

# Controls of iron isotope spatial variability across the Changjiang River basin: Sources, fractionation, and implications

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Iron isotopes have been increasingly used to trace the sources and biogeochemical cycling of Fe in Earth's surface environments. Despite the clear significance of riverine input of Fe to the primary productivity throughout the oceans, uncertainties still remain over the riverine Fe sources and fractionation of Fe isotopes. Here we present Fe isotope data in suspended particulate matter (SPM) and dissolved load of the Changjiang River throughout the basin from headwater to estuary.

We demonstrate that SPM carried ~99% of the Fe and have  $\delta^{56}\text{Fe}$  values (from -0.15‰ to 0.16‰) that are lower than, or similar to that of the upper continental crust. The significant correlation between  $(\text{Fe}/\text{Ca})_{\text{SPM}}$  and  $(\text{Na}/\text{Ca})_{\text{SPM}}$ ,  $(\text{K}/\text{Ca})_{\text{SPM}}$ ,  $(\text{Mg}/\text{Ca})_{\text{SPM}}$  and  $(\text{Al}/\text{Ca})_{\text{SPM}}$  confirms that clay minerals are important components of SPM. Furthermore, evidence from the Fe and Zn enrichment factors (relative to Al) and their relationships with  $\delta^{56}\text{Fe}$  in SPM reveals that Fe input from anthropogenic activities is limited. We suggest that the spatial variations of  $\delta^{56}\text{Fe}$  in SPM reflect changes in the types and intensities of weathering which controls Fe isotope fractionation during the formation of secondary phases. This interpretation is supported by the close relationship between the chemical index of alteration (CIA) value and  $\delta^{56}\text{Fe}$  in SPM. In contrast, the dissolved  $\delta^{56}\text{Fe}$  vary over a larger range from -1.51‰ to 0.20‰, whereas Fe concentrations vary over two orders of magnitude (7-146 nmol/kg). Once again, we find little evidence for an anthropogenic impact on dissolved Fe concentration and  $\delta^{56}\text{Fe}$ . Rather, the main control is the variable sequestration of heavy Fe to secondary particulate phases, likely Fe-oxyhydroxides, both in soils and in the river itself. This is consistent with the decreasing trend in dissolved Fe concentration and  $\delta^{56}\text{Fe}$  from the upper-middle reaches to lower reaches, both reaching a minimum value close to the estuary. Altogether, the new data confirm the robustness of Fe isotopes for tracing Fe sources and processes that occur during mineral dissolution, transport, and phase transformations in river systems, with key implications for the use of such an isotope approach in the fingerprint of tectonic- and climate-driven changes in continental weathering.