Triple sulfur Isotope Fractionations during Sulfate Reduction in Modern Marine Sediments: New Interpretation on Sulfur Cycles

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The linear correlations between $^{33}\theta$ value (defined as $\ln^{33}\alpha / \ln^{34}\alpha$) and $1000\ln^{34}\alpha$ as a function of sulfate reduction rate, demonstrated by pure culture experiments on sulfate reducing microbes, have been increasingly used to reconstruct microbial process or ancient sulfur cycle through geological record. However, the $^{33}\theta$ vs. $1000\ln^{34}\alpha$ correlation has never been verified in modern marine environment. In this study, we applied a reaction transport model to model a triple sulfur isotope composition of pore-water sulfate profile that was collected from a typical continental margin (muddy sediment, depth ~1100m) in northern South China Sea. The data below the surface mixed layer (below 2.4m) show that the $^{34}\alpha_{net}$ value is 0.967±0.003 and $^{33}\theta_{net}$ value is 0.5176±0.0006. Active extracellular reoxidative sulfur cycles does not operate below 2.4m, as is supported by pore-water $(\Delta DIC+\Delta Ca^{2+}+\Delta Mg^{2+})/\Delta SO_4^{2-}$ ratios (<2). Hence, the $^{33}\theta_{net}$ and $^{34}\alpha_{net}$ value are characteristic of microbial dissimilatory sulfate reduction. Interestingly, the θ obtained from our field-based data is distinct from the expected $^{33}\theta$ value of 0.5110-0.5120 (with $^{34}\alpha$ about 0.967) according to the linear fits of published $^{33}\theta$ and $1000\ln^{34}\alpha$ for pure culture experiments. This study highlights the need to revisit the practice of using triple sulfur isotope relationship to reconstruct sulfur cycles in the past.