Gorkavyi et al. (hereafter "Gteam") have three primary thrusts in this manuscript on the Raikoke volcano plume of 2019, to 1. characterize the stratospheric plume SO2 and sulfate evolution, 2. Introduce a limb-view retrieval artifact called the "arch effect," and 3. Follow a compact plume element termed a coherent circular cloud (CCC). These themes are carried out by invoking five different satellite-based measurement/retrieval products, three nadir-imaging and two vertical profiling.

The above themes, data, and methods are an appropriate fit for AMT. It is evident from the data presented by Gteam that the Raikoke volcanic cloud is scientifically important, even remarkable, and needs to be thoroughly characterized in the literature. Moreover, Gteam present a novel method for limbview aerosol profile handling to deal with a known limitation in the limb approach to quantifying perturbations that are inhomogeneous and/or geographically small in the context of instrument resolution.

My summary assessment of GTeam is that each of the three thrusts need substantial revision to qualify as publishable. In each respect, the material has either been presented in prior literature and not fully recognized herein, or the current presentation lacks clarity, validity, or motivation. Details of these primary concerns are next. Following these is a list of minor concerns. Technical concerns and questions are handled by comments and annotations to the manuscript, provided separately.

# Primary Concerns

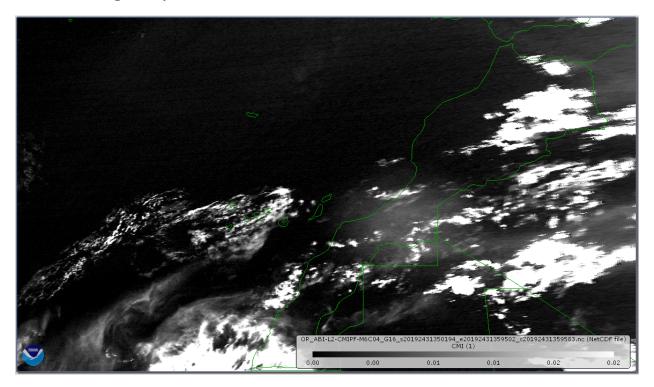
Gteam's foremost new content regards the "arch effect." Here they focus on a fundamental uncertainty with respect to limbview spectroscopy, that being the accuracy of retrieving extinction and altitude of an object that is small with respect to the instrument's (OMPS-LP in this case) line-of-sight field of view (FOV). The problem common to all such stratospheric limb viewers is that they peer through a roughly 200 km atmospheric path length (~180 km quoted by Gteam for OMPS-LP) composed of a wide altitude range in the Earth-centric reference frame. Generally, the assumption is made that any object in the view path occupies the entire FOV. To the extent that any object (like a meteorological cloud or aerosol plume) is smaller (vertically or horizontally) than the instrument's FOV, one or both of the retrieved extinction and feature altitude will be biased low. In highlighting the "arch effect," Gteam seem to be suggesting that the Raikoke sulfate plume presents a source of systematic error in OMPS-LP extinction and plume-height results. Hence the motivation for their focus on an adjustment to mitigate the "arch effect."

The "arch effect" argument, to be persuasive, requires at least one additional data item independent of OMPS LP to

characterize cloud- or plume-object height, vertical thickness, and horizontal extent all within the OMPS-LP FOV. I.e. OMPS-LP on its own is under-constrained for such an assessment. Gteam present the arch effect argument by invoking only OMPS-LP profile data. Moreover, the illustrative example of the arch effect is for a scene that has a Raikoke plume object that is ~40 days old. Even though it is presumed to be compact with respect to the OMPS LP FOV, this (and most other ) Raikoke plume elements have spread over distances greatly exceeding the OMPS LP FOV. With the data that Gteam present as an example of the arch effect—a consecutive sequence of OMPS LP aerosol profiles—it is not possible to know the true size of the Raikoke plume object precisely in the FOV. It is known, however, that the CCC the Gteam is following has horizontal

