Alteration of iron solubility and isotopic composition of industrial particles by atmospheric processing

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Iron (Fe) plays a major role in ocean biogeochemistry and thereby the carbon cycle. Low dissolved Fe concentrations limit primary production in >30% of the surface ocean, and thus strong interest lies in constraining Fe inputs to the ocean from various sources on different spatial and temporal scales.

Large fluctuations in atmospheric deposition fluxes of continental particles (e.g., mineral dust, volcanic ash) to the ocean may have played a part in climate perturbations in Earth's history. With an increase in human activity, anthropogenic particles (e.g., from metal working, biomass burning) are now also recognised as Fe sources to the ocean, with Fe solubilities typically exceeding those of natural dusts. However, uncertainties remain in the deposition fluxes and Fe solubilities of these various materials, hindering assessment of their relative importance to ocean Fe biogeochemistry.

Distinct Fe isotopic signatures (δ^{56}Fe) reported for natural versus anthropogenic particles serve as a potential tool for tracing atmospheric Fe sources to the ocean. However, the effect of physicochemical processes (e.g., acidification, photochemistry) - that likely enhance Fe solubility in airborne particles before fallout - on their δ^{56}Fe remains unknown.

In this study, particles from an Fe-Mn alloy factory were subjected to simulated cloud processing for 1 hour in pH 4.5 perchloric acid, in the presence/absence of oxalic acid and/or solar radiation. The Fe solubility (0.1 to 7 wt.%) was highest in particles exposed to both oxalic acid and solar radiation, and the δ^{56}Fe/IRMM-014 range (-0.18 to +0.30 ± 0.01‰) indicates varying extents of fractionation relative to unprocessed particles (-0.38 ± 0.01‰). An apparent inverse relationship between Fe solubility and Fe isotope enrichment in the particles may be due to an imprint in less-dissolved samples of early preferential ^{54}Fe release and ^{56}Fe retention in the sample surface. Variable δ^{56}Fe/IRMM-014 values across experiments may also reflect an influence of different fractionation processes related to proton-promoted, ligand-controlled and reductive dissolution.

These findings as well as implications for tracing atmospheric Fe sources to the open ocean will be discussed.