

## **Review of: Observationally constrained analysis of sulfur cycle in the marine atmosphere with NASA ATom measurements and AeroCom model simulations by Bian et al.**

Bian et al. compares the spatial and temporal distribution of 4 sulfur-containing species (DMS, SO<sub>2</sub>, MSA, SO<sub>4</sub>) from the ATom airborne measurements with five global aerosol models participating in the AeroCom-ATom experiment. The study finds significant differences in the sulfur species concentrations between the models and the observations and identifies the reasons of these differences for the individual species. For DMS, the authors conclude that the emissions should be revisited, for SO<sub>2</sub> and SO<sub>4</sub>, the authors suggest that in the models the chemical conversion of SO<sub>2</sub> to SO<sub>4</sub> is too rapid and that the vertical transport of SO<sub>2</sub> is too weak. The study investigates further the model results by identifying the largest source of sulfur over ocean regions and finds that the largest contribution is dominated by anthropogenic sources followed by volcanic emissions and oceanic sources.

Given my expertise, my comments will mostly cover the modelling part and the logic of the study. The manuscript is well-written and the text well-structured. The methods and results are exposed in a logic and clear way and the whole manuscript can be easily followed and understood.

On the other hand, it is not clear what is the added value of this manuscript. The authors lack to highlight what is new in this study compared to previous literature. This is evident in the Introduction, where there is no mention of results from previous/recent studies using the ATom measurements nor using the AeroCom models. This is a major concern because the manuscript, as it is, looks like a comparison exercise with no follow-up message.

Because of this lack of novelty, I find the conclusions quite weak, and the relevance is not clearly articulated. More details are provided in the “Major comments” section below.

In light of these comments and those below, my recommendation is that the manuscript can be considered for publication in ACP once all comments below are properly addressed.

### **Major Comments**

- Lack of “what’s new”. The manuscript does not convey the message of what is new compared to previous literature. The Introduction needs to contain references to previous literature using the ATom measurements and the AeroCom models for studies of sulfur-containing species over land and ocean. I think that one paragraph for ATom and one for AeroCom would suffice. The Abstract and the Conclusions must also state what is new in the study.

This study can be relevant if, for instance, it is the first using the ATom airborne measurements. If this was the case, it must be emphasized considerably more in the Abstract, the Introduction (like P1L93-97 but much better/stronger) and the Conclusions (and throughout the text).

This study can be relevant also if a comparison between AeroCom models and ATom measurements has not been done before. In this case also, the authors should emphasize the originality of such intercomparison.

- Conclusions. Based on the major comment above, the Conclusions need to be strengthened. Once the novelty of the study will be identified (again, first intercomparison?), the Conclusions must be modified accordingly. Here follow the two main conclusion points that

need to be addressed:

1) It seems to me that the authors conclude that the dominant source of atmospheric sulfur (SO<sub>2</sub> in particular) over oceans is anthropogenic. It is well known that the largest contributions to the sulfur budget (SO<sub>2</sub> included) is due to human activities. Hence, despite the short lifetime of the sulfur compounds, it does not seem so striking that the major source of atmospheric sulfur is anthropogenic even over oceans (e.g., Chin et al., 2000). Thus, I have the impression that this conclusion does not add anything significantly new to the current knowledge.

In order to make this conclusion more appealing, I suggest increasing its relevance by emphasizing its unicity. I couldn't find any other modeling study asserting that the sulfur over the oceans is mostly caused by human activities. This means that the authors are the first proposing that conclusion (as far as my literature research goes), and that is certainly worth mentioning. However, I suggest to carefully check the literature for modeling studies looking at the atmospheric sulfur over the oceans. If the authors find a relevant study, it would be interesting to compare their findings with yours.

