients in volatile compositions of planetary and asteroidal precursors may at least partially explain Mercury's anomalous composition, as well as some meteorite parent body compositions. This model predicts low Ca/Al, and very low FeO content of Mercury's surface rocks.

[1] Robinson & Taylor (2001) *Meteor & Planet Sci* **36**, 841-847. [2] Drake & Righter (2002) *Nature* **416**, 39-44. [3] Ebel (2006) In: Lauretta & McSween (Eds.), *Meteorites and the Early Solar System II*, U. AZ, pp. 253-277. [4] Messenger *et al.* (2003) *Science* **300**, 105-108. [5] Lodders (2003) *Ap.J.* **591**, 1220-1247.