Using radium isotopes to determine the residence time of circulated seawater in coastal sediments

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The circulation of seawater in seafloor sediments and the resultant fluxes of water to the water column are highly important to oceanic mass balances of certain elements and to coastal water quality. The residence time of this water in the sediments is critical to the kinetics, therefore the effectiveness of water-rock interaction, which further affects the delivery of chemicals from the sub-seafloor to the ocean.

We used radium isotopes to decipher about the residence time of circulated seawater in coastal sediments. Seawater is radium-poor, with the activity of ²²⁶Ra (half time of 1605 yrs) on the order 0.1 dpm/L, and those of the short-lived isotopes, ²²⁴Ra and ²²³Ra (half lives of days), being close to zero. On the other hand, activities in pore water of both short and the longlived isotopes may be 2-3 orders of magnitude higher. Another important consideration is that while the pore water activities of the long-lived 226Ra are pretty much controlled by desorption, those of the short lived isotopes are mainly controlled by recoil during the disintegration of their thorium radioactive parents. Gonnea et al. (2008) have shown that pore water attains secular equilibrium for ²²⁶Ra within a few hours, may be less. On the other hand, since ²²⁴Ra in pore water is enriched by recoil due to the disintegration of sediments' ²²⁸Th, the activity of this nuclide may be built-up during a period of up to several weeks, which could be more relevant to the proces of seawater circulation (hours to years).

We pumped pore water using mini piezometers from depths of 0.5-2 m in the granular sediments (quartz sands) of Dor Bay (eastern Mediterranean, northern Israel). Water salinities were between 30-39 (eastern Mediterranean water salinity is ~40, psu scale). ²²⁶Ra activities were mostly 2-2.5 dpm/l, though in a few shallow cases (0.5 m), activites dropped to <1 dpm/l. Experiments showed similar ²²⁶Ra ctivities of 2-3 dpm/l, which suggests that the residence times of pore water are longer than a few hours. Activities of ²²⁴Ra varied between 1.5-6.5 dpm/l, with the deeper water (1.5-2 m) significantly more enriched than the shallow ones (0.5-1 m). In chromatography experiments with the same sediments and coastal seawater, ²²⁴Ra increased from <0.5 to 4 dpm/l at residence times of 1 and 24 hrs, respectively. Assuming uniform sediment characteristics. this suggests that the average age of pore water at 1-2 m depth is about 0.5-2 days.

Low-T hydrothermal fluid evolution

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The chemical and isotopic composition of calcium carbonate veins precipitated within the oceanic crust can be used to infer the composition of their parent fluid [1], and trends of fluid compositions with temperature [2] can be extrapolated to estimate ancient ocean water compositions [3, 4]. Previous studies focused on carbonates from drillcores in ancient oceanic crust, but ophiolites could be used to extend the record of ocean water chemistry further back in time. We evaluated the potential of ophiolites as paleoseawater archives by analysing carbonates from the Cretaceous Troodos Ophiolite of Cyprus.

Variations of fluid Sr/Ca, Mg/Ca, ⁸⁷Sr/⁸⁶Sr and ⁴⁴Ca/⁴⁰Ca with temperature (δ^{18} O) in the volcanic section of the Troodos Ophiolite display similar trends to data for fluids [2] and carbonates [3, 4] from drilled oceanic crust, indicating that similar processes acted upon the fluids. In accordance with experimental results [5] we found that the formation of saponite and palagonite, and the subsequent precipitation of anhydrite, are responsible for modifying the composition of seawater as it percolates downwards through the oceanic crust. Our data and thermodynamic calculations indicate that anhydrite may form at much lower temperatures ($\vartheta < 70^{\circ}$ C) than previously thought [6], if seawater interacts with basalt.

Our results allow not only to identify fluid evolution pathways affected by mixing of seawater with a more evolved fluid (e.g. Juan de Fuca), but also to show that ophiolites can be used to determine past seawater compositions, and to study the chemical evolution of fluids in greater detail than is possible using samples drilled from oceanic crust, where recovery of interstitial carbonates is less complete.

[1] Coggon et al. (2004) EPSL 219, 111 - 128. [2] Elderfield et al. (1999) EPSL 172, 151-165. [3] Coggon et al. (2010) Science 327, 1114-1117. [4] Rausch et al. (2013) EPSL 362, 215-224. [5] Seyfried & Mottl (1982) Geochim. Cosmochim. Acta 46, 985-1002. [6] Bischoff & Seyfried (1975) Amer. J. Sci. 278, 838-860.