We thank the reviewers for constructive feedback that has improved our manuscript. The reviewer comments are given below in black font followed by our answers in this blue color.

This paper presents a new method deriving vertical profiles of SO2 representative of volcanic emissions by combining space-borne observations of SO2 by AIRS/Aqua and of aerosol by CALIOP/Calipso. The profiles are constructed by connecting the CALIOP high resolution aerosol profiles with the AIRS SO2 columns using trajectory calculations. The interest of providing SO2 profiles at the period of a volcanic injection in the stratosphere is definitely clear for the community especially for the initialization of Chemistry-Transport and Climate Models that aim at estimating the chemical and radiative impacts of volcanoes. So I found the idea behind the manuscript of very high scientific return. I found the manuscript structure pertinent, well-written and going straight to the obtained results (but a bit too much). Although I estimate that this work is really worthy of publication in AMT, there are some methodological elements that still need to be clarified throughout the text.

General comments:

It can be difficult to grasp how the trajectories interconnect both satellite datasets (e.g. how they intersect, typical trajectory lengths). An illustration of a specific case linking CALIOP at a given altitude level and a AIRS swath would be helpful for reader. The relevance of using forward trajectories starting from CALIOP aerosol profiles (aerosol are a product of SO2 so I much better clearly understand the use of backward trajectories) can be clarified too . Why in the example of the command file the simulation lasts for 6 days?

We ran all FLEXPART simulations for the same time-span (June 14th – 23rd) for programming simplicity. (Forward trajectories were run from the CALIOP sampling time to the 23rd June, and backward trajectories were run from the CALIOP sampling time to the 14th). However, we only used the parts of the trajectories needed to connect the CALIOP profiles to the AIRS swath of interest.

The reason for using both forward and backward trajectories is that it enables us to use the information from as many CALIOP swaths as possible for each AIRS swath. Height information from each CALIOP swath was transported by FLEXPART to the AIRS (at their specific time and location of the AIRS swaths). Also using forward trajectories enabled us to use more CALIOP swaths, and hence more height information, in our analysis. In order to make this clearer we have included a conceptual figure of the method in the introduction (Fig. 1 in the new version of the manuscript).

To me (but I may be mistaken!) the authors should find CALIOP and AIRS swaths capturing the volcanic plume every day, allowing them to compute trajectories of a few hours only to connect both satellite observations.

There are CALIOP and AIRS swaths every day, however only using CALIOP data from the same day as the AIRS swath limits our analysis to a very small part of the volcanic cloud since the swath width of CALIOP is very narrow. We include information from CALIOP swaths from several days to have better height information from a larger part of the volcanic cloud.

We understand that this was not properly addressed and have changed the text (in the abstract as well as in the introduction) to clarify the advantage of using multiple CALIOP swaths.

A clearer description of the limitation in the method is missing. For instance, since what would be the impact of the sedimentation of the aerosols in the time length of the trajectories that on the attribution of the retrieved SO2 plume heights? I guess the impact would be limited but I would suggest the authors to mention it.

Yes, the sedimentation rates for fine mode particles are negligible in the time-frame and vertical resolution of our study. We agree that this is worth mentioning and added text to the manuscript (section 2.2).

In case of the presence of ash which has been reported for some eruptions (Kelud, Raikoke) and as derived from CALIOP depolarization/colour ratios, how the method would be affected? Can the CALIOP instrument adequately distinguish between ash and sulfate to properly associate the aerosol detection with the corresponding SO2 observation away from it?

As the reviewer suggests, the sedimentation of ash would be spotted by CALIOP as a layer of larger particles (from the color ratios) with high depolarization ratios below the sulfate layer. We did not observe this in our study, but it could be important if studying the two eruptions mentioned by the reviewer. For Kelut, the sedimentation rate was small (~500 m/month) (Vernier et al., 2016) compared with the time-frame (and vertical resolution) in our study. However, studying ash rich eruptions may require further development of our method.

The relevance of the methodology for other reported eruptions could be more discussed.

We agree that this was little discussed in the manuscript. We added a few sentences on the topic in the Discussions section.

Specific comments:

Introduction: I found the comment by Xue Wu of high interest and I strongly recommend

to add their references about their similar method in the introduction.

