Reviewer’s Comments

This manuscript presents a valuable microscale isotope dataset from deep fracture-hosted barite-pyrite-calcite assemblages in the Fennoscandian Shield. The authors combine new SIMS $\delta^{34}S$ and $\delta^{18}O$ analyses of barite with previously published $\delta^{34}S$ data from pyrite and $\delta^{13}C$–$\delta^{18}O$ data from calcite to explore microbial sulfate reduction, sulfate-dependent anaerobic oxidation of methane, and organoclastic sulfate reduction in the ancient deep biosphere. The analytical work appears careful, the supplementary dataset is extensive, and the topic is well suited to Biogeosciences. The study provides a useful contribution by showing how barite $\delta^{18}O$–$\delta^{34}S$ systematics, when considered together with calcite and pyrite isotope records, may help distinguish different microbial sulfur-cycling regimes in deep subsurface fracture systems. I find the manuscript publishable after minor revision, mainly requiring clarification of several interpretive and statistical points.

General comments

The main strength of the manuscript is the integration of multiple microscale isotope systems across barite, pyrite and calcite. This approach has clear potential for improving the interpretation of biosignatures in ancient deep subsurface fracture systems. My main concern is not the quality of the isotope data, but the extent to which some interpretations are developed beyond what the current evidence directly constrains. In particular, the two-group Gaussian Mixture Model classification, the interpretation of Group 1 and Group 2 as dominantly associated with AOM and OSR, the inference of different sulfate reduction rates, and the proposed preservation bias against extremely ^{34}S-enriched barite all need to be presented with greater transparency. The manuscript would be strengthened by distinguishing more clearly between direct observations, model-dependent interpretations and speculative but plausible explanations.

Specific comments

1. Statistical support for the two-group GMM classification should be made more transparent.

The two-group division of barite $\delta^{18}O$–$\delta^{34}S$ data is central to the manuscript. However, the Methods state that the Bayesian Information Criterion indicated three components as the optimal GMM solution, whereas the authors selected two components because the three-component model produced negative regression slopes and because two components are consistent with the expected AOM–OSR end-member framework. This choice may be reasonable, but it requires clearer justification.

Please report the BIC values for one-, two-, three- and preferably four-component models. The rejected three-component solution should be shown in the Supplement, together with a short explanation of why negative slopes are considered geochemically implausible rather than reflecting mixed growth generations, local fluid mixing, non-coeval sulfate reservoirs, or sampling heterogeneity. I also recommend a sensitivity test showing whether the two groups remain robust when Forsmark and Laxemar are treated separately, or when individual crystals/samples rather than individual SIMS spots are used as the statistical unit. Because multiple SIMS spots may come from the same crystal or fracture, they are not fully independent observations; this should be considered when evaluating the robustness of the clustering and regression results.

2. Quantify the basis for the AOM- and OSR-dominated interpretations.

The wording that Group 1 was “dominantly associated” with AOM and Group 2 was “dominated” by OSR is reasonable. However, because Group 1 also includes barite associated with less strongly ^{13}C-depleted calcite, and because OSR-related barite appears to occur in both groups, the basis for “dominantly” should be stated more explicitly.

I suggest adding a short summary table or a few sentences indicating how many samples or analyses in each group are associated with strongly ^{13}C-depleted calcite, moderately depleted calcite, and non-depleted calcite. It would also be useful to distinguish cases where the calcite is petrographically co-genetic with barite from cases where average δ^{13}Ccalcite values from the barite-bearing sample were used. This would strengthen the process-level interpretation without requiring any change to the main conclusion.

3. Present the preservation-bias interpretation more cautiously or support it with a simple calculation.

The proposed explanation that extremely ^{34}S-enriched barite is absent because barite becomes undersaturated during advanced Rayleigh distillation is interesting. However, the current discussion is mostly qualitative. I encourage the authors either to provide a simple first-order calculation showing how sulfate concentration, Ba^{2+} availability and barite saturation state may evolve during Rayleigh distillation, or to slightly soften the wording of this conclusion.

In particular, the maximum observed δ^{34}Sbarite value of approximately +52‰ should be presented as the highest value recorded in the dataset rather than as a demonstrated saturation threshold. If no quantitative saturation calculation is added, the manuscript should more clearly acknowledge alternative explanations, such as temporal decoupling between pyrite and barite formation, limited Ba availability during late-stage sulfate reduction, post-formation dissolution, open-system mixing, or sampling bias.

4. Clarify the inference of sulfate reduction rates and sulfate/electron-donor controls.

The interpretation that differences in $\delta^{18}O/\delta^{34}S$ slopes reflect variations in sulfate reduction rates, fractionation and sulfate/electron-donor ratios is reasonable and consistent with previous conceptual models. However, the manuscript would benefit from a clearer distinction between directly observed isotope patterns and inferred microbial or hydrogeochemical controls.

For example, higher sulfate availability does not necessarily imply higher sulfate reduction rates if electron donors are limited. Similarly, apparent $^{34}\varepsilon$, sulfate concentration and cell-specific sulfate reduction rate are related but not interchangeable. I suggest revising this section to state more explicitly that SRR differences are inferred from isotope systematics rather than independently measured. This small clarification would improve the logical flow of the discussion.

5. The comparison with Antler et al. and marine sedimentary systems should be expanded.

The manuscript notes an important discrepancy: the AOM-influenced Group 1 has a steeper $\delta^{18}O/\delta^{34}S$ trend than the OSR-dominated Group 2, and the Group 2 trend is shallower than OSR-related trends reported from marine sedimentary systems. This difference is important because it supports one of the manuscript’s broader implications, namely that isotope interpretation frameworks developed for marine barite cannot be directly transferred to deep subsurface fracture-hosted barite.

This point deserves a more mechanistic discussion. The authors should compare the deep fracture system with marine SMTZ settings in terms of sulfate concentration, Ba availability, electron-donor limitation, fluid residence time, openness of the sulfate reservoir, temperature, and barite saturation state. A short comparative table would be useful. Such a comparison would help readers understand whether the observed $\delta^{18}O$–$\delta^{34}S$ trends are a predictable consequence of low-sulfate deep subsurface environments, or whether they require modification of existing conceptual models for sulfate oxygen and sulfur isotope evolution during microbial sulfate reduction.

Minor comments

1. Please check isotope notation throughout the manuscript. Symbols such as $\delta^{34}S$, $\delta^{18}O$ and $^{34}\varepsilon$ should be formatted consistently in the abstract, main text, tables, figures and captions.
2. Figure 4 would benefit from uncertainty envelopes around the regression lines. The caption should also clarify whether each plotted point represents one SIMS spot with paired $\delta^{34}S$–$\delta^{18}O$ data, one crystal, or one sample.
3. In the caption of Figure 5, two panels appear to be referred to as "(d)". Please correct the panel labeling.
4. The BSE images in Figures 5 and 6 are useful but would be easier to interpret with clearer arrows or labels marking the key petrographic relationships discussed in the text. For the interpolated $\delta^{34}S$ heatmap in Figure 6, please clarify that the interpolation is illustrative and should not be over-interpreted beyond the density of SIMS analyses.
5. Reference list formatting: The reference list should be revised carefully for consistency with the Biogeosciences style. In particular, please standardize journal-name formatting, as some journals are written in full whereas others are abbreviated. Please also check article-title capitalization, as some titles appear to use inconsistent capitalization styles. In addition, article titles

containing isotope notation and chemical formulas should be checked for correct superscript/subscript formatting, for example $\delta^{34}S$, CH_4, and H_2S, rather than plain-text forms such as δ34S, CH4 or H2S.