2) Another conclusion consists in the description of the significant differences between the observations and the models. This is somehow discussed in the text with the attribution of the differences to emissions (DMS, e.g., P9L379-394), chemical conversion (SO<sub>2</sub> to SO<sub>4</sub>, e.g., P10L449-451) and transport (SO<sub>2</sub>, e.g., P8L365-367) depending on the model, with a nice final discussion (P13L584588). However, these parts (especially P13L584588) can be explored further with a more detailed discussion of the models. Here's a few ideas for further discussion: 1) what are the common aspects among the models? and their differences? Can we discern something from similarities/differences 2) It is possible/feasible to perform additional sensitivity tests to assess model performances (not for this manuscript but as material for discussion)? 3) What would be the added value of satellite data in this comparison (again, for future studies): would we learn something more thanks to the denser temporal and spatial sampling compared to airborne measurements)?

3) Within the Conclusions, the authors can also dare to provide some outlook for future model developments by suggesting ways to improve the current models (e.g., update the chemical scheme, increase horizontal/vertical resolution, identify and try to improve relevant parametrization for transport).

- Tracer-tag. It would be very helpful if the authors added more detail to the methodology of the tracer-tagging concept used to obtain the source origins shown in Fig. 13. My understanding is that these sensitivity tests were conducted by the GEOS developers and made available to the authors. Even though the authors did not perform the experiments, the concept of tagged data needs to be explicitly discussed (currently it is superficially mentioned in Section 2.2) so the non-expert reader can understand the logic behind such sensitivity test.
- Figures. The figures are generally too busy – there is a lot of information that is not always entirely discussed. For example, the AMS measurements at 1s and 60s in Fig. 2 are barely discussed. In that regard, the authors state that they use 10s merged data for AMS in the text unless otherwise stated (P6L241-242). Based on that, I suggest removing the 60s and 1s lines from Figure 2 (maybe moving the 1s, 10s, 60s discussion to a separate paragraph in the

methods?). This way, the authors could merge the observations and models together in the same panels (like Fig. 3) and make Fig. 2 easier to read.

Because of the complexity of the figures, the discussion is often confusing in the sense that the authors do not specify any panel when presenting the results. For example, the analysis of Fig. 2 (P5L212-P6L264) never mentions one panel specifically. This makes it difficult to discern which flight (ATOM1-ATOM4) is considered.

In addition, the box-and-whisker panels are seldom mentioned, and they can be probably removed from there (and maybe merged and moved to Section 2 or moved to the supplement?). As a general rule, if a panel/figure is not discussed, it should not be shown.

For Figure 3, the statistics are briefly mentioned in P6L280-282. Please provide more discussion of the statistics shown in Fig. 3 or consider moving the figures/panels to the supplement, given the large number of figures in the manuscript.

Figure 4 also contains a large number of panels – (and the box-and-whisker panels are again shortly mentioned). As my comment above, please provide more discussion or move the panels that are not discussed in the Supplement. Furthermore, the addition of panels c,f,i,l breaks the continuity with respect to the previous figures, which contain only PDFs and statistics. These panels should belong to a separate figure (and paragraph) that discusses the vertical information from the observations/model comparison (maybe merged/moved to Section 3.2?).

A final note on the box-and-whisker panels: if the authors think that the statistics should be shown, maybe it can be shown as tables depending on the sulfur species instead of panels in the figures.

For Figures 9-11, I would suggest to average together the horizontal regions that do not differ much one from another (e.g., Fig. 9a, A40S-20S with A70S-40S) and adapt the discussion accordingly.

Once the figures will be simplified, the discussion will benefit from that and there will be more room to guide the reader through the different panels of each figure.

#### **Minor Comments.**

- P1L1: “[...] and ecosystems.”. Could you add one or two sentences on how the sulfur cycle plays the key role (e.g., pollution, acid rain)?
- P2L62-63: “wreak havoc”. Could you replace this expression with something like “devastate, destroy”?
- P2L83-84: “the TUT and above are observation-sparse regions”. I would rephrase this part as something like “...observations in the TUT region and above are sparse”.
- P2L91: Is it possible to specify what “DC-8” means?
- P3L123: “(i.e., ~0.2-12 km) (Thompson et al., 2021)” --> (i.e., 0.2-12 km, Thompson et al., 2021).
- P4L148-150: I recommend rephrasing this sentence to something like “Two instrument were used [...]: the California [...] and the NOAA [...] (Table 1)”.