We agree, but were not aware of their study before. We have added a few sentences on their work in the introduction section. Furthermore, we decided to contact the authors regarding their data, and have added it to the figure with comparison of vertical SO2 profiles (old Fig.7, now Fig. 8). See also comment #3 by reviewer #1.

P1 line 22: the temperature impact of the Pinatubo aerosols is debated in the community and I suggest to add more recent references such as Canty et al., Atmos. Chem. Phys.,13, 3997–4031, 2013 http://www.atmos-chem-phys.net/13/3997/2013/

We agree that our statement was somewhat exaggerated and changed to *"tens of a degree Celsius"* (citing Kremser 2016 and Canty et al., 2013)

P3 line 22: The authors should cite also the work of Günther et al. (Atmos. Chem. Phys., 18, 1217–1239, 2018https://doi.org/10.5194/acp-18-1217-2018) who present a synthesis of the emitted SO2 masses and height ranges (see their Table 1).

We have now included the SO2 profiles from Günther et al., and Höpfner et al., (2015) (referenced in Günther et al. (2018) to Fig. 8.

P4 about AIRS: Do the authors know how this dataset compares with other SO2 datasets such as IASI (Clarisse, L. et al.: Retrieval of sulphur dioxide from the infrared atmospheric sounding interferometer (IASI), Atmos. Meas. Tech., 5, 581–594, https://doi.org/10.5194/amt-5-581-2012, 2012)? If available, I would suggest to add this information in section 2.1.

We decided to limit the discussion of different SO2 instruments since our method does not use their vertical information.

Figure 1: following the general comment given above, adding the CALIOP paths superimposed to AIRS swaths would be helpful for the reader to see which part of the SO2 plume is closely captured by CALIOP. Please specify the exact date and/or time range of each swath on Figure 1.

CALIPSO/CALIOP and AQUA/AIRS ran in the same satellite constellation (A-Train). The concurrent CALIOP swaths are almost in the center of the AIRS swaths. We used 75 CALIOP swaths together with the AIRS dataset, and most of these did not occur at the same time as the AIRS observations used in Figure 2. Adding the CALIOP paths to the figure would not provide any information on which parts of the volcanic clouds that have been sampled by CALIOP since the SO_2 cloud move and change shape during this time lag. We therefore wish to keep the figure layout as simple as possible. In response to the general comment above, we added a concept figure (new Fig.1) illustrating the relation between the CALIOP and AIRS data.

Furthermore, we have added the time of each AIRS swath to the figure.

P6 line 11: why the investigation of the proportion of the SO2 plume located in the troposphere, which is by the way an interesting information, cannot be done automatically? Is it due to errors in following air masses using the trajectories in the troposphere (especially along isentropes)?

It is difficult to do this automatically with CALIOP, due to frequent presence of ice-clouds in the troposphere. The volcanic aerosol is difficult to separate from these ice-clouds and we therefore excluded the troposphere to prevent signals from ice-clouds.

P6 line 23: I am fine with the method using depolarization and colour ratios from CALIOP observations to point out the presence of ash and ice but there is a lack of information here. At least please refer to Vernier et al. (2016) who describes the method using depolarization ratio and add the information accordingly.

We agree with the reviewer. We have fused that section with the next one where Vernier et al. (2016) and further references on depolarization ratios of ice and ash are discussed (at the end of section 2.2).

P7 line 24: Judging by the difference in vertical resolution between ERA5 (several hundreds of meters) and CALIOP (60 m) in the stratosphere and also horizontally, please specify that the meteorological fields are spatially interpolated to release trajectories from each CALIOP profile.

We released particles at specific heights in FLEXPART corresponding to the CALIOP heights. The interpolation of the ERA5 data is done within FLEXPART and not by us. This type of interpolation is standard in dispersion models.

P7 line 27: I do not really understand what the authors mean here with the 2 standard deviations (in meter and mbar) provided here. Do they correspond to the final vertical resolution of the SO2 profile?

Yes, this is the vertical resolution of the profiles with height coordinates transformed from potential temperature to geometric altitude and pressure. We have rewritten this sentence to make our point more clear.

