Review of "*Methodology to obtain highly resolved* SO₂ *vertical profiles for representation of volcanic emissions in climate models*" by Oscar S. Sandvik and co-authors

The manuscript describes a method employing trajectory calculations to combine information from two satellite instruments and deduce the vertical distribution of SO₂ in fresh volcanic plumes. Such a multi-sensor analysis method that involves some modelling is probably at the edge of the scope of Atmospheric Measurement Techniques, but because the output can be regarded as higher level observational data, I would still rate it within the journal's scope. The manuscript is appropriately structured and well written and the figures are well composed and of high quality.

The one thing that I find not (yet) convincing is the value and purpose of the dataset created by the new method. And there are a few aspects of the method that are not fully clear to me and that may bear some caveats, which in my opinion are not properly discussed. I will touch on these issues in some general points below and then point out a few more specific points that may need attention but that are probably minor in nature.

General points:

- 1. At the end of the abstract, a "gridded high vertical resolution SO₂ inventory that can be used in Earth system models" is mentioned. The retrieval of the SO₂ height distribution in AIRS swaths is well described, but I'm not sure I would classify that as "gridded inventory". Of course, for global Earth system models, SO₂ data on a regular horizontal/vertical/temporal grid would be of great value. If your data product is anything like such a larger gridded inventory, then please do describe this in more detail (on page 10, you describe gridding the FLEXPART output on a 0.5 x 0.5 degree grid, but that is prior to linking it to the AIRS information, so it is not your final "product", or is it?).
- 2. In the same context, the simpler (compared to a "gridded inventory") vertical information for volcanic plumes/clouds/sub-clouds may still be very useful for modellers. But I wonder if hing-ing them to the AIRS swaths in space and time is the most useful thing that can be done. If you assume that the vertical distribution of the aerosols observed by CALIOP is representative for the SO₂ distribution, then why not calculate everything right back to the volcano? Vertically resolved emission plumes could then be "injected" into models of any scale.
- 3. There has been a rather comprehensive study on the Sarychev plume and its dispersion by Wu et al. (2017), and I was rather surprised to see no mention of it in your paper!

I suggest you look carefully at this previous work that also combines satellite observations and trajectory calculations. It should probably be mentioned in your introduction, and a comparison of the two studies (What was done similar? What different? Are the results in agreement?) in your discussion is clearly warranted.

Note that while I have been working on this review, a short comment was uploaded by the authors of the Wu et al. (2017) paper. This comment contains some excellent ideas and suggestions as to how connections between the two studies could be made, and I strongly recommend you to carefully look at this short comment and follow the recommendations.

- 4. According to your method description, you use ERA-5 reanalysis data to drive the FLEXPART trajectory model and MERRA-2 to determine tropopause heights and potential temperature levels of the CALIOP data (Page 7, lines 22 25). Would it not be more consistent to use one single reanalysis data set for both?
- 5. Can you give more detail on how horizontal transport and transforms between different vertical coordinates is realized? On page 9, lines 6 10, you explain that potential temperature "is stored" for the released tracer particles. On page 10, lines 24 26, you state that you "did not use the FLEXPART output vertical coordinates" and "set a single height interval between 0 and 50 km in the output grid specification". I find this confusing. Do you mean that you transport the parcels from the CALIOP SWATH converting geometric altitude to potential temperature in the beginning and then convert the same potential temperature value back to geometric altitude at the end? And when you say "locations" on page 9, line 10, does that include time in that case?

If the potential temperature of an air parcel is indeed not allowed to change during transport in your simulations, then the method would obviously not work in the tropics, where cross-isentropic vertical transport is common. Also, self-lofting in fresh volcanic plumes would not be accounted for, correct?

Some clarifications and a more extensive discussion of this would help.

- 6. I do wonder to what extent the issues and uncertainties I mentioned under points 4 and 5 above could factor in producing the bimodal distribution in the sum of your study that is shown in Figure 7, and also on the height of your SO₂ plume being somewhat higher than the earlier studies you mention. First, the relationship between potential temperature and geometric altitude is probably not the same at the different locations (I guess the profiles from all the earlier studies are close to the volcano in space and time?), so when you strictly transport along isentropes, it may be better to do this comparison on a potential temperature vertical coordinate. Second, self-lofting could explain at least part of the difference in altitude and should be mentioned.
- 7. As you state on page 3, lines 8/9, an important assumption for your method to work is the horizontal and vertical co-location of volcanic SO₂ and aerosol following the eruption. While this assumption is probably reasonable for a wide range of eruptions and conditions, I still suggest a more detailed discussion to further support this assumption: what do we know from the literature with respect to the SO₂/aerosol co-location? Are there conditions, under which the assumption may not hold, and on what time scales will it hold? How about ash-rich eruptions, where particle sedimentation rates may be higher (you mention this at the end of page 6)?

This may all not be critical for your particular case looking at rather short time scales not so long after the eruption for a non-tropical eruption. But as you describe a new method that should hopefully be applicable to other cases, such caveats and limitations should be made more clear.

Specific and minor points:

<u>Page 2, line 15:</u> The Watts (2000) reference for OCS sources and sinks is in the middle of the timeline of possible references, i.e. it is neither the first one nor is it up to date. I suggest to cite Kremser et al. (2016, already in your reference list), which contains a fairly recent OCS budget together with a discussion of source and sink processes.

<u>Page 9, lines 1 – 5:</u> I'm not sure that I have fully understood the scaling of the number of FLEXPART particles. Are you saying that you are releasing *x* FLEXPART particles per one CA-LIOP pixel, and *x* scales with β_{aer} of that pixel?

<u>Page 12, lines 2 – 3</u>: As the reason for the lack of trajectories matching some of the AIRS SO_2 clouds, you mentioned that these SO_2 detections could be located below the tropopause, while only the stratospheric aerosol scattering observed by CALIOP is "transported". Have you tried testing this by, e.g., using TP - 2km instead of TP as vertical threshold?

References:

Wu, X., Griessbach, S., and Hoffmann, L.: Equatorward dispersion of a high-latitude volcanic plume and its relation to the Asian summer monsoon: a case study of the Sarychev eruption in 2009, Atmos. Chem. Phys., 17, 13439–13455, <u>https://doi.org/10.5194/acp-17-13439-2017</u>, 2017.