Mass-independent fractionation of sulfur isotopes for all S-bearing components of Archean sediments

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Mass-independent isotope fractionation of sulfur (MIF-S) signatures in Archean sediments have been often used to infer the presence of an O_2-poor atmosphere, as UV photolysis of atmospheric SO_2 can produce MIF-S in the absence of an ozone layer [e.g. 1]. However, recent theoretical [2] and experimental [3,4] studies have shown that non-photochemical processes can also produce MIF-S. Hence, key factors controlling the production of MIF-S signatures in Archean sediments remain unclear. In this study, we performed δ^{34}S-δ^{33}S analyses of S-bearing fractions and bulk samples of ~2.7 Ga black shales from drill core from the Jeerinah Formation, Western Australia. Elemental sulfur (S^0), sulfate (water-soluble and HCl-soluble fractions), pyrite and organic-bound sulfur (kerogen-S) fractions were obtained by solvent extraction and gravity separation,.

A slope of ~1 is observed for many bulk samples in a plot of δ^{33}S vs. δ^{34}S, but also for the S-bearing fractions (Fig. 1). The S-bearing fractions show large intra-sample heterogeneity (up to 8‰ for δ^{34}S), and positive Δ^{33}S values for all fractions (up to 6‰). The presence of MIF-S for all S-bearing fractions indicates multiple sulfur sources thr-oughout the Arch-ean sulfur biogeo-chemical cycles, and suggest that possible modern weathering has had little impact upon S-isotope signatures.

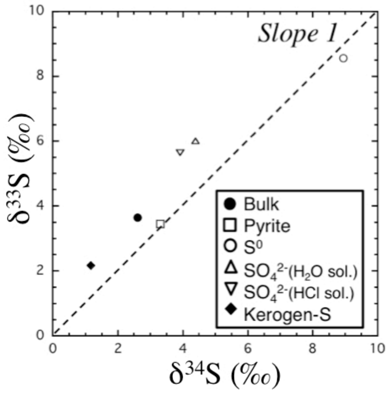

Figure 1. Intra-sample δ^{34}S-δ^{33}S heterogeneity in Archean sediment.

[1] S. Ono *et al.*, *EPSL* **213**, 15 (2003) [2] A.C. Lasaga *et al.*, *EPSL* **268**, 225 (2008) [3] Y. Watanabe *et al.*, *Science* **324**, 370 (2009) [4] H. Oduro *et al.*, *PNAS* **108**, 17635 (2011).

Use of homogenized sediment in experimental set up: Re-stabilization of redox fronts and artifacts due to sieving methods

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In experimental microcosms where the addition of marine sediments is needed to mimic natural benthic systems, sieved and homogenized natural sediment is often used (e.g.[1]), but the consequences of sieving methods on early diagenetic processes and their possible effects on experimental results are often neglected.

Our experiment investigated the effects of two different sieving methods on organic matter concentration and mineralization, exchanges at water/sediment interface and geochemical steady state reaching times for the main chemical species (O_2, Fe^{2+}, PO_4^{2-}, NH_4^+, ΣNO_3, H_2S, SO_4^{2-}).

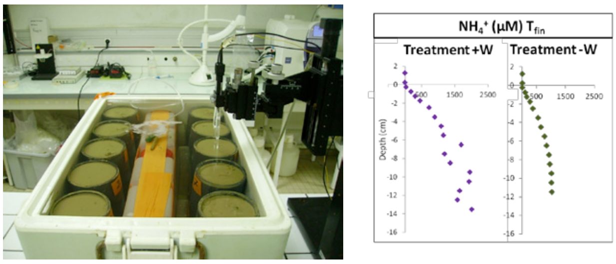

Figure 1 Experimental set up

One set of sediment samples was sieved (<38μm) with natural seawater (+W). The other was sieved without additional water (-W). The results show that while oxygen penetration depth in both treatments is stable within 7 days, the other chemical species show higher concentrations and longer time to reach steady state after the -W sieving treatment compared to the other sieving method. This is suggested to be due to the removal of organic matter during the +W treatment.

[1] Koho *et al.* (2011), *FEMS Microbiol. Ecol.* **75**, 273–283.

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