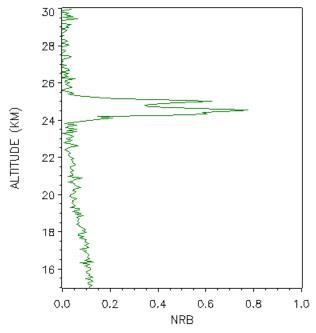
dimensions as great as ~600 km, based on the CALIOP curtain one day later, shown in Figure 13. It is conceivable that the 31 August OMPS example given for the arch effect was a case of the limb view sampling a peripheral part of the CCC, presenting a much smaller horizontal distance. But insufficient independent information is provided to inform the reader.

Exploring complementary data sets for the Gteam 31 August 2019 case of the arch effect, it becomes apparent that indeed the OMPS center slit sampled CCC they are tracking. The GOES East visible reflectance image shown below illuminates the CCC over coastal northwest Africa at OMPS measurement time: ~13:50 UTC. The visible meridional extent of the CCC is ~3.4 deg. Latitude, approximately 380 km. This is roughly doule the OMPS tangent path FOV.



The NASA micropulse lidar (mplnet.gsfc.nasa.gov) at Santa Cruz, Tenerife, (due west of the OMPS extinction-profile curtain in Figure 4) measured the CCC as it blew west. A snapshot of the normalized backscatter ratio profile at 22UTC 31 August is below.

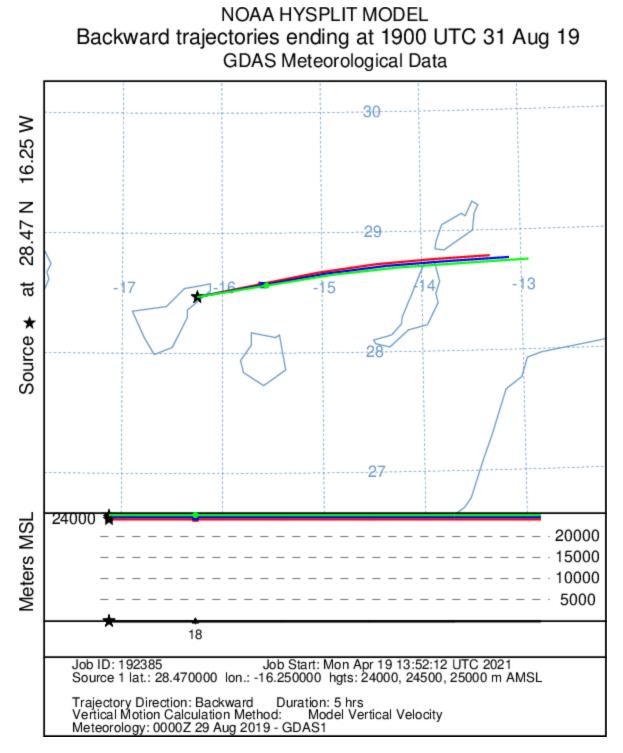
MPLNET Santa\_Cruz\_Tenerife 2019-08-31 22:10:30 V3\_L1\_NRB: NRB



Energy: 6.518 uJ (Set Point: 6.244 uJ) Box Temperature: 23.883 C (Set Point: 23.380 C) Detector Temperature: 26.841 C (Set Point: NaN C) Laser Temperature: 29.920 C (Set Point: NaN C) Solar Background: 4.320E-05 MHz

QA Flag: Good

Back trajectories to OMPS time 31 August show that the plume over Tenerife indeed passed over the OMPS curtain at OMPS measurement time.



The nadir GOES image and the precise lidar profile tightly constrain the plume geometry encountered by OMPS. Thus it appears that the CCC conditions encountered by OMPS are

vastly inconsistent with the limited horizontal cloud extent used to illustrate and motivate the arch effect principle. See the annotations to Gteam's schematic in the separate manuscript.

