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Deciphering the sulfur isotopic distribution of dissolved and sedimentary sulfides in sediments within and below the OMZ off central Namibia

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The stable sulfur isotope composition of sedimentary sulfides is an integral part in the paleoenvironmental reconstruction of benthic marine conditions. Yet, to date the full potential of the sulfur isotope composition has not been fully exploited because the overall sulfur isotope effect is produced by multiple microbial and geochemical processes in the sulfur cycle that interact in a complex manner. The continental shelf and slope off central Namibia is overlain by a permanent oxygen minimum zone that extends from the inner shelf to 350 m water depth. Anoxic bottom waters occur almost perennially in the innermost shelf areas. The change in bottom water oxygen concentrations across the shelf and continental slope provides an ideal opportunity for a systematic investigation of the effects of changing oxygen concentrations and sulfate reduction rates on the stable sulfur isotope composition of sedimentary sulfides. In the inner shelf environment, high dissolved sulfide concentrations and perennial benthic anoxia prevent oxidative cycling of sulfide. The resulting isotope effect is exclusively from bacterial sulfate reduction and only 4.6 ‰. Further offshore, on the central shelf, abundant large nitrate-storing sulfur bacteria catalyze up to 55 % of total sulfide oxidation and increase the isotope effect to values between 30 and 35 ‰. Outside the oxygen minimum zone, in water depths greater than 600 m, more than 95 % of the sulfide is oxidized at the sediment water interface. Sulfide recycling likely proceeds through disproportionation of sulfur intermediates. The effective sulfur isotope effect in these sediments is as high as 65 ‰ and produces iron sulfides at the sediment-water interface with an isotopic composition as low as -45 ‰ vs. VDCT. Strong ^{34}S depletion of sedimentary sulfides is therefore not indicative of euxinic water column conditions.

With burial, all sedimentary sulfides become enriched in ^{34}S due the effects of anaerobic oxidation of methane and the concomitant ^{34}S enrichment of dissolved sulfide. This alteration does not depend on the availability of reactive iron, because concentrations of sedimentary sulfides do not increase gradually with depth of burial. Apparently, sulfur atoms exchange between dissolved sulfide, acid-extractable iron sulfides, and chromium-reducible sulfide. The secondary ^{34}S-enrichment of iron sulfides formed at the sediment-water interface due to the effects of the anaerobic oxidation of methane is about 20 ‰ in the shelf sediments and 13 ‰ in the sediments on the continental slope. Detailed mass budgets of concentration and isotope composition of sedimentary sulfides allow a quantitative assessment of the extent of diagenetic overprinting in the sediments.