- P4L177: “(CMIP6) (Feng et al., 2020)” → (CMIP6, Feng et al., 2020).
- P4L178: “(.... System (GFAS))” → (... System, GFAS).
- Section 3.1: I suggest homogenizing the discussion here. As it is, Fig. 3 is less discussed than Fig. 2 and Fig. 4. The authors could enhance the discussion for Fig. 3 with the addition of a couple of paragraphs on SO<sub>2</sub> or reduce the discussions of the Figs. 2 and 4.
- P6L252-264: The Atom flights are not discussed separately for Fig. 2 (SO<sub>4</sub>). I suggest providing some discussion about the different flights to maintain consistency with the following discussions of Figs. 3 and 4 (where the different flights are mentioned).
- P7L305: “Fig. 4c,f,i,l”. This is the only place of Section 3.1 where specific panels are mentioned. Since no other panel of Figs. 2, 3 or 4 is explicitly discussed, I suggest 1) either discussing explicitly the remaining panels or 2) removing the explicit mention to Fig. 4c,f,i,l and move these panels into another figure (as suggested in my major comment about Figures above).
- P7L309: I suggest removing “Apparently”.
- P7L317: I suggest rephrasing “...is uniquely having a....” to “..... is unique because it has a .....”
- P8L344-345: I suggest rephrasing “Despite that improvements are needed, ...” to “Despite the need for improvements, the models are generally able to capture the shape of the SO<sub>4</sub> profile.”
- P8L346-354: I find the introduction of the model Groups 1, 2 and 3 a little confusing (also, these groups are not mentioned again throughout the manuscript). I suggest keeping the model names as there are only 2 models per group and Group 2 consists only in E3SM.
- P8L358: I suggest rephrasing “... an order of magnitude, but around ....” to “... an order of magnitude around ....”.
- P8L359: The authors say that SO<sub>2</sub> is better in IMPACT in the NH and CAM-ATRAS and OsloCTM3 in the SH, but compared to what? Please specify the reference (I suppose that would be GEOS and E3SM?).
- P8L360-361: Please be more specific with regard to how the models should improve the SO<sub>2</sub> simulations (too large/little concentrations wrt observations?). I understand that the differences are thoroughly discussed in the following sentences, but it would be nice to have a preliminary “hint”.
- P8L369-370. “All models [...] ATom-1 observation”. I do not quite understand what the authors mean with this sentence. Could you please explain it?
- P8L371-375. Since this part refers to a figure in the supplement (Fig. S5), I suggest reducing this discussion here.
- P9L384: “The parameterization....”. Do the authors refer to the parameterization of Nightingale et al., (2000)? If yes, I suggest rewriting the beginning of the sentence with something like “That/This parameterization ....”.
- P9L396: “... than the observed one,” I suggest providing an example of a panel showing this steeper gradient (e.g., Fig. 7 54N-90N Atlantic). It would be nice to add panel numbers/letter to these vertical profiles to facilitate the navigation during the discussion.
- P9L399: I suggest rephrasing “tease out” with “obtain”.
- P9L396-410: Very nice discussion!
- P9L414: I suggest rephrasing “These behaviors are inconsistent with....” with “These patterns do not agree with....”.
- Section 3.3: Concerning the definition of the altitude ranges, I suggest keeping the same nomenclature both in the text and in the figures. Specifically, in the text the authors use

words like “free troposphere” or “boundary layer (BL)”, while in the figures they show only altitude ranges in kilometers (0-1.5, 1.5-6, .... km). My suggestion is to add the name of the layer to the range in kilometer in the Figures (e.g., “0-1.5 km” becomes “0-1.5 km (BL)”, etc.).

