

Interactive comment on “Dynamic interactions between iron and sulfur cycles from Arctic methane seeps” by Pauline Latour et al.

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We appreciate the comments and feedbacks from Prof. Dr. Sabine Kasten, which we find them very helpful. We agree with most of the comments from Dr. Kasten and will gladly revise the manuscript accordingly. Similar to what we responded to reviewer #1, we would like to point out that one of the greatest challenges is to have enough constraints for the model results, especially in the case of pyrite formation. We are aware that our current model has only a simplified reaction pathway. However, this is mostly due to the limited constraints we have; i.e., we have only porewater sulfide and Fe(II) concentrations and CRS abundance to constrain three different processes in the current model. We feel like this is perhaps the optimal setup we can have at the moment. Future improvement is possible with constraints from the isotopic signatures

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of these porewater species and information from other intermediate sulfur species. Here we reply to the major concerns of the reviewer upfront and provide detailed point-to-point explanations later.

(1) The formation pathway of pyrite: We reply to the same question from reviewer #1 as follow. The assigned reaction pathway in our model is inspired by Rickard and Luther (2007), the same literature as the reviewer #1 suggested. To better justify our choice, we summarize some of the conclusions from the paper. Rickard and Luther (2007) reviewed the processes proposed in the literatures about pyrite formation in section 7.4. The most often mentioned pathway: $\text{FeSm (the amorphous iron sulfide form)} + \text{S(0)} \rightarrow \text{FeS}_2\text{p}$ (Berner et al.) was concluded by the authors to be "could not describe a mechanism since S(0) is in the form of S₈, which would make this an impossible multi-molecular reaction step". Rickard and Luther also concluded that "Certainly, it has been unequivocally demonstrated experimentally and in natural systems that FeSm does not "transform to pyrite" in the sense of a solid-state transformation. FeSm, where it occurs, dissolves, and pyrite forms from the reaction between dissolved iron and sulfur species to which the products of the FeSm dissolution reaction contribute". This conclusion justifies our choice not to involve the amorphous iron sulfide phase in the reaction network. Rickard and Luther (2007) suggested that the Bunsen reaction (polysulfide controlled) and Berzelius reaction (sulfide controlled) are two more likely pathways, in which these reactions involve an aqueous FeS intermediate (Eq, 45, 46, and 51 in the paper). More importantly, both pathways produce H₂ as a by-product. Rickard and Luther (2007) also pointed out these two pathway can be fast under low-temperature condition with the presence of certain microbes operating sulfur disproportionation. As the formation of this aqueous intermediate, FeS, is not a rate-limiting step of the overall reaction, it is adequate to use the reaction we assigned in the model. We also include some newly-obtained d₃₄S data from both CRS and AVS to shown that the two sulfur fractions underwent very different history and may use different pools of hydrogen sulfide when forming. We feel that these new data included will also help justify our choice of pyrite formation pathways.

(2) The contribution from organic matter: We entirely agree with the reviewer's opinion that the deposition of organic matter will also have large impact on the Fe cycle. In the original manuscript, we discussed briefly this affect by comparing two sites with different organic matter degradation pathways (Fig. 9). We are aware that this may not be obvious for the readers so we will extend this discussion and focus a bit more on this aspect as the reviewer requested.

(3) The application of pyrite abundance as paleo-SMT indicator: Thanks for the suggestions from Dr. Kasten, we will revise the literatures we cited in the discussion of sulfur record in the Black Sea. We will also soften our criticism about the application of pyrite as paleo-SMT indicator. We agree such application may work in other locations. However, based on the data we presented for some of our sites, we see that pyrite abundance is not necessary applicable for this purpose at least for our study sites.

Please see our point-to-point reply in the attached file.

Please also note the supplement to this comment:

<https://www.biogeosciences-discuss.net/bg-2018-223/bg-2018-223-AC2-supplement.pdf>

Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2018-223>, 2018.

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