Given these two additional views of the CCC encountered by OMPS LP, it is largely uncertain as to how to interpret the OMPS extinction profiles in Figures 10 and 4. This does not appear to be a candidate for the arch effect, and the maximum extinction in the CCC—according to the example of it shown in the 1 September CALIOP curtain (Figure 13)—may be as much as an order of magnitude greater than retrieved by OMPS (CALIOP backscatter x lidar ratio of 50 sr). Considering all these factors, Gteam is encouraged to either clarify and bolster their arguments for this arch effect example, or find another case where it can be independently shown that a sub-FOV-filling plume element is sampled by OMPS.

If such an example can be demonstrated, it still seems unlikely that the aging/spreading Raikoke plume as sampled by OMPS LP justifies wholistic application of the arch-effect adjustment. The shear variety and complexity of plume presentations (e.g. the various CALIOP curtains illustrated by Gteam) to OMPS during the analysis time frame indicate that compact-to-thepoint-of-sub-FOV plume elements are rare. This includes the CCC and the meridionally broader Raikoke layers at lower altitudes do not meet the size-limited view as depicted in Figure 3. It is essential to characterize the schematic angles and "object" (also referred to as a "cloud") in terms of their geophysical horizontal and vertical dimensions. It is all important to know what size range of cloud in the along-track FOV direction creates the arch effect. It is difficult to know from the schematic how realistic the cloud object is, but it appears to be tiny in relation to the 180 km OMPS LP FOV.

To make a compelling argument for the arch effect, three things would have to be presented that are lacking. 1. A case study involving an independent plume-object physically characterized, 2. Such an element being located within the OMPS LP FOV (i.e. a space-time match), and 3. The plume element being demonstrably smaller than the FOV (as drawn schematically in Figure 3). An example of such a case study is provided in Penning de Vries et al. (2014) (cited by Gteam), who attempted to reconcile SCIAMACHY limb-scatter profiles of the Nabro volcano stratospheric plume with simultaneous nadir SO2 imagery. It is notable that the Nabro example was when the plume was less than two days old; demonstrably compact.

How is an "arch" identified in the OMPS data? How do we know when an arch shape might be geophysically accurate vs. one that is an artifact of a tiny cloud? Gteam describe a wholesale processing of the OMPS LP extinction profiles, applying the arch effect correction to the whole set. At least that is how I understood the method description. If this is the case, does that suggest that Gteam considers the arch effect a global vulnerability? Much more clarification of the correction application is needed.

ANSWER 1: The first 7 pages of the Primary Concerns are based on a misinterpretation of Figure 3. We agree with the reviewer that the CCC was much larger than 180 km (this is evident, for example, from the Figures 12-14). But the arch effect occurs if the horizontal length of the cloud is less than 1100 km, and not 180 km (for a cloud height of 25 km). We improved Figure 3 and made additional Figure 3b, which clearly demonstrate the effect of the arch or the observed decrease in the height of the cloud when it is displaced from the tangential point. We have added the following text to the article:

"The arch effect is observed when the length of the visible part of the cloud is less than ~1100 km (at a cloud height of 25 km). Figure 3b shows cloud  $F_0G_0$ , 1 km thick and 226 km long, centered above tangential point T. Due to the curvature of the globe, such a cloud has an observed thickness of 2 km (see Figure 3b). If we take a cloud 226 km long and with a real thickness of N km, then the observed cloud thickness will be N+1 km. Thus, the real average height of a thin (1-2 km) cloud is underestimated by 0.5-1 km even under the most optimal observation conditions. Consider a cloud FG, the center of which is displaced from the tangential point by 273 km (or by  $\varphi^{2.5}$  degrees). The real height of the FG cloud is 24-25 km, but its observed height varies from 13 to 22 km. If we consider the F<sub>0</sub>G cloud with a length of 499 km, then its real height above the earth's surface will be 24-25 km, and the observed height is 13-25 km. Let us take into account that the limb profiler assigns the latitude of the tangential point to any extended cloud. Therefore, a single cloud shown in Figure 3a in five different observed positions, instead of one real geographic latitude, receives several "observed" latitudes, which creates an arch effect. Let the region  $F_0G_0$  be a gap in a continuous cloud. Then this gap, together with the arch effect, will lead to a decrease in the maximum observed height of the cloud layer by 1 km (see Figure 3b)."

