

Interactive comment on “Modeling study of the impact of SO₂ volcanic passive emissions on the tropospheric sulfur budget” by Claire Lamotte et al.

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Dear Authors,

I have seen your manuscript "Modeling study of the impact of SO₂ volcanic passive emissions on the tropospheric sulfur budget" in ACPD and it captured my interest. Unfortunately, while reading the manuscript, I have found a number of possible major flaws that I would like to mention during this Discussion phase. Please find it in the following.

The manuscript introduces a new and more detailed volcanic emission inventory (by

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Simon Carn), input to MOCAGE CTM modelling, and evaluates the improvements brought in the global and regional sulphur budget with respect to older inventories using satellite observations as reference. The topics of this manuscript is of certain interest for multiple communities (atmospheric modellers, atmospheric scientists, climate scientists and volcanologists) and is worth attention. Unfortunately, I have found the following major flaws that, in my opinion, invalidate the results of this work that I think should not be published in the present form. I'll be available to review a possible revised manuscript in case the Editor thinks it useful.

My best regards,

Pasquale Sellitto

Major comments:

1) In the introduction lines of Sect. 3 (L150-155), it is said that SO₂ is the main volcanic effluent and is the only volcanic emission considered in this work. This is absolutely not true. The single most important volcanic effluent is not SO₂ but water vapour, with water vapour/SO₂ emitted mass ratios reaching values as large as a few hundreds. I think that many emissions and near-source volcanic processes can safely be neglected, as a first approximation, like halogen emissions and their impact on sulphate formation, transition metal contribution and other interactions of SO₂/sulphate with ash including heterogeneous chemistry; nevertheless, volcanic water vapour emissions cannot absolutely be neglected, as well as their in-plume effects on sulphate formation and SO₂ depletion. In my perspective, neglecting water vapour emissions (as said, the dominating gaseous effluent in volcanic degassing) invalidate the results of this work.

2) I agree with Referee #1 on the fact that ACSAF GOME-2 retrievals are not a good choice for the validation of the MOCAGE simulations. I'd also mention that, differently to what said at L287, GOME-2 data are not completely independent on the OMI and TOMS input data to your inventories: the three instruments operate in the UV spectral range and use similar spectral ranges and SO₂ absorption structures for the retrieval.

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Why not using infrared instruments as IASI, instead?

3) Also, the choice of MODIS AOD is quite debatable strategy. MODIS AOD is linked to all aerosols, not only sulphates. how do you separate sulphate aerosols from the other aerosol typologies/composition/sources? For example, in "Region 3 (Mediterranean)" dust is, on average, overwhelmingly dominant with respect to sulphate aerosols: how can you check the improvement of volcanic SO₂ sources in such an environment, due to the expected small sulphate signal?

4) In addition, how to interpret the results of the comparisons in Tables 3 and 4? Am I wrong to say that observations and simulations compare very weakly? This is also the case if looking at the (necessary in the main text) Figure S1. The simulations and the observations seem to not describing the same SO₂ fields. Results for aerosols compare better but, in my opinion, only because the aerosol fields are dominated by other aerosols (and MODIS is more sensitive to higher altitudes aerosols than boundary layer aerosols, so again probably large dust plumes lofted by convection).

5) In general, the manuscript is poorly written and needs a thorough linguistic review. The description of the scientific context is quite approximative and a lot of key references are lacking – please see specific comments in the following.

I also add here specific and technical comments so to help improving the manuscript.

1) L1: "The contribution...": what contribution?

2) L9: "negliable": "negligible"?

3) L20: Here and all the following discussions (including comparisons with your results): recent assessments of sulphur budget should be discussed here, like (for volcanically quiescent conditions, so of large interest for your study):

- Sheng, J.-X., Weisenstein, D. K., Luo, B.-P., Rozanov, E., Stenke, A., Anet, J., Bingermer, H., and Peter, T. (2015), Global atmospheric sulfur budget under volcanically quiescent conditions: Aerosol-chemistry-climate model predictions and validation, J.

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Geophys. Res. Atmos., 120, 256– 276, doi:10.1002/2014JD021985.

4) Also, for climate impacts, this should be cited and possibly discussed:

-Kremser, S., et al. (2016), Stratospheric aerosol-Observations, processes, and impact on climate, Rev. Geophys., 54, 278– 335, doi:10.1002/2015RG000511.

5) L22: "variation of climate": You mean "climate forcing"? (in this case, please specify that you're not talking about SO₂ but sulphate aerosols)

6) L23-24: "SO₂ emissions had become a major concern in environmental policies, leading to strong reductions in anthropogenic emissions in recent decades." : Not everywhere. Please differentiate geographically between decreasing, stationary and increasing emissions regions and add a reference.

7) L24-25: "Thus, the relative proportion of volcanoes in the total sulfur emission sources tends to increase.": Due to different regional trends of anthropogenic emissions, this statement sounds just arbitrary (unless you have specific references that I don't know)

8) L26-27: "is greater in altitude": you might mean: "increases with altitude"

9) L27-28: "Thus, we now...emissions": not clear, please rephrase

10) L27: "longer": "for longer time periods"?