P8 line 17: Please better define what you mean by a pixel for CALIOP. How do you obtain pixels from a smooth signal (i.e. from fig 4b to 4d)? This is an important step I think.

We have changed the word pixel to grid-cell here.

P9 line 2: "backscatter" instead of "scattering" OK, we changed accordingly.

P9 line 4: When mentioning "95,000" particles, do you mean in total for one subcloud or for each pixel? What is the time step of the particle release? Every 6 hours?

We mean maximum 95 000 particles in total per CALIOP swath (per simulation), i.e. all pixels and sub-clouds identified in that swath. However, two of the CALIOP swaths have been divided into two Flexpart simulations when the sub-clouds were far away from each other. We release particles only once per Flexpart simulation, at the time of the CALIOP swath (or the closest half hour mark).

We have made changes to this section to clarify this.

P10 line 30: the authors do not provide details about the role of the weighting of FLEXPART outputs and how they do it. Please clarify.

We have written a more clear explanation on how the weighting was performed.

P11 line 3: I do not really agree with this statement. E-folding of SO2 is about 13-17 days

(see Haywood et al., 2010; Lurton et al., 2018). It can be mentioned here that 9 days is

lower than the reported e-foldings for the Sarychev proving that SO2 is still present in high

quantities over the 9-day time length of the trajectories.

We agree that the transformation can have some impact on our analysis. It is difficult to estimate this impact since the e-folding time for SO2 is uncertain. We have changed the text in section 3.3 to: *"…The time it takes for the aerosol to form could thus affect the representation of the cloud. The nine days used here is shorter than previous estimates of volcanic SO_2 conversion in the stratosphere (Andersson et al., 2013), but ongoing SO_2 conversion adds uncertainty to our estimation. The short time span covered in the present study can thus be assumed to have a small effect on β_{aer} per SO_2. This can be seen in Fig. 8 in Friberg et al. (2018), where the stratospheric aerosol load from Sarychev peaks in September, i.e. months after the last swath used in this study…"*

Figure 5: I suggest the background of the AIRS swaths to be coloured in grey rather than

dark blue to better highlight SO2 fields.

Setting a threshold for the background SO2 concentration is difficult. We have used the same color scale for the swaths in Figures 1, 5, and 6, and wish to keep it that way.

Did the authors focus on the 18th? Is it because there is concentrated SO2?

Yes, we focused on the 18th since AIRS had good coverage of all volcanic sub-clouds during that day. That way we could obtain an integrated view of all the Sarychev eruptions taking place in June 2009.

Why trajectories are calculated over 9 days? .

Our wish is to use as many CALIOP swaths as possible to get the best possible representation of the vertical distributions in all sub-clouds.

In figure 5, how many CALIOP profiles have generated each mapping of transported aerosols and matched with AIRS swaths? If I understand well, all trajectories over the 14-22 June period have been used to reconstruct one single AIRS swath but why not focussing on a day-by-day basis, i.e. considering only trajectory calculations from the CALIOP tracks on the same day of the AIRS swaths? The number of CALIOP profiles used to generate the vertical profile of each AIRS swath is given in the figure caption for figure 6. The different CALIOP swaths capture different parts of the sub-clouds. Hence, using CALIOP swaths of several days, instead of only one day, leads to better coverage of the SO2 clouds in the AIRS swaths. This is illustrated in the new concept figure (Fig. 1). This would possibly limit effects of aerosol sedimentation (as a result of growth and coalescence) that can bias the mapping if trajectories are calculated on a too long period. Sedimentation rates of fine mode particles are too short to influence our data in the time-frame and vertical resolution used here (Please see comments above).

Figure 6: Not all the labels are visible on fig. 6a because of the colour choice. 11 AIRS swaths are labelled in fig. 6a but only 9 are shown on the top left list. For figs. 6b, c, d I am wondering if using a log-scale will make all profiles corresponding to the AIRS swaths in fig. 6a more visible.

We did not compile vertical profiles for those swaths, since the volcanic SO2 concentration was too low and no CALIOP data could be matched with these AIRS swaths. We have now added this information to the figure caption. We changed the label fonts to make it more accessible to the reader.