I could find no discussion of the vulnerability of OMPS-LP extinction profiles to saturation in the presence of optically thick aerosol plumes. Given the likelihood that the Raikoke sulfates presented widespread scenes of such optically dense conditions, as did previous eruption plumes like Nabro, Sarychev Peak, and Kasatochi (Fomm et al., JGR, 2014; Lurton et al., <u>https://doi.org/10.5194/acp-18-3223-2018</u>) it is important for Gteam to directly address if/how this issue was dealt with in their various visualizations of OMPS LP extinction and SAOD.

ANSWER 2: Although Raikoke was the largest volcanic eruption seen by OMPS LP, it is still considered a mid-size eruption and nowhere near the size of Pinatubo or even El Chichon eruption. As noted by Rieger et al., (2019) (see below), the "saturation bias" cited by Fromm et al, 2014 and Lurton et al. 2018 was caused by the OSIRIS V5.0 algorithm conservative approach in masking any data when the extinction exceeded  $2.5 \times 10^{-3}$  km<sup>-1</sup>. OMPS LP algorithm has no such restriction, and we are not aware of any detector saturation caused by this volcanic plume.

Rieger, L. A., Zawada, D. J., Bourassa, A. E., and Degenstein, D. A.: A multiwavelength retrieval approach for improved OSIRIS aerosol extinction retrievals, J. Geophys. Res.-Atmos., 124, 7286–7307, <u>https://doi.org/10.1029/2018JD029897</u>

Figure 14 and related discussion of CCC: Chouza et al (2020) https://doi.org/10.5194/acp-20-6821-2020 do a very similar tracking of the CCC (although they do not describe in CCC terms). It is not cited by Gteam. There appears to be a similarity in how the CCC is tracked but then a divergence occurs in late August. Because of the similarity and relevance to this paper, Gteam are encouraged to read Chouza et al. and evaluate the similarities and discrepancies between the two treatments of the CCC.

ANSWER 3: We have added the following text to the article:

"Figure 15 was published in December 2019 at AGU-2019 (Gorkavyi et al, 2019). On September 24, 2019 the CCC was observed by lidars in Hawaii (Chouza et al, 2020). Chouza et al (2020) traced the trajectory of this cloud back to July 17, 2019. Although the two studies were done independently, they came up with very similar results. Chouza et al (2020) consider this cloud as a Raikoke plume, but we prefer to call it CCC because it is a very small part of the Raikoke plume."

Kloss et al. (2021) present a SAOD analysis very similar to Figure 7. In fact, the same version of OMPS LP retrieval is used for both. I did not see in GTeam what I expected, an acknowledgement of this previously published analysis and a motivation for presenting the analysis anew. The only substantial differences in the Gteam plot are an addition of two markers for pyroCb sources and a slightly later endpoint. Neither of these visual differences are taken advantage of, from my reading. If I missed it, please advise. Regardless, it would be important for Gteam to assess whether the figure should stay and also cite Kloss et al. for the earlier rendition.

ANSWER 4: We have replaced Figure 7 with plots of OMPS LP aerosol extinction at three different altitudes, 14.5, 18.5, and 20.5 km.

## **Minor Concerns**

Note: Text below in quotation marks is directly from Gteam.

P4, L118: The schematic (Figure 3) shows a "feature" that is limited both horizontally and vertically. Why does this statement call out only the vertically limited aspect? What's the relative importance of limitations in the vertical versus horizontal?

ANSWER 5: See Answer 1

P4, L122: "If we believe that these lower altitude values do not represent a true aerosol signal..." How is this belief ascertained?

