

Editor Decision: Publish subject to minor revisions (review by editor) (04 Feb 2021) by Drew Gentner

Comments to the Author:

Please fully consider and address the comments in the recent re-review (copied here for your convenience):

This version of the manuscript has been improved relative to original submission, addressing several of previous reviewers comments. I commend the authors for this revision. However, while this is relevant to the scope of ACP, I have still some concerns with regards to the analysis of the results and to the evaluation of the updated emissions. I suggest minor revisions to address the following concerns before publication:

Response: We have addressed all the comments requested. We really agree with the idea to make a more targeted comparison with measurements and satellite data and we now believe that the manuscript has now improved substantially. Please note that we have also shortened the abstract and the conclusions, as there were details that were probably repetitive.

General comments

1/ I note that a particular effort has been done on the evaluation of the results but I still think that the evaluation section could be reinforced. The easiest way to see the effectiveness of the inversion is to show a gridded bias between the observations and the both the simulations using EGG or NE emissions. In this sense, you should complete the Figure S11. In addition, in my opinion, the evaluation against CrIS satellite data should be done at the regional scale in addition to the global one. It would not be an evaluation of the IASI data or of the model but of your updated emissions. It would complete the analysis of the evaluation against surface stations at least over Europe and over the US where your inversion can degrade the fit to the measurements compared to the prior EGG inventory.

Response: We really appreciate for this comment, as we had not thought about this kind of presentation previously. Since it is a very valuable figure, we have added the mapped station bias not in the supplements, but as Figure 8 of the main manuscript (see Track Changes). We have also added a paragraph where we try to explain what the main conclusion from this map is (see lines 572-580 in Track Changes).

In addition, we also agree that the comparison with the CrIS data should not refer to all datasets but only to the one we present here (NE). For this reason, we have restricted the comparison only for the NE emissions and only for North America, Europe and Southeastern Asia, as suggested by the Editor. For this we have substituted the previous figure with Figure 9 (see manuscript with Track Changes). Since section 4.2 became really small after limiting the comparison to the NE emission data only, we merged it with section 4.1, which is now entitled "Validation against ground-based observations and satellite products" (see manuscript with Track Changes).

2/ A comprehensive overview about the existing literature is still missing for the analysis of SO₂ changes. Figure S2 is not in agreement with Krotkov et al., 2016, ACP, showing strong decrease of SO₂ between 2005 and 2015 at least over Eastern US and over Eastern Europe with the OMI data themselves. These features could be explained by the choice of the authors to analyze their results for Europe or for the

US as a whole but it should be discussed in the text. The section 3.2, circa p.320-340 would be more complete with analysis not only for the North China plain, even if I understand the particular interest for this region.

Response: We have tried to clarify that the observed decrease after 2015 is only due to our choice to present global averaged, whereas others (Krotkov et al.) have already seen decreased concentrations after 2015 (Track Changes line 369-375). As already mentioned, the reason why we chose North China Plain to investigate changes in NH₃, reactants and precursor species is because the largest anomalies of SO₂ and NO₂ were seen there. We believe it would be beyond the scope of this paper to investigate in detail more regions in this respect, as the only reason for examining SO₂ and NO₂ was to explain any changes of ammonia's reactants, which would explain change in NH₃ emissions. We believe that Krotkov et al. has done a very complete analysis on SO₂ and NO₂ from OMI, much more detailed than what we have done in the present.

3/ The fact that the bi-directional exchange with surfaces is not taken into account should be mentioned. The potential impact on the inversion results should also appear in the text.

Response: Dear Editor, please do correct me if I did not understand well this query. If you mean that we do not discuss in the text the effect of the exchange of ammonia in/out from the hypothetical box (due to atmospheric transport) that we used as a proxy to calculate emissions, then we do not agree. We have dedicated a full section (4.3 Limitations of the present study), where we explicitly state that the exchange due to transport is an issue of the selected methodology and that we believe it is fair to assume it negligible due to the very short lifetime of NH₃ in the atmosphere. In case you meant something else, please be more specific and we will act on the manuscript to correct any issue.

Specific comments

Line 329: "To" instead of "to"

Response: A full stop was missing there, as the sentence stops before "To". It has been corrected (see Track Changes).

Line 330: "NO_x" instead of "NOx"

Response: We have now added a subscript at this point (see Track Changes).

Line 329-330: The sentence is not clear. Please rephrase"

Response: We now think the sentence is clear after adding the missing full stop. If not, please help us improve this point further.