Fe isotope ratios as a tracer for anthropogenic aerosol Fe sources

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Atmospheric supply of iron (Fe) to the oceans plays a key role in marine biogeochemical cycles, due to Fe's role as a limiting nutrient. Aerosol Fe comprises a mixture of both natural and anthropogenic dust, which may have very different solubilities in seawater [1]. Understanding both the relative contributions of these different types of aerosol Fe and their spatial distributions is thus important for constraining soluble Fe supply to the surface oceans.

Here, we present the Fe isotopic signature (δ^{56}Fe) of North Atlantic marine aerosol particles, collected during U.S. GEOTRACES GA03 winter cruises (Lisbon to Woods Hole, via Cape Verde, 2010-11). A range of sampling locations close to Portugal, Mauritania or North America allowed us to investigate the variability in δ^{56}Fe induced by different regional dust sources. We measured δ^{56}Fe relative to IRMM-014 in the the bulk aerosol phase (HF-HNO_3 digested), and in ultrapure water aerosol fraction (the Fe obtained with a 10 second 'instantaneous' leach with ultrapure water). Our results show that bulk aerosol Fe was characterised by near-crustal δ^{56}Fe values of +0.09±0.12‰ (1SD) with only slight variability between airmasses, pointing to the dominance of mineral dust. In contrast, the water-soluble fraction showed much more variability; aerosols collected from European and North American airmasses had remarkably isotopically light δ^{56}Fe (-0.74‰ to -1.56‰), compared to those from Saharan airmasses (+0.09‰). By comparing our data to previous model predictions of soluble Fe from mineral, industrial, and biomass burning [1], we find our data is best fit assuming a heavier δ^{56}Fe for mineral and combustion Fe (both close to +0.1‰) and a distinctly lighter δ^{56}Fe for biomass burning Fe (-1.6‰). This work demonstrates the utility of soluble aerosol δ^{56}Fe as a tracer of Fe source in marine aerosol samples, as previously suggested from bulk aerosol δ^{56}Fe [2].

[1] Luo, C. *et al.* (2008), *Glob. Biogeochem. Cycles,* **22**, GB1012.
[2] Mead, C., Herckes, P., Majestic, B. J., and Anbar, A. D. (2013), *Geophys. Res. Lett.,* **40**, 5722-5727.