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Spatially resolved Fe- and S-isotope composition of sedimentary pyrite

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Past studies of iron and sulfur isotope records of sedimentary sulfides over geological time have placed important constraints on the biogeochemical cycle of sulfur and iron and the evolution of ocean chemistry. Since biogeochemical cycles of Fe and S are closely coupled in marine systems, Fe-limitation and S-limitation for pyrite formation in black shales should leave an imprint on the isotopic record of both elements. We developed a technique for accurate and spatially-resolved measurement of 34S/32S, 33S/32S, 56Fe/54Fe, and 57Fe/54Fe isotope ratios in sedimentary pyrite using a combination of solution and laser ablation analysis. Fe- and S-isotope ratios were measured by high-resolution MC-ICP-MS (ThermoElectron Neptune), enabling us to resolve major isobaric interferences on S isotopes and Fe isotopes from O_2+, ArN+, and ArO+. A CETAC LSX 213 nm laser was used as the ablation source with He as the sample carrier gas. Fe- and S-isotope ratios were calibrated against several pyrite standards using the conventional “sample-standard bracketing technique”. Instrumental mass bias of Fe and S isotopes were also corrected through an internal normalization technique using respectively Ni and Mg of known isotope composition. The long-term reproducibility of S- and Fe-isotope compositions was typically better than 0.2 per mil.

We investigated the fine scale variations of d56Fe, d34S and d33S values of diagenetic pyrite nodules in several Devonian, Paleoproterozoic and Archean black shales in order to (1) explore biosignature potential of co-variations of Fe- and S-isotopes at the grain-size scale; (2) assess potential diagenetic effects on Fe-isotope fractionation during sulfide formation; and (3) assess potential mixing between isotopically distinct Fe- and S-pools using multiple S isotope data. Those results will be presented together with bulk stratigraphic S- and Fe-isotopic variations and Fe speciation data in order to establish an Fe isotope mass balance in black shales and its relation to global environmental changes in the Precambrian oceans.