

Evidence for anthropogenic zinc in the south Pacific Ocean (GEOTRACES GP21)

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Oceanic zinc (Zn) is vital for the growth and functioning of marine primary producers. However, large gaps in our understanding of the modern marine Zn cycle remain. The light isotope composition of dissolved Zn ($\delta^{66}\text{Zn}$) is pervasive throughout most of the low-latitude upper ocean yet cannot be accounted for by biological uptake. Two very different mechanisms have been suggested to drive the low dissolved $\delta^{66}\text{Zn}$ of the upper ocean: (1) removal of heavy Zn isotopes through scavenging onto particles, or (2) addition of isotopically light Zn from atmospheric aerosols, likely anthropogenic in origin. These two processes are also reflected in the Zn isotope composition of marine particles, which allows in-depth quantification of their relative controls. However, the vast majority of oceanic $\delta^{66}\text{Zn}$ measurements were focused on the dissolved phase, with marine particles receiving much less attention.

Here, we report $\delta^{66}\text{Zn}$ in labile and refractory phases of suspended marine particles and atmospheric aerosols sampled during the GEOTRACES GP21 cruise in the south Pacific Ocean. Furthermore, $\delta^{66}\text{Zn}$ measurements are coupled with lead (Pb) isotopes, an established tracer of pollution-derived inputs to the oceans.

Zinc/phosphorus ratios in bulk upper ocean marine particles (<500 m) mostly exceed those expected for phytoplankton quotas, indicating an excess of Zn relative to biogenic material. Upper ocean labile particulate $\delta^{66}\text{Zn}$, which represents either adsorbed or external anthropogenic Zn, ranges between -0.1 and +0.3‰ (1–2441 pmol L⁻¹). These $\delta^{66}\text{Zn}$ values are lower than the dissolved $\delta^{66}\text{Zn}$ subducted into the thermocline from the Southern Ocean (~+0.4 - +0.6‰). Since scavenging is hypothesized to render the labile particulate pool isotopically heavy, the low particulate $\delta^{66}\text{Zn}$ values imply negligible Zn scavenging in upper-ocean waters advected northwards from the Southern Ocean. In addition, low particulate $\delta^{66}\text{Zn}$ is generally associated with particulate Pb isotopes bearing anthropogenic signatures, pointing to possible coupled human-sourced inputs of Zn and Pb. The results are further discussed in the context of aerosol $\delta^{66}\text{Zn}$ and Pb isotopes and elemental ratios to quantify the relative controls of biological Zn uptake, scavenging, and anthropogenic inputs, as well as to evaluate the extent of pollution-derived Zn in the south Pacific Ocean.