Our response to the comments of the Reviewer 1

(1) Although many modifications have been made, this new version of the manuscript maintains a biased approach in the discussion and presentation of the data. CapD33S values are interpreted considering 'sizeable' or 'meaningful' values without any reasonable scientific basis. This concern was already raised in the previous version of the manuscript, but it has not been properly addressed. Terms of comparison are not used, so that when it is said 'small' or 'big CapD33S values' the meaning of the sentence results misleading.

As we clearly stated in the manuscript, we use the updated S-MIF definition by LaFlamme et al. (2018) according to the reviewer's suggestion. We do not use the unclear terms 'sizeable' or 'meaningful' in the revised manuscript. Also, we use the terms of comparison in the revised manuscript as the reviewer suggested to avoid vagueness. In most cases in the Result and Discussion sections, we give numerical values for comparison of different populations.

(2) In the discussion, processes imparting variation in S isotope signatures are evaluated in a superficial way. For example, decrease in d34S is associated with BSR since some CapD33S values are negative. The same patter could have been caused by many other processes such as TSR, or precipitation at variable pH-Eh conditions. To argue that BSR played a role, more convincing evidence, such as the presence of biological remains, should have been provided.

We admit that MSR is only one of several possible processes of seawater sulfate reduction. Both biotic and abiotic reduction scenarios are possible as the reviewer suggested and it is difficult to constrain reduction processes in the older Onverwacht and Fig Tree sediments. What we would like to suggest here is that, regardless of its reduction process, the seawater sulfate was likely a sulfur source of the observed rounded pyrite grains with negative $\Delta^{33}S$ values. Our former description was inappropriate and we modified the sentences to "Thus, seawater sulfate was likely a sulfur source of the observed rounded pyrite grains with negative $\Delta^{33}S$ values. The seawater sulfate may have been reduced and the produced sulfide was added to the older Onverwacht and Fig Tree sediments. Both biotic and abiotic processes are possible mechanisms accounting for sulfate reduction in these older sediments. For example, MSR may have occurred during the deposition of older source sediments. The sulfate may have been reduced with $^{34}S/^{32}S$ fractionation factors in the range of 0-10‰ at low sulfate concentration (<200 μM) in the Paleoarchean oceans [44]. Abiotic thermochemical sulfate reduction (TSR) is another possible candidate for seawater sulfate reduction ([45,46]). Within the sediments after deposition, interstitial sulfate may have been reduced by organic matter or Fe^{2+} at temperatures over 100◦C, with $^{34}S/^{32}S$ fractionation up to 20‰ [47]. Although it is difficult to constrain the original reduction processes of seawater sulfate leading to the negative $\Delta^{33}S$ signals observed in the rounded Moodies pyrite, several previous studies suggested the involvement of MSR for some pyrite in the older Onverwacht and Fig Tree groups [48-51] (Fig. 6). It is consistent with our interpretation that the Paleoarchean seawater sulfate was a sulfur source of the rounded pyrite with negative $\Delta^{33}S$ values in the Moodies." (new line no.: 321-335). We also revised Figure 6.

(3) Another example of superficial interpretation is the sentence in L.427-428. "fast-accumulated sediments may have substantially diluted sulfur derived from an atmospheric source during their deposition." There are no references in support, no is process explained or model provided. This appear to be a pure speculation.

We clearly stated that "our argument of a fast Moodies sedimentation and its influence on the sulfur records is rather an implication than our main conclusion" in our former response to the comment (4) from the Reviewer 2. We admit that our interpretation is one of several possible scenarios without strong evidence for it. That is why we substantially modified and weakened our description of the argument in the former revised manuscript, although it was apparently not enough. Hence, we added a sentence in the re-revised manuscript to show that our interpretation is an implication so far and the relationship between fast sedimentation and the sulfur isotopic record of the Moodies should be checked in more detail (new line no.: 441-449).

(4) Also, previous comments have not been addressed properly. For example, when I pointed out that: "(17) L.341: "Archean sediments are generally characterized by sizeable S-MIF". This statement is too generic and not backed by a statistical analysis. Firstly, it is not clear what represents a "sizeable" S-MIF signature. Also, according to the data of the Global Sedimentary Sulfur Isotope Database provided by Selvaraja et al. (2017; http://www.cet.edu.au/research-projects/special-projects/gssid-global-sedimentary-sulfur-isotope-database) 2746 analyses over a total of 4776 (of Archean sediments older than 2.4Ga) have a CapD33S between +1 and -1‰, which has been considered in this manuscript as "unclear S-MIF". I recommend the authors to access the data from Selvaraja et al. (2017), revise, and discuss more in detail this statement." The answer from the authors has been: "New L.415: See our responses to general comments (1)

by the reviewer. The reviewer's major concern about this statement is due to our unclear definition of S-MIF in the former manuscript. In the revised manuscript, we followed the S-MIF thresholds proposed in LaFlamme et al. (2018). We judged that the present statement is unnecessary here and thus removed it.''' No, my concern here was not the threshold definition, but about your interpretation of sizeable MIF-S, which, although no more contained in this sentence, it is still utilised as a recurring concept in the manuscript. I find this answer inadequate and dismissive.

