

Anonymous Referee #3

This paper presents a much appreciated (much needed?) upgrade of global estimates of emissions of sulfur dioxide (SO₂) using a combination of bottom-up information (HTAP) and satellite measurements (OMI).

Each source of information has its pros and cons, but not necessarily the same, so it is worthwhile to try to integrate them into a combined bottom-up/top-down SO₂ emission inventory (HTAP-OMI).

The paper convincingly shows that, using the emission inventories in model simulations and comparing model results with independent observations of atmospheric SO₂, that the combined OMI-HTAP emission database provides better results.

The study reveals many discrepancies related to slight mis-allocation of sources, as well as well-known issues with regard to representativeness of measurements, satellite data quality, and documented emission sources like underreported regions.

Overall, the approach is a step forward towards the integration of bottom-up information with top-down information. The method also provides a direction for increased and integrated use of upcoming new satellite missions to better estimate worldwide sources of emissions.

Overall, the paper is well written and well structured, and publishable after consideration of the remarks and suggestions below.

Response: We thank Referee #3 for the encouraging comments. All comments and suggestions have been considered carefully and well addressed below.

[1] general remark

I know the use of “higher/lower” is commonplace science publications when referring to other things than heights, but Personally, I prefer to use “larger/smaller” rather “higher/lower” when discussing anything unrelated to heights, in particular in atmospheric science, in order to avoid confusion.

I leave it up to the authors to decide, but if agreed, please check the entire paper and modify accordingly.

Response: We thank you for the suggestions. We have checked the entire paper and modified it accordingly.

[2] typos and suggestions for minor grammar changes. Please note the remark regarding P9, L17-L19. Also note some suggestions are provided for improving a few figures.

P1, L31. Change to “We focus for the validation ...”

P1, L32. Change to “... and for which a relatively large number of ...”

P1, L33. Change to “... improves the agreement between the model and observations.”

P1, L34-35. Suggest to move “Additionally, our ... detected by satellites” to earlier in the abstract in L28, after “... with such missing sources.”. Then, suggest to change “Additionally” to “In addition”.

P1, L39/P2, L1-2. Suggest to move last line of abstract to after the previous suggestion (P1, L34-35).

P1, L37. Remove “For example,”

Response: Thanks. We have corrected them based on above comments in the revised manuscript.

P2, L13. “earlier” could be made more specific (SO_2 emission regulations in the US and EU are introduced in the 1980s)

Response: Thanks. We have changed it in the revised manuscript, as follows:

“in particular earlier (since the 1980s for power plants) in the US and Europe (Crippa et al., 2016)”.

P2, L21. I think it should be “removal”, not “removals”

P2, L21. Suggest to change to “The spatial distribution of emissions is even more uncertain”

P2, L21-22. Change to “... are in most cases allocated by spatial proxies rather than ...”

P2, L23-L24. Move “developing” to the beginning of the sentence: “In addition, developing ...”

P2, L26. Replace “indicate” with “identify”

P3, L5. “Gaussian distributions” or “Gaussian dispersion”

P3, L11-L12. I think what is meant here is “that combines information about large SO_2 sources from ...”

Response: Thanks. We have corrected them based on above comments in the revised manuscript.

P4, L3-5. Please provide a brief justification or explanation (or reference) for the source strength dependence on parameter L.

Response: Thanks. We add the reference of Fioletov et al.(2016) in the revised manuscript.

P4, L7. Unclear where “In this way ...” refers to.

Response: We agree that the phrase is confusing. We deleted the sentence in the revised manuscript.

P5, L1. “monitoring systems” (plural)

P5, L20. Latest: later, more recent?

P5, L24. Replace “it is located” with for example “emissions are located” or something similar

Response: Thanks. We have corrected them based on above comments in the revised manuscript.

P5, L30. Explain what is meant here with “profile”. In atmospheric sciences, a “profile” generally refers to a vertical distribution of a parameter.

