

## Colloid Facilitated Migration of Radioelements - Mechanisms, Significance, and Needed Conditions

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Colloids are fine-grained particulates (1 nm – 1 μm) present in near surface geologic materials. They result from i.) the degradation of organic detritus, ii.) mineral weathering, and iii.) various in-situ precipitation reactions. Colloidal materials enhance the sorption capacity of the solid phase when aggregated or bound to the surfaces of larger, immobile particles because their high surface area, electrostatic properties, and surface functional groups favor inorganic solute complexation. Under aqueous conditions that promote their dispersion and stability, colloidal material can facilitate the migration of radioelements that strongly sorb to them leading to more widespread dispersal. Here we describe accounts of colloid facilitated radioelement migration, generalizing controlling factors such as colloid identity and concentration; aqueous phase conditions; and contaminant sorption mechanism, strength, and reversibility. Then, the subsurface migration of <sup>137</sup>Cs will be evaluated in a high-level nuclear waste vadose zone plume at the Hanford site where alkaline, high-ionic strength <sup>137</sup>Cs –containing solutions have migrated 30 m through the subsurface, yielding an anomalous distribution profile. <sup>137</sup>Cs<sup>+</sup> is a strongly sorbing cation whose migration is facilitated by high Na<sup>+</sup> in the waste-water. Low ionic strength meteoric waters currently infiltrate the site that dilute pore-water Na<sup>+</sup> and increase adsorptive affinity and colloid migration potential. Laboratory experiments documented the occurrence of waste-induced mineral dissolution and precipitation reactions leading to the formation of fine-grained zeolitic phases including sodalite and cancrinite. These phases have comparable <sup>137</sup>Cs adsorptivity to the indigenous sediment sorbents (phyllosilicates) and exhibit surface charge characteristics conducive to particle migration. Column studies were performed with pristine and contaminated sediments that demonstrate the potential for colloid migration of a portion of the adsorbed <sup>137</sup>Cs pool. The implications of colloid migration to the existing contaminant plume and future migration are discussed.

## Evidence for Li isotope fractionation during subduction

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Compared to the mantle, subducted oceanic crust is generally heavy in Li isotope composition, potentially making Li isotopes a powerful tracer for the subduction component in island arcs. However, preliminary data (Zack et al., 2001; Goldschmidt 2001) has shown that subducted basalts (orogenic eclogites from Trescolmen, Switzerland) can be isotopically very light. The two lightest eclogites have  $\delta^7\text{Li}$  values of -8.5‰ and -9.6‰, with replicates of both samples being within the established analytical  $2\sigma$  uncertainty of 1‰. Mechanisms that can be ruled out as producing such light compositions (Zack et al., submission to EPSL) include:

(1) High-T alteration. One amphibolite-facies ocean floor basalt measured by Chan et al. (1992; EPSL 108: 151-160) is light in terms of  $\delta^7\text{Li}$ . However, Trescolmen eclogites have chemical patterns characteristic of low-T altered basalts, e.g.  $\delta^{18}\text{O}$  up to +7.5‰ (Wiesli et al., 2001; Int. Geol. Rev. 43: 95-119) and elevated Na and LILE concentrations.

(2) Fluid influx from surrounding garnet mica schists. Although there is widespread evidence for fluid exchange between eclogites and adjacent metapelites at high P conditions, this process can have only elevated  $\delta^7\text{Li}$ , as the mica schists have consistently heavier  $\delta^7\text{Li}$  (+2.4 to +3.8‰).

(3) Amphibolite-facies overprint. Eclogites show variable, although minor, signs of retrogression due to hydration by an amphibolite-facies fluid influx. Such retrogressed areas can be excluded by analysing carefully selected pure omphacite mineral separates, as omphacite contains most of the Li in eclogites. The  $\delta^7\text{Li}$  of eclogite whole rocks overlaps with that of corresponding omphacite separates within analytical uncertainty, demonstrating that post-eclogite-facies retrogression has not changed the Li isotope composition significantly.

We suggest that the very light  $\delta^7\text{Li}$  values (down to -9.6‰) in the eclogites is produced by isotopic fractionation during early devolatilization of low-T altered basalts. Here, fluid loss by Rayleigh distillation between 100-300°C efficiently releases heavy Li to the fluid while the residue is subsequently depleted in <sup>7</sup>Li. These data support previous suggestions that isotopically heavy Li is released into the forearc mantle wedge in subduction zones, while isotopically light Li is subducted deeply, forming a distinct mantle reservoir.