We apologize that our former reply was inadequate and did not answer correctly the reviewer's comment. In order to examine the significance of the present Moodies sulfur records, we statistically compared the Moodies data set with older Paleoarchean records according to the data from Selvaraja et al. (2017). t-tests confirmed that, in many cases, the mean $\Delta^{33}S$ value of the Moodies data set is significantly different from the mean value of the data set of other formations in the Paleoarchean. For example, the mean $\Delta^{33}S$ value of the Moodies SIMS data set (+0.06‰) is significantly smaller than the mean $\Delta^{33}S$ value of sulfide from the Kromberg Formation (3.34 Ga) in the Onverwacht Group (+0.48‰) (p-value < 0.05%). We added the sentences: "Based on the global database of previously-published Paleoarchean sulfur isotope records [58], statistic t-tests demonstrates that the $\Delta^{33}S$ values of Moodies sulfides are significantly lower than the values of sulfide of most of the Onverwacht and Fig Tree groups. For example, the average $\Delta^{33}S$ value of the present Moodies SIMS data set (+0.06‰, n=130) is significantly lower than the average value of a SIMS data set from the 3.34 Ga Kromberg Formation of the Onverwacht Group [48] (+0.74‰, n=49) (p-value < 0.05%)." (new line no.: 427-432). We also accordingly modified Abstract to emphasize this point.

(5) Other comments below. In reference to the responses to the reviewer 1: (1) LaFlamme et al. (2018) MIF-S thresholds have been included, but Farquhar and Wing's (2003) are still used. This makes the figures and the data interpretation confusing. Remove Farquhar and Wing (2003) thresholds, or justify them under a physical point of view.

Revised as suggested. We removed the Farquhar and Wing (2003) thresholds from Figure 6 to avoid confusion.

Comments on the new version of manuscript:

(6) *L.25 - "a typical Archean S-MIF signal"*

It is unclear what a typical MIF-S signal is supposed to be. From the Global Sedimentary Sulfur Isotope Database (Selvaraja et al., 2017) the CapD33S median values of sediments older than 2.6Ga is +0.16 per mill, and the mean is 0.64 per mill. Are this the values which are considered as typical?

We admit that the term "typical" is unclear as the reviewer suggested and is not suitable to be used here. Thus we removed this term and revised the sentence to be clearer (new line no.: 23-26).

(7) *L.27-29 and 427-428 - "Fast* accumulation (~0.1-1 mm/year) of the Moodies sediments might have substantially diluted sulfur derived from an atmospheric source during their deposition [...]"

It is unclear how the rate of sedimentation could have affected S-isotope signatures. There are no references regarding this alleged process, nor any physical explanation or modelling.

See our response to the comment (3). We admit that our interpretation is one of possible scenarios without strong evidence for it and thus added a sentence explaining that in the re-revised manuscript. We also changed the sentence here in the Abstract to tone it down (line 29-31). We understand that the term "diluted" is misleading in this context, the idea and physical explanation is that higher sedimentary accumulation rates lead to a reduced accumulation of atmospheric sulfur. We changed the wording accordingly in Abstract and in Discussion (new line no.: 441-449).

(8) *L.71 - "The Archean S-MIF has been conventionally defined by a sizeable (> 0 ± 0.2‰) Δ 33 S value [16]"*

The convention cited in this line refers to an information box in Farquhar and Wing (2003) which states: 'We note that the definition of CapD33S used in this paper specifically refer to the terrestrial distribution of sulfur isotopes and CapD33S values within ± 0.2‰ of zero can arise through the normal operation of mass-dependent fractionation processes.' This explanation is not anymore adequate to define the MIF-S thresholds, after 17 years of research and new data published the current understanding of MIF-S threshold has changed as discussed in many papers, among which LaFlamme et al. (2018). Please justify the motives for using Farquhar and Wing (2003) MIF-S thresholds. It is my opinion that Farquhar and Wing (2003) thresholds do not have anymore a coherent justification, therefore they should not be used.

We agree with the reviewer's comment and use the updated S-MIF definition by LaFlamme et al. (2018) as we clearly stated in the manuscript. We also removed the Farquhar and Wing (2003) thresholds from Figure 6. Nevertheless, we would like to emphasize that this sentence is part of the introduction that summarizes the notation of S-MIF in Archean strata and our citation of Farquhar and Wing (2003) is appropriate since this is one of the first papers on the topic (new line no.: 74-75).