Response: We changed “monthly profile” to “monthly variations” in the revised manuscript.

P5, L33. “OMI-HTAP inventory”

P6, L4-L5. Delete “On the contrary,”

P6, L24. Change “less” to “smaller”

P6, L33. Maybe replace “reported” with “documented” to avoid double use of “report” in the sentence?

Response: Thanks. We have corrected them based on above comments in the revised manuscript.

P7, L1. “Long standing experience”, would still be nice to add some references.

Response: We add the reference of Hoesly et al. (2018a) in the revised manuscript.

P9, L14. “identifies” maybe change to “shows” or “reveals”?

Response: We changed it to “reveals”.

P9, L17-19. I think it would be nice to mention here (or in the conclusions) that OMI-HTAP changes 2008-2008 are more consistent with EPA estimates than HTAP. Important finding.

Response: We pointed out the better consistency in the revised manuscript, as follows:

“The magnitude of SO₂ emissions decreases by 25% in OMI-HTAP during 2008 to 2010, consistent with the decline of 25% reported by EPA (EPA Air Pollutant Emissions Trends Data; available at <https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data>) and much larger than the decline of 9% in HTAP.”

P11, L3. Change “knowledge of” to “information about”

P12, L1. “my” should be “may”

P12, L6. Replace “less” with “fewer”

P12, L14. “averaged” to “average”

P12, L31. Maybe use “The OMI-HTAP emission database developed ...”?

P13, L1. “leading”, maybe “widely used”?

P13, L5. “since decades” to “for many decades”?

P13, L12. “The exact location of ...”

Response: Thanks. We have corrected them based on above comments in the revised manuscript.

[3] figures

Figure 2, upper panel. Most land areas are color coded in dark blue/purple. Does this include the 0.0 value, or does this mean that emissions indeed are larger than zero. If not zero, this also begs the question how SO₂ emission over most land areas are larger than zero. If indeed zero, it is worth considering color coding land areas with zero SO₂ emissions in grey or white. Please clarify in caption and/or modify the figure.

Response: The legend does not include the 0.0 value. We have clarified this in the revised caption.

The figures below display the maps for HTAP SO₂ emissions from the energy (top) and the residential (bottom) sector for year 2010 at the resolution of 0.1° × 0.1° (available at http://edgar.jrc.ec.europa.eu/htap_v2/index.php). In principle, emissions are spatially distributed based on their locations. As shown in the map for the energy sector (top), emissions from large point sources like power plants are allocated to grid cells based on their coordinates and thus there are many land areas showing no emissions. However, this is not the case for sectors with areal sources as dominated sources, like the residential sector (bottom). Emissions are distributed to grid cells using spatial proxy due to the lack of information on locations and thus most land

areas are distributed to emissions. In addition, emissions are regridded at much lower resolution ($1^\circ \times 1^\circ$) for illustration in Figure 2. It further improves the possibility of being distributed to emissions for individual grid cells, as the size of grid cells is larger.