ANSWER 6: If we compare the lidar observations of the CCC (Figure 14) and the limb observations of this cloud (Figure 4), it becomes clear that all parts of the arch below 23 km are artifacts. There are many such examples. For example, PMCs are always 80 to 85 km high. According to the data of the limb sensor (see below, unpublished), these clouds have apparent heights of up to 25 km. All parts of the PMS observed below 80 km are artifacts from the arch effect.

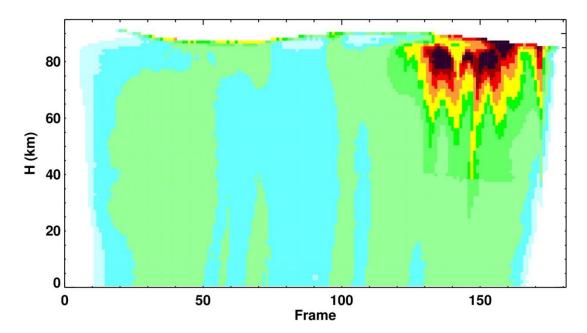


Figure 3 caption: The caption needs to describe the meaning of the horizontal solid and dash-dot line. Presumably these show

the OMPS instrument vertical FOV. But if so, why are the line styles different?

ANSWER 7: Now all lines are similar.

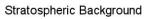
Figure 4. Please add an x-axis marking/labeling in deg. latitude, to give the reader a useful geographic refence frame.

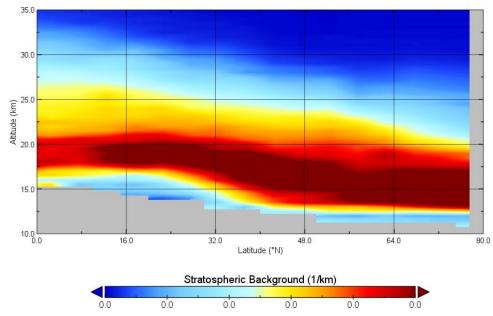
### ANSWER 8: Done

P6, L153: The Junge layer peaks much higher according to Kremser https://doi.org/10.1002/2015RG000511 citing Junge. An examination of CALIOP data shows lots of Raikoke aerosols widespread, lower down, on both sides of the CCC.

ANSWER 9: Our statement "The aerosol layer at an altitude of 18-20 km with a decrease to the north to 15-16 km is the Junge layer" (P6, L153)" is in full accordance with LARC/NASA data (see figure below) and does not contradict the classical works: "stratospheric aerosol occurs in a distinct layer between 15 and 25 km altitude with a peak near 20 km [Junge et al., 1961]" (Kremser et al, <u>https://doi.org/10.1002/2015RG000511</u>). Also we added in text next statement after Fig.4:

"Note that CCC has been the southernmost part of the Raikoke plume since late July 2019 (see section 3.3). During this period, only the usual Junge aerosol layer was located south of latitude 20°N."





P6, L158: Please define/describe "artifact density." Why is the term "density" used? Why not "extinction?" ANSWER 10: "Density" was replaced by "extinction".

P6, L157-161: This paragraph has several vague or unintuitive terms, such as "real," "artifact density," "cleaned." Please replace these terms or define them.

ANSWER 11: "real" was replaced by "true"; "density" -> "extinction"; "cleaned" -> "corrected".

P6, L158: How is an "isolated" feature determined from OMPS data alone? There is no discussion of complementary data (e.g. OMPS nadir) informing the limb data.

ANSWER 12: We use CALIOP (and SAGE – see new Fig. 10) as an additional source of information.

P7, L185: "The sensitivity of the satellite data we examined..." What satellite data? OMPS NM? According to Figures 5 and 7, the aerosols are detected much longer.

ANSWER 13: We improve our statement: "The sensitivity of the satellite data (OMPS LP, CALIOP) we examined is such that the CCC from the Raikoke volcano was observable for 3 months following the eruption and the increase in the Junge aerosol layer was observed even longer."