The authors have chosen the 18-19 June for the application of their method. However, at this stage, the SO2 plume is already geographically extended. Then, does the method properly capture the more localized and main injection event (i.e. June 15th in Haywood et al. and Lurton et al. studies)? This is of primary importance for robust initialization of models that account for SO2 chemical cycle producing sulphate particles.

Fig.2 shows all the eruptions from Sarychev, where different eruptions usually are found at different geographical locations. No SO_2 clouds are found outside the SO_2 swaths from AIRS shown in the timeframe of Fig. 2. This figure is the central time of the computations, containing all the Sarychev eruptions that have taken place before the 18-19 June, including June 15th. By using data from several days that we connect to different 18-19 June AIRS swaths by trajectory computations, we obtain a large number of profiles describing the different eruptions. That way the number of horizontally narrow, but vertically highly resolved CALIOP observations are increased. By this approach e.g. the AIRS swath containing most SO_2 was intersected by 34 CALIOP profiles (caption of Fig. 6).

All sub-clouds could not be fully captured before the 18th. Furthermore, observations of fresh dense SO2 clouds may result in saturation of the signal and underestimation of the SO2 mass so it is not necessarily a better approach to use data directly after the event.

Figure 7: I suggest also to indicate the date of the profiles in the mentioned studies.

We have added the dates of the other profiles to the figure caption.

P14 lines 12-13: I am not sure that if the model studies indeed missed the highest cloud of SO2 (i.e. around 15.5 km in fig.7) the consequence on the retrieved aerosol space-time distribution is significant. In Lurton et al. for instance, the agreement between the simulated aerosol profiles and in

situ observations are matching pretty well even if the initial SO2 injection is underestimated by considering the results in fig. 7 (red profile). This may be due to the coarse vertical resolution of global models in the lower stratosphere (~1km) which dilutes (or spreads out) the vertical distribution of the SO2 profile and limits the impact on the vertical profile of the subsequently produced aerosols. Also, the model results tend to show longer residence times than in the observations. I suggest the authors to mention this possibility.

We agree that it is important to mention this and have added the following sentence to the manuscript."…Nevertheless, the effects of missing the highest SO_2 clouds depend on the vertical resolution of the models in the stratosphere and how well they capture the circulation there. "

I would suggest to remove the sentence ("Their release…") We agree with the reviewer, and changed.

P15 lines 27-29: "Our deduced…sulphate particles." I suggest to remove or modify this statement since 1) the SO2 vertical distribution (which is visible only over the first weeks after the eruption) cannot be directly connected to the one of sulphate particles several months after the eruption as a result of vertical motion (Brewer-Dobson Circulation, sedimentation) sustained by the particles over such a long period

We believe that our statement is correct and wish to keep it with some modifications. Sarychev exploded in the season with minimum subsidence from the stratosphere and aerosol was mixed to the tropics in a shallow BD branch, prolonging the influence of the eruption. The Sarychev aerosol did not reach the upper BD-branch and were thus not subject to upwelling in that branch. Furthermore, it is evident from the figure 8 in Friberg 2018 that more than half of the volcanic aerosol reached above the 380 K isentrope. This is seen already in July, and the shallow BD-branch holds more than half of the volcanic aerosol load until December (when the stratospheric subsidence maximizes (Appenzeller et al. 1996). We decided to add information to the manuscript on this in both the discussion and conclusions sections, clarifying that half of the volcanic aerosol was located above the 380 K isentrope already in July. and 2) no modelling study has been conducted yet to quantify the effect of the new vertical profile of SO2 after the publication of the authors'work. We agree that modelling would be valuable. In an upcoming study, we will simulate aerosol formation and climate response with ours, and others, vertical SO2 profiles.

References:

Appenzeller C. , Holton J. R. , Rosenlof K. H . Seasonal variation of mass transport across the tropopause. J. Geophys. Res. 1996; 101: 15071–15078.

Fairlie, T. D., Vernier, J.-P., Natarajan, M., and Bedka, K. M.: Dispersion of the Nabro volcanic plume and its relation to the Asian summer monsoon, Atmos. Chem. Phys., 14, 7045–7057, https://doi.org/10.5194/acp-14-7045-2014, 2014.