Mass-dependent and -independent fractionation of Hg isotopes in lakes: Effects of smelter emissions and pH

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The stable isotope composition of Hg in cores from three lakes located 3.8, 9.4, and 21 km from a smelter [1], and hence polluted to different degrees by smelter emissions, was determined by multicollector inductively coupled plasma mass spectrometry [2]. Comparison of isotopic and chemical data for these lakes and for sediments deposited before and after the onset of pollution within each lake revealed evidence for both mass-dependent and mass-independent fractionation of Hg isotopes by natural processes [cf. 3, 4], whose effects are sensitive to smelter fallout and water chemistry. Mass-dependent fractionation is inhibited by Cu, Cd, and As from the smelter; but mass-independent fractionation, which causes selective depletion in isotopes of odd mass number (^{199}Hg and ^{201}Hg), implying a role of nuclear spin, is inhibited by Zn and Pb and is also affected by ambient pH. The mass-independent effect is greatest in sediments predating pollution by the smelter and smallest in the most polluted sediments, the difference increasing with pH over the range 4.40-7.56. The results suggest that the mass-dependent and mass-independent isotope effects are mediated by different species of bacteria which differ in their sensitivity to specific toxic pollutants and are subject to different limiting factors. Core profiles of organic matter show damped oscillations in the zone of pollution except in the most acidic lake, suggesting that smelter fallout perturbed the population dynamics of the biota, including bacteria that mediated mass-independent isotope effects. Only in the lake closest to the smelter does Hg appear to have retained the isotope signature of the source of pollution, probably because particulate Hg(II) from the smelter is concentrated there and because suppression of isotope-fractionating bacteria by toxic smelter fallout is greatest there, preventing obliteration of the signature. The signature is attributable to abiotic fractionation in the smelter.

[1] Telmer *et al.* (2006) *Geochem.: Explor. Environ. Anal.* **6,** 187-202. [2] Performed by Activation Laboratories Ltd. [3] Jackson *et al*. (2008) *Appl. Geochem.* **23**, 547-571. [4] Bergquist & Blum (2007) *Science* **318,** 417-420.

Ephemeral evaporation history of the first solids in the early Solar System

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Ca-Al-rich inclusions (CAIs) are the oldest known solids to have formed in the early solar system. They therefore bear witness to the physical and chemical conditions in the early solar system at time of their formation. The refractory mineralogy and chemical composition of CAIs suggest that they condensed out of a cooling gas of solar composition. However, the depletion of Mg and Si in Type B CAIs relative to what is predicted from equilibrium condensation suggests they experienced subsequent loss of these elements through evaporation. This evaporation should also be accompanied by mass dependent isotopic fractionation of Mg. To investigate the timescale of condensation and evaporation of CAIs in the early Solar System, we obtained stable isotopic composition of Mg (δ^{25}Mg) together with δ^{26}Mg* from the decay of the short lived ^{26}Al ($T_{1/2}$ ~0.7 Ma) in whole rock fragments and mineral separates from seven well characterized Type A and B Allende CAIs. The Mg isotopic composition of CAIs A33, A39, A43, A44A, AJEF, AM10-21-CAI-1, and AM5-63-RI-1 has been measured by MC-ICP-MS at UC Davis. The total range of δ^{25}Mg in the CAIs is -4.04 to +5.65‰. Except for CAIs AM10-21-CAI-1 and AM5-63-RI-1 with negative δ^{25}Mg, δ^{25}Mg in Type B CAIs is generally correlated with the 1/Mg ratios suggesting that fractionation of Mg isotopes and 1/Mg ratio was dominated by evaporation. Assuming CAIs started with a solar δ^{25}Mg=0 (given $\delta^{25}\text{Mg}_{\text{DSM-3}}=\delta^{25}\text{Mg}_{\text{Orgueil}}$), kinetic distillation modeling of the Mg isotopes with α=0.991 [1] shows that CAIs lost between 14-47% of its original mass. The two Type A CAIs with negative δ^{25}Mg most likely are condensates from a nebula gas already depleted in the heavier Mg isotopes. Together with the ^{26}Al-^{26}Mg isochron obtained for all our CAI samples which yields a well defined intial ^{26}Al/^{27}Al ratio of $(5.11\pm0.14)\times10^{-5}$ [2], stable Mg isotope data strongly suggests that condensation and evaporation in Allende CAIs occurred within ca. ± 20,000 years and that ^{26}Al was homogenously distributed in the solar system at the time of their formation at 4567.44 Myr ago [2].

[1] Richter *et al.* (2007) *GCA* **71**, 5544-5564. [2] Jacobsen *et al.* (2008, in review) *EPSL.*