Figure 5 caption: "5: The daily zonal mean (45-85N) SO2 mass (assuming a cloud height of 13 km) and the aerosol extinction coefficient at 675 nm (13-18 km.)." This is 6 OMPS LP altitude bins. Which extinction is it? Max, min, average?

ANSWER 14: We have clarified the caption for Fig. 5: "the average aerosol extinction coefficient at 675 nm (summed up over 13-18 km and divided by 6 (km) to get the average extinction coefficient)."

Figure 5 caption: "ash fallout" How does the reader know how much ash fallout is observed? What does ash fallout have to do with SO2? Please explain.

ANSWER 15: The statement about "Ash fallout" was deleted.

Figure 5 caption: "...extinction starts to decrease due to gravitational sedimentation, but very slowly (from OMPS LP and NM data)" What does this mean? How do these inform about sedimentation? Sedimentation is one of several processes that an diminish extinction. How is sedimentation demonstrated herein?

ANSWER 16: We have clarified: "decrease due to gravitational sedimentation and other processes"

Figure 6 caption: "main plume" How is the "main plume" defined?" This is the first invocation of "main plume."

ANSWER 17: We have clarified: "the main plume (45-85°N)"

Figure 6 caption: "south branch" Is "south branch" synonymous with CCC, or is it more general? Perhaps just stick with "CCC" if they are one and the same.

ANSWER 18: "south branch" was replaced by CCC.

P9, L227: "After that, the detection limit of the instrument..." What id the detection limit? Please state the value.

ANSWER 19: We added next statement: "OMPS in the stratosphere can typically detect 0.2-0.3 DU of SO2".

P9, L239: No pyroCbs are mentioned by Kloss et al. as far as I can tell. If they are, please provide some more detail. If not, what is the justification for adding these symbols to the figure? There is no evident increase of SAOD clearly attributable to these symbols in the plot.

ANSWER 20: We are fixed the Fig. 7 with a caption and have clarified:

"In addition, Figures 7b and c also show the aerosol transport to subtropics and tropics at higher altitudes. Increased aerosol loading in the lower stratosphere can also be attributed to two pyroCumulonimbus (pyroCb) events that took place before and after the eruption, Alberta fires (June 18) and Siberian fires (July 2) (Kloss et al., 2021). OMPS LP detected both plumes in the stratosphere at 12-13 km, although it became difficult to separate them from Raikoke plume once it spread around the NH."

P10, L241: "The top panel in Fig. 8 shows that the maximum altitudes of the plume are around 25 km, when the plume

penetrates the tropics." How is "when" shown? Figure 7 seems to show that Raikoke material only got to the subtropics.

ANSWER 21: We have replaced Figure 7 with plots of OMPS LP aerosol extinction at three different altitudes, 14.5, 18.5, and 20.5 km. The new figure clearly shows the plume transport to the tropics at altitudes 18.5 and 20.5 km.

Figure 8(a): Please explain the two apparent outliers of ~25 km layers over Northern Europe. They seem to be all by themselves; no indication of a ramp up to those altitudes from nearby layer observations.

ANSWER 22: We agree with the reviewer that the original figure was confusing. We have now modified the figure to show the day number instead of the maximum altitude since the reader can match the altitude in Fig8b with the location and day number in Fig 8a. In addition, we changed the maximum altitude period to only show the first four months of the plume altitude, which is 150 days since June. The maximum altitude estimate uncertainties increases as the plume move around and subside following the first 150 days. Fig 8a period now matches Fig8b white line, which wasn't the case in the previous version. The only difference is that Fig8b shows the location of the plume every two days for 150 days, while Fig8b is plotted every day for the same period. We have now modified the text accordingly.

Figure 8(a) caption: What is the number of days shown? Span of time? Are all the reported dates consecutive, or are there gaps? If there are significant gaps, these dates should be mentioned.

ANSWER 23: See our reply to the previous comment. The new caption and figure are clearly showing the period, which is 150 days since June, plotted every other day.

