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Assessing the Alteration of Depositional Sedimentary Sulfur Isotopes Under Thermal Alteration Using Secondary Ion Mass Spectrometry

SELVA MARIANA MARROQUÍN, HANNA KONAVALUK, TYLER B. BLUM AND CHLOE BONAMICI
University of Wisconsin - Madison

The sedimentary sulfur record is critical for interpreting how marine environments have evolved throughout Earth's history as the burial of reduced sulfur species (i.e. in pyrite and kerogen) impacts global climate through drawdown of atmospheric CO_2 and buildup of O_2. Furthermore, depositional conditions, such as ocean chemistry, microbial activity, and redox conditions in deep geologic time are recorded in the pyrite sulfur isotope composition (i.e. $\delta^{34}S$). However, it remains an open area of study whether depositional $\delta^{34}S$ signals are retained after deep burial and exposure to the geothermal gradient, later stage metamorphism, and hydrothermal fluid flow. Though modelled in laboratory experiments, few field studies have examined how $\delta^{34}S$ records are impacted by post-depositional alteration at fine spatial scales. In this work, we seek to assess the response of iron sulfide $\delta^{34}S$ during high temperature diagenesis and hydrothermal fluid flow, using in-situ sulfur isotope analyses of individual sulfide grains. Samples were collected within a single stratigraphic horizon of the Upper Cretaceous Pierre Shale (Raton Basin, NM) along a 22 m transect perpendicular to a Cenozoic mafic intrusion (9 m). Here, we present Secondary Ion Mass Spectrometry (SIMS) $\delta^{34}S$ data on individual sulfide grains and additional mineralogical and lithological constraints (e.g. X-ray diffraction, total organic carbon content, and total sulfur content). The thermal influence of the mafic dike, as well as fluids evolved from the dike or through heating-induced dehydration reactions, has driven variable amounts of recrystallization of framboidal pyrite to larger pyrite grains, and the formation of pyrrhotite. Both recrystallization of primary pyrite and reaction to pyrrhotite appear systematically within the transect, along with a decrease in total organic carbon, and an increase in total sulfur content with proximity to the dike. Sulfur isotopes in samples distal and proximal to the intrusion show different patterns of sample-scale variability, but $\delta^{34}S$ values average around -33‰. Despite evidence for sample-scale reorganization of sulfur, we find that the average sedimentary sulfur isotope signatures in the studied bed of the Pierre Shale remained broadly preserved across the hydrothermal alteration halo of the intrusion, where temperatures reached, up to ~300°C.