Response to anonymous referee 1's comments

First of all, the authors acknowledge the referee for his constructive comments and suggestions. In the revised manuscript, the authors made an effort to improve the quality of the English and the figures. The English of the revised manuscript was checked by a native English speaker. The modifications are indicated by italic and red bold fonts in the revised manuscript.

- **Specific points:**

Referee 1 : I would suggest adding in the introduction some recent works that have shown how significant explosive volcanic eruptions can be for the stratospheric dynamics, by affecting large scale trace species transport and age of air, via radiative perturbations due to volcanic aerosols (Ray et al., 2014). Although the largest emphasis has been given to major tropical eruptions and their induced dynamical effects (Pitari et al., 2016a), extratropical eruptions in the last 15 years may also have had a significant role in lower stratospheric trends of key dynamical quantities (Kremser et al., 2016).

Authors **:** This part of the introduction in link to the helpful role of the explosive volcanic eruption on the stratospheric dynamic was improved by including the references suggested by the referee 1 (See revised manuscript).

Referee 1 : Page 4, Lines 11-15: Regarding QBO effects on mid-latitude transport of the volcanic plume, I suggest citing Pitari et al. (2016b). In this paper, the e-folding time of the stratospheric sulfate plume caused by four major past tropical eruptions has been studied in a modeling experiment focusing on the QBO role. Their conclusion is in agreement with those in the Trepte and Hitchman (1992) paper.

Authors : We thank the referee 1 for this relevant reference. As suggested by the referee 1, this reference was included in the revised manuscript.

Referee 1 : Page 10, Lines 16-27: Again, the only studies cited here are the ones from Trepte in 1992-1993. I feel that the addition in the discussion of Pitari et al. (2016b) would enrich the discussion by offering further evidence of the behaviour of the aerosol plume under different QBO regimes.

Authors : We thank the referee 1 for this suggestion. The publication mentioned by the referee 1was in included in the revised manuscript.

Referee 1 : Overall, the authors need to discuss much better the differences between Lidar, OMPS and CALIOP in paragraph 3.2.1. In particular, in Fig. 6 and Fig. 7 the differences are far too big between the panels. CALIOP doesn't see anything at all in the June-July period. A proper, good explanation should be given by the authors, I do not think a simple remark on vertical resolution is sufficient.

Authors : The reasons for these discrepancies may be multiple but effects due to different spatial samplings cannot be excluded. As mentioned by the referee 1, the difference in vertical resolution between ground-based LiDAR and satellites (OMPS, CALIOP) which could be one possible cause at the origin of discrepancies. The discrepancies between the satellites and ground-based LiDAR could be significant when the difference of vertical resolution is high between these devices. We note that the vertical resolution of OMPS is 10 times lower than the ground-based LiDAR with 0.15 km and 1.5 km respectively (Jaross et al., 2014)[1]. Thus, the structures of the plume look smoother than those obtained from the ground-based LiDAR. In the case of CALIOP where the vertical resolution is better (~ 3 times less to the ground-based LiDAR), the differences in the structure of the plume are less.

Moreover, the