(9) *L.125 - "(sample 12-007-1: 25°42'4.3" S, 31°04'46.1" E)"*

In my previous comments I asked 'not to report sample coordinates in the text, but provide a sample list as a Electronic Supplementary Material (ESM) specifying coordinates, sample names, sample types, and any other relevant information useful to characterise the samples' A sample list is still missing and coordinates are still within the main text. Modify accordingly.

As we already stated in the former response, we judged that it is useful for readers to describe the sample locations and their coordinates in the main text. We additionally showed the coordinates of the base and top of the analyzed sequence and a stratigraphic thickness of each sample on the Supplementary Table 2.

(10) *L.237 - Figure 5*

The mixing area drawn in Figure 5 is wrong. When BSR is involved mixing must account for non-atmospheric MIF-S. Revise Figure 5 following the indications provided in Ono et al. (2006), Aoyama and Ueno (2017, DOI: 10.1111/gbi.12268).

We agree with the reviewer's suggestion that a mixing line is precisely a curve on the $\delta^{34}S$-$\Delta^{33}S$ cross plot and our former mixing lines of the figure were incorrect. However, in our revision, we slightly modified the estimated seawater sulfate field on the $\delta^{34}S$-$\Delta^{33}S$ cross plot (Figure 6), based on the Paleoarchean barite records in the BGB (Bao et al., 2007; Roerdink et al., 2012). Accordingly, we modified the model and the former mixing between sulfide derived from seawater sulfate and hydrothermal sulfide is unnecessary anymore. In the revised model, the observed rounded pyrite grains with negative $\Delta^{33}S$ values in the Moodies can be directly explained by a reduction of seawater sulfate with $\delta^{34}S$ fractionation (revised Figure 6). We think the revised model is simpler and more straightforward than the previous one. Nonetheless, we agree that MSR is only one of several possible processes of seawater sulfate reduction, as the reviewer suggested. Both biotic and abiotic reduction scenarios are possible and it is difficult to constrain the reduction process in the older Onverwacht and Fig Tree sediments. We stated that in the revised version of our manuscript at new lines 330-332. What we would like to suggest here is that the seawater sulfate was likely a sulfur source for the formation of the rounded pyrite grains with negative $\Delta^{33}S$ values in the Moodies. Also see our response to the comment (2).

(11) *L.311 Discussion (general comments)*

"a small MIF signature (Δ 33 S < 1.0‰)"

Relative comparisons must have a term of comparison. Small/big compared to what?? Marin-Carbonne et al. (2014, http://dx.doi.org/10.1016/j.epsl.2014.02.009) documented zoned sedimentary pyrite nodules with both positive and negative CapD33S signatures, consider that sedimentary sulfur being dissolved and remobilised, it would be 100% sedimentary with an overall CapD33S proximal to zero. How can the author argue that since the gold deposits have a a small MIF signature, thus little sedimentary sulfur was involved. The term of comparison, in this part of the discussion is pivotal, as a consequence the current interpretations are entirely speculative.

We use the terms of comparison in the revised manuscript as the reviewer suggested. Agangi et al. (2016) concluded that the MIF-sulfur that was leached from older volcano-sedimentary successions was a sulfur source of the Sheba and Fairview gold deposits, based on the $\Delta^{33}S$ values of ore sulfide up to +1.0‰. As the reviewer suggested, the sulfide with S-MIF was likely dissolved into fluids, remobilized, and eventually precipitated with the gold bearing sulfides at 3.1-3.0 Ga, forming the ore. The $\Delta^{33}S$ frequency histogram of the sulfide of the gold deposits is similar to that of Moodies sulfide (Figure 11) and we infer that the precipitation of euhedral/subhedral pyrite in the Moodies sediments were associated with the gold mineralization episode in the central BGB. We newly formed the section 6.3 to emphasize our interpretation of the timing of supposed metasomatism in the Mesoarchean (new line no.: 398).

(12) *“Role of BSR”*

In order to suggest that bacterial sulfate reduction played a role in the modification of the original sulfur isotope signatures more robust evidence must be provided. Thermochemical sulfate reduction could have been active too, variation in pH during deposition could have caused similar shift in d34S. Many other processes and imparted a shift in d34S are reasonable candidates for explaining the d34S variation presented in this manuscript. For example, traces of biological remains should be provided in order to argue that BSR played a role. Otherwise this interpretation is conjectural.

See our response to the comment (2). We modified the sentences (new line no.: 359-363) and Figure 6, because our former description was inappropriate as the reviewer suggested.