

Tracing atmospheric iron supplied to the subarctic North Pacific by stable iron isotope ratios

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Deficiency of dissolved iron (Fe) limits primary production over much of the surface ocean, especially in the subarctic North Pacific. In this study Fe stable isotope ratios ($\delta^{56}\text{Fe}$) were applied in bulk and size-fractionated marine aerosol particles and dissolved Fe of surface seawater across the subarctic North Pacific on Japanese GEOTRACES cruise GP02 (Summer 2017) as a tracer to clarify the relative contribution of combustion and natural Fe in them. The bulk aerosol particles in the coastal regions of both East Asia and western North Pacific have total $\delta^{56}\text{Fe}$ values that are as low as -0.5‰ when compared to crustal ($+0.1\text{‰}$), with both the water-soluble phase and the fine particles even more fractionated (as low as -1.9 and -2.8‰ , respectively). The negative correlation between the aerosol $\delta^{56}\text{Fe}$ signatures and the enrichment factors of Fe and other elements dominated by anthropogenic sources (e.g. Pb) in these coastal regions indicates the presence of Fe emitted from high-temperature combustion sources. Combustion Fe accounts for 4–13% and 13–45% of the total and water-soluble aerosol Fe in the coastal regions, respectively. The results demonstrate that soluble aerosol Fe sourced from combustion Fe can be equivalent to that sourced from natural dust Fe in these coastal regions. By contrast, the aerosol particles in pelagic regions were near crustal $\delta^{56}\text{Fe}$ in all particle size fractions, indicating the dominance of natural Fe and little to no combustion Fe. The $\delta^{56}\text{Fe}$ of the dissolved Fe of the surface seawater does not match that of the simultaneously collected water-soluble aerosol Fe; the surface seawater has a higher $\delta^{56}\text{Fe}$ by up to $+1.5\text{‰}$. We attribute these elevated surface $\delta^{56}\text{Fe}$ values to the effect of biological uptake. New Fe fluxes from both the atmosphere and deeper depths are limited at least in summer compared with the biological uptake in the open ocean of the subarctic North Pacific.