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Changes in sulfur concentration and sulfur isotopic composition within authigenic sulfide minerals from sediments of Miocene age to the present, ODP Site 995, Blake Ridge, offshore southeastern United States

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The Blake Ridge area (offshore southeastern United States) consists of deep-water continental margin sediments overlying deposits of methane gas hydrate. Authigenic sulfide minerals (elemental sulfur, S⁰; iron monosulfides, ~FeS; and pyrite, FeS₂) are actively deposited in these sediments due to sulfate reduction and focused anaerobic methane oxidation (AMO) at the sulfate-methane interface (SMI). Sulfide minerals at the present SMI are enriched in ³⁴S relative to those produced by sulfate reduction of organic matter higher in the sulfate reduction zone. ³⁴S enrichments preserved in the sediment column may identify relatively high amounts of AMO and thus record periods of high methane delivery to the SMI.

We extracted sulfide sulfur from sulfide minerals in sediment samples using chromium reduction (Holes A and B, Site 995, Ocean Drilling Program Leg 164). Sediment samples spanned a depth of 0 to 703.80 meters (Miocene). We made two separate sulfide sulfur extractions. The first extraction measured sulfide sulfur concentration by precipitating zinc sulfide (ZnS) and subsequently titrating dissolved sulfide via iodimetry. The second extraction precipitated sulfide sulfur as silver sulfide (Ag₂S) for sulfur isotopic analysis.

Baseline solid-phase sulfide sulfur concentrations are generally low (0.2 weight percent sulfur, dry weight) to 200 m but increase below 250 m (~0.4 wt %). Superimposed on these baseline values are peaks of sulfide sulfur concentration as high as ~1 wt %. Baseline sulfur isotopic data (δ³⁴S) generally show relative enrichments of ³⁴S between 0 and 200 m (~ -30‰), and relative depletions in ³⁴S below 250 m (~ -40‰). Some of the high concentration samples are correlative with large ³⁴S enrichments (+2.6 to 44‰) in both depth zones. These correlative peaks may represent authigenic sulfide mineral formation at the SMI, where enrichments in ³⁴S are expected. This is especially likely in samples above 200 m. Baseline shifts indicated by the sulfide sulfur data possibly represent changes in organic matter delivery over time, affecting the geochemical importance of AMO versus sulfate reduction.

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