Figure 8 caption: The sAOD panel is not described.

ANSWER 24: We have added the following to the figure's caption "Figure 8(b): (top) Stratospheric aerosol optical depth (x  $10^3$ , sAOD) at 675 nm for latitudes and period similar to Figure 8b. The sAOD is derived by integrating aerosol extinction profiles above the tropopause to 35 km"

Figure 8(b) caption: How are the data comprising the white line determined? The white line doesn't conform closely to the color shading. I.e. it crosses extinction contours in time. Please explain fully.

ANSWER 25: The plume altitude is derived using the OMPS LP cloud algorithm, which can identify enhanced aerosol layers in the stratosphere. We have now added this to the text. The reason for the mismatch between the white line and the extinction contour lines is the color table used, which favors larger aerosol extinction values. We have now changed the color table to better show small extinction values.

Figure 9 caption: The Junge layer is higher than the range indicated here.

ANSWER 26: see Answer 9.

P16, L309: Specifically, how were the CCC detections done out to 22 September? I could not find a definitive explanation.

ANSWER 27: We added after Fig.13 (new): "Using the data of CALIOP, OMPS LP and NM as well as OMI"

Figure 14 caption: "OMI" is mentioned here for the first time as part of this analysis, but it is not mentioned in the Data and Methods section. This should be done, unless OMI is not part of the present analysis.

ANSWER 28: See ANSWER 27, also we added to end 2.1: "We also used OMI data as an additional source."

P18, L349: "A carbonaceous aerosol plume associated with wild fires in British Columbia in August 2017 reached the stratosphere a few days following the initial injection above the tropopause..." Confusing. It is stated that the plume was injected above the tropopause, but only reached the stratosphere a few days later. Both clauses are attributed to Torres et al. Please clarify.

ANSWER 29: The text has been changed: "A carbonaceous aerosol plume associated with wild fires in British Columbia in August 2017 reached the stratosphere a few days following the initial injection and resulting from selflofting triggered by solar heating (Torres et al., 2020; see similar effect for Australian fires - Khaykin et al., 2020)."

P18, L352: "...19 to 26 kilometers is observed for a cloud consisting of sulfate aerosol." From the material presented to this point, how does the reader know it is sulfate? If compositional information has not been provided, a citation is needed. Alternatively, the paper could just state that sulfate is assumed.

ANSWER 30: The text has been changed: "observed for a cloud presumably consisting of sulfate aerosol"

P18, L376: What are "large-scale mixing events?" Please explain using a meteorological perspective.

ANSWER 31: The text has been changed: "large-scale mixing events" -> "turbulent mixing"

P18, L279: "SO2 reduction" is vague and general. Provide more detail to this description, such as "e-folding time"

ANSWER 32: The text has been changed: "SO2 reduction" -> "e-folding time"

P18, L383: "...the CCC circled the globe almost three times at the latitude of 30oN..." The CCC ended up almost 10 deg. south of this according to figures herein. Perhaps elaborate a little more on the CCC's slow movement even south of 30.

ANSWER 33: We added: "(during September, the CCC shifted south to 15-20°N)"

P18, L383: "...increased its height from 19 to 28 km." "28" is higher than shown if Fig. 14. The text in the body of the paper only gives pressure and theta. Suggestion: enhance the description of the endpoint altitude in the main text and state "26 km" or whatever actual number the authors think is represented by the figure.

#### ANSWER 34: The text has been changed: "28 km" -> "26 km"

P18, L387-389: This sentence is confusing, combining 3 ideas: arch effect, 45 km background, and aerosol that can pollute the background. Please flesh out these points more exhaustively.

ANSWER 35: We transfer this sentence to end 2.2 with clarification.

More generally, the Conclusion section is too thin, and doesn't flow logically from points made in the body. A thorough rework of this section is called for.

ANSWER 36: The conclusion has been improved and expanded.

ANSWER 37: We have corrected all errors and took into account all the comments of the reviewer made in the comments to the text of the article.