- P10L422: I suggest explaining what the Figures contain. Something like “In order to analyze [...], Figs 9-11 shows histograms of XX concentrations as a function of altitude (rows) and latitudinal band (columns)”.
- P10L432-433: “The most high [...] (NH spring).” Could the authors specify the altitude range here?
- P10L438: I suggest rephrasing “more polluted” with “tends to simulate higher SO<sub>4</sub> concentrations”.
- P10L438-440: “SO<sub>4</sub> concentrations [...] the Supplement.” I suggest expanding this part by moving here the relevant parts of the Supplement.
- P10L429-440: There is no mention of the difference between Pacific and Atlantic regions. I suggest adding a couple of sentences discussing this separation.
- P10L443-444: “..... and pollution affects ....” It is not clear to me what the authors mean with pollution. Could the authors explain?
- P10L444: I suggest changing “Areas where free tropospheric SO<sub>2</sub> pollution is relatively polluted ....” to “Areas where free tropospheric SO<sub>2</sub> concentrations are relatively large .....”.
- P10L442-451: there is no mention of ATom 2 in this paragraph. I suggest including one sentence discussing the most relevant pattern for ATom 2.
- P10L454: I suggest rephrasing “.... when the hemisphere is in spring” with “.... during springtime”.
- P10L458-459: This sentence concerning the BL sounds a bit redundant with part of the sentence before (P10L457, discussing the free troposphere) except for the point (3). I suggest merging the discussions together to avoid repetition.
- P10L460: I suggest rephrasing “The convoluted effort can be somehow alleviated by ....” with “Additional insights can be obtained by ....”.
- P10L463: I suggest replacing “giving” with “because”.
- P10L453-465: Also here, very nice discussion.
- P11L468: I suggest adding a sentence or two explaining the need for this Section (why it is important to look at the sulfur budget in model in light of what was shown before and what will come in Section 5).
- P11L489: I suggest rephrasing “... are pretty much the same as they should be” with “... are as expected”.
- P12L524: I suggest replacing “... is at its most active” with “is the largest”.
- P12L525: I suggest removing “and event”.
- P12L530: I suggest removing “where and”.
- P12L536 “... continental areas” I just have a question: is there any anthropogenic source of sulfur over the oceans (ships?). If yes, it would be nice to mention it.
- P12L554: “It has a clear...” What does “It” refer to? Tropospheric SO<sub>4</sub>? I suggest the authors to specify here.
- P12L556: “There are two ...” This sentence needs to be introduced. Something like “Concerning volcanic sources, emissions from volcanoes are of two types.”
- P12L556-P13L561: The two types of volcanic contributions need to be separated more clearly. I suggest something like “One type is the volcanic degassing [...]. The other type consists in the volcanic eruptions [...]”.

- P13L560: I suggest removing “eruption”.
- P13L571: I suggest adding “airborne” before “ATom”.
- P13L571-573: This nice sentence is the goal and “what’s new” in the manuscript. I suggest expanding this in the Introduction with references to previous studies (and a similar sentence in the Abstract). See my major comments on this aspect.
- P13L590: “over remote oceans” Again, this is the goal and should be stated more clearly in the Introduction.
- P13L599: “... this proportion is increased ...” Do the authors know the amount of the increase?
- Figure 9: In the caption, I suggest adding the seasons corresponding to the different Atom flights.
- Figure 10: The contribution from biomass burning (BB) is hardly discussed in Section 5 (which is logical!). Therefore, I suggest renaming BB to something like OTHER and state that it includes biomass burning (and any other sulfur source) and that it will not be discussed in Section 5 because its contribution is negligible compared to AN, VOL and OCN.

### **References.**

Chin, M., Rood, R. B., Lin, S.-J., Müller, J.-F., and Thompson, A. M. (2000), Atmospheric sulfur cycle simulated in the global model GOCART: Model description and global properties, *J. Geophys. Res.*, 105(D20), 24671–24687, doi:10.1029/2000JD900384.