11) L29: "these variations": which variations?

12) L44-45: Please change the phrasing here: there are very few "easy-to-access" volcanoes (Masaya can be mentioned, maybe), while normally the internal processes themselves build "uneasy-to-access" morphological structures for volcanoes.

13) L51: "information on injection altitude is available": The information on the altitude is still very limited. These are observing systems that have a few units of Degrees of Freedom in vertical profile observations of SO₂, mostly between 1 and 2.0-2.5, so not

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allowing for detailed altitude information. Please mention this in the text and smooth this statement.

14) L56: what do you mean with "more numerous and qualitative data"?

15) L63-65: "In contrast, few studies focus on the impact on tropospheric composition including air quality, with the exception of case studies of volcanic eruptions...": This is not true. Please look at the following papers of my research group, that aimed at the impact of volcanic activity, including passive degassing of selected volcanoes, on the tropospheric composition and air quality, and the many references therein:

- Sellitto, P., Zanetel, C., di Sarra, A. et al., The impact of Mount Etna sulfur emissions on the atmospheric composition and aerosol properties in the central Mediterranean: A statistical analysis over the period 2000–2013 based on observations and Lagrangian modelling, *Atmospheric Environment*, Volume 148, 2017, Pages 77-88, <https://doi.org/10.1016/j.atmosenv.2016.10.032>.

- Sellitto, P., Salerno, G., La Spina, A. et al. Small-scale volcanic aerosols variability, processes and direct radiative impact at Mount Etna during the EPL-RADIO campaigns. *Sci Rep* 10, 15224 (2020). <https://doi.org/10.1038/s41598-020-71635-1>

Please correct the wrong statement and cite the previous work mentioned above.

16) Section 2.5: What about the vertical transport, which can pose problems for the modelling of confined plumes, like volcanic plumes, and is discussed in the following paper?

- Lachatre, M., Mailler, S., Menut, L., et al., New strategies for vertical transport in chemistry transport models: application to the case of the Mount Etna eruption on 18 March 2012 with CHIMERE v2017r4, *Geosci. Model Dev.*, 13, 5707–5723, <https://doi.org/10.5194/gmd-13-5707-2020>, 2020.

17) Section 3: see Major Comment 1

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18) L165: "calm": What do you mean with "calm"? "Non-eruptive"?

19) L167: the reference to molecular structure sounds strange here. You might want to say that "SO₂ and ozone have absorption bands at overlapping spectral regions" (which is linked to molecular structure) or something like this.

20) L181: "...as one of the largest passive emitters.": Clumsy phrasing. Please rephrase.

21) L181-182: Please add details on the sources of these flux information.

22) L183-184: This is very unclear. Please clarify.

23) L188-189: You mean that volcanic SO₂ is emitted at the surface (including orography)? Is orography "smoothed" by the average in-grid topology? This aspect is very important e.g. for Etna. Even in case of passive degassing, its emissions are released at, at least, 3000 m altitude and episodic eruptions can reach, for Etna and Kilauea, quite higher altitudes.

24) Section 3.2: there are many repetitions. In general, all the paper should be condensed and repetitions should be suppressed.

25) L224-226: "We implemented...emissions": Why this parameterisation is not described in details here? How it compares to established parameterisations like the one of: ?

- Mastin, L. G. (2014), Testing the accuracy of a 1-D volcanic plume model in estimating mass eruption rate, *J. Geophys. Res. Atmos.*, 119, 2474-2495, [doi:10.1002/2013JD020604](https://doi.org/10.1002/2013JD020604).

26) L239: "Finally, the availability of emission heights in this inventory gives a better description of the emission.": At this point I think it is necessary to discuss the limitations in the vertical characterisation of volcanic emissions in the new inventory and the satellite observations used to build it, so to not oversell your new simulations

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- 27) Figure 1 and most figures: Please use larger text and labels.
- 28) L269: "lowest eruptive. . .negligible in 2013": How much this is "low"? Is it really negligible? How do you qualify this as "negligible"?
- 29) L272: reference to summer and winter: Please correct to "northern hemisphere summer/winter" and adapt the discussion.
- 30) L284-285: "'Due" and "since" in the same sentence is quite clumsy. Please rephrase."
- 31) Section 4: see Major Comments 2-3.
- 32) Section 4.2.2 title: "MODIS Aerosol Optical Depth"
- 33) L349: Please check altitude of Mount Etna, this is not the right altitude.
- 34) Section 5: see Major Comment 4
- 35) Section 5: It looks like some of the Figures in the Supplements are needed here in the main text, e.g. S1
- 36) L430: "(industries. . .": and dust, of course
- 37) Figure 8: This figure would be largely more useful with an altitude vertical axis (instead of pressure).
- 38) L517: "This corresponds. . .eruption": This is quite straightforward interpretation of these results, but it is important to stress the fact that 2013 is not a "normal" year as even a small number of explosive volcanic eruptions can change the vertical distributions of Figure 8 at the global scale. This has to be discussed and the limits of your simulation (a "predominantly passive degassing" year) must be clearly stated.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1040>, 2020.