

Anthropogenic fractionation of zinc isotopes

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Zinc (Zn) is used in many industrial processes, including the galvanization of steel and the vulcanization of rubber. Due to its relatively low volatilization temperature, Zn is emitted to the atmosphere in greater quantities than any other trace metal worldwide. The fractionation of stable Zn isotopes during anthropogenic processing provides a potential opportunity to understand and evaluate the sources, transport, and accumulation of pollutant Zn in the environment.

Here we present the results from a series of investigations of Zn isotopes in anthropogenic systems, including a new investigation of lake sediment cores from seven urban lakes and one reference lake in the USA. The time-resolved records of sediment (and Zn) accumulation suggest that, on average, the $\delta^{66}\text{Zn}$ (measured relative to JMC 3-0749L) of Zn from sediments deposited during intense urbanization is $0.09 \pm 0.07\text{‰}$ ($n = 12$), while the average isotopic signal for sediments deposited prior to urbanization (and in the non-urban reference lake) is $0.33 \pm 0.07\text{‰}$ ($n = 23$). The anthropogenic isotope signal for Zn fits well with previous investigations of Zn processing related to combustion, which may suggest that automobile emissions and waste burning are important contributors to urban Zn pollution. The natural background isotopic signal of Zn for lake sediments is similar to that reported for many igneous rocks. This may suggest that processes like weathering, transport, and biological uptake do not appreciably change the isotopic signature of the natural lake sediments.

This lake sediment study, as well as previous investigations of Zn-contaminated sediments, water, and atmospheric particles confirm that Zn isotopes can be useful to help distinguish between natural and anthropogenic Zn sources. However, because of the small amounts of fractionation and overlapping isotopic signatures, Zn isotope investigations are most successful when they are combined with other chemical and/or isotopic measurements.

Antigorite dissolution rates as a function of pH at 25 and 80 °C

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Antigorite ($\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$) is a common serpentine mineral that could provide the divalent metal cations required for mineral carbonation as part of carbon storage efforts. Such carbonation could occur via the dissolution of acidic CO_2 saturated H_2O . Antigorite dissolution into this fluid would both release aqueous Mg and neutralize the fluid promoting carbonate precipitation [c.f. 1].

In an attempt to better quantify this carbonation process antigorite dissolution rates have been measured as a function of pH from 2 to 10 temperatures of 25 and 80°C in closed system reactors. Rates were calculated from measured aqueous Mg and Si concentrations

$$r = (dC/dt)v/S$$

where r is the dissolution rate, dC/dt is the time derivative of Mg or Si concentration, v is the fluid volume in the reactor and S is the surface area.

Magnesium is released preferentially at the beginning of most of the experiments compared to silica suggesting the importance of Mg for proton exchange reactions in the antigorite dissolution mechanism. Also similar to the behavior of other Mg-silicate minerals including talc and tremolite [2], antigorite dissolution rates decrease systematically with increasing pH over the whole pH range considered in this study; antigorite dissolution rates at pH 2 are ~1.5 orders of magnitude faster than corresponding rates at pH 10. The addition of ~0.1 mol/kg citrate to the aqueous fluid appears to increase constant pH rates by less than an order of magnitude suggesting that the addition of organic ligands would be an inefficient method to accelerate the carbonation of Mg silicate minerals.

[1] Oelkers *et al.* (2008) *Elements* **4**, 333-337. [2] Saldi *et al.* (2007) *GCA* **71**, 3446-3457.