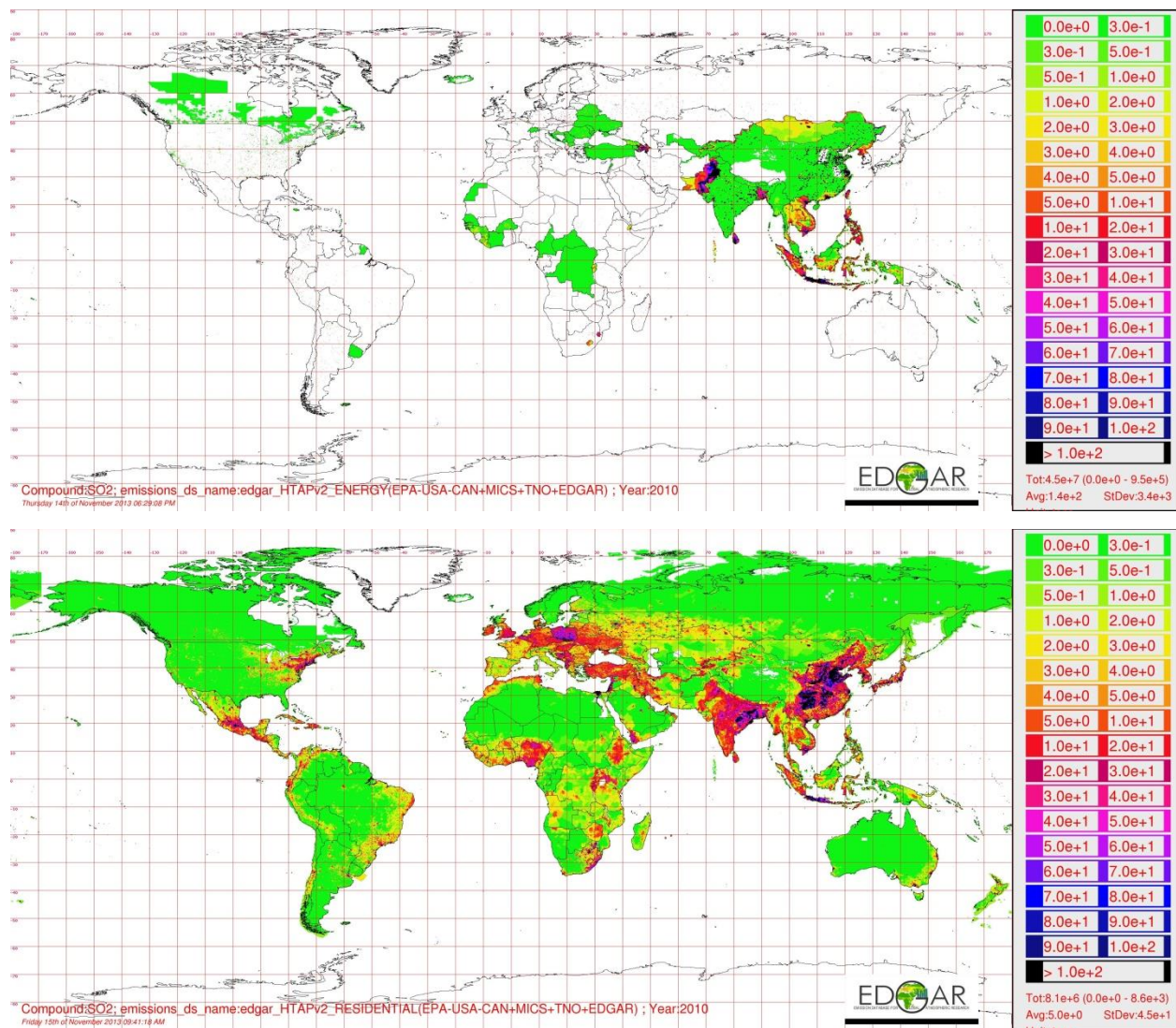


Figure 2, lower panel. Is not necessary insightful because of the small pixels. Is it possible to use symbols like a filled circle that for example indicate the size of the difference? I don't know if that would work, but I think it is worth trying to see if that helps for the usefulness of the panel. Also, it might be worth to color code non-zero differences between ± 5 Gg/yr/grid with greys (see discussion about the upper panel). As the panel now stands, there is no separation between zero emission locations and locations with small differences.

Response: Thanks. We changed the symbol accordingly and color coded small differences with light yellow in the revised figure. The areas without emissions changes are separated by grey as well.

Figure 3, upper panel. Add whites for (near) zero concentrations, as now everything is mostly blue, not very appealing. And similar to previous discussion, maybe separate small concentrations from zero concentrations.

Response: Thanks. There are no grid cells with zero concentrations at the modelling resolution of $0.5^\circ \times 0.5^\circ$. We have added whites for near zero concentrations in the revised figure.