Goldschmidt 2000
September 3rd–8th, 2000
Oxford, UK.

Journal of Conference Abstracts
Volume 5(2), 1103
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Extreme Sulfur Isotope Fractionations During (Single-step?) Bacterial Sulfate Reduction in Hypersulfidic Interstitial Waters of the Great Australian Bight, ODP Leg 182

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The shelf of the Great Australian Bight has been the site of cool-water carbonate sedimentation since Eocene time, resulting in an almost 1 km thick succession, and it is now the largest area on the globe composed of such sediments. Ocean Drilling Program Leg 182 drilled several transects through this shelf with an average penetration depth of 500 m. Porewater profiles measured during Leg182 indicate that the margin of the ramp contains a complex system of possibly seawater-derived brines, reaching a maximum chlorinity of 1681 mM. In consequence, high concentrations of sulfate are observed in the interstitial waters throughout the cored intervals (up to 49 mM at 541 mbsf at Site 1129). The, for carbonate sediments, comparatively high content of organic matter (up to 0.8 wt%) triggers intense microbially mediated sulfate reduction, resulting in total alkalinity values as high as 298 mM and hydrogen sulfide concentrations of up to 13 mM. These extreme conditions are reflected in the isotope composition of the dissolved and sedimentary sulfur species. Hydrogen sulfide and sulfate show fractionation factors as high as 69 permil, which is significantly larger than the highest values reported from culture experiments of sulfate reducing bacteria. It has been suggested that such high values are the result of a multi-stage process involving the re-oxidation of hydrogen sulfide to sulfur intermediates and subsequent dis-proportionation to sulfate and hydrogen sulfide by dis-proportionating bacteria. Re-oxidation of hydrogen sulfide may be caused by a) chemical oxidation with oxygen, b) aerobic bacterial oxidation, c) anoxic chemical oxidation and d) anaerobic oxidation by chemotrophic bacteria. The possibilities (a) to (c) can be excluded, as the fractionation factor remains high to great depths (>190 mbsf) below the redox boundary. Possibility (d) appears unlikely too, because at these depths reactive iron is already converted to pyrite. Furthermore, the high concentrations of hydrogen sulfide would energetically inhibit the metabolism of sulfur-disproportionating bacteria. This suggests that the sulfur biogeochemistry in the interstitial waters of the Great Australian Bight sediments is either controlled by: a) a multistage process which involves a yet to be discovered oxidising agent and hydrogen sulfide tolerant sulfur-disproportionating bacteria, or b) a single stage process which involves sulfate reducing bacteria which cause unusually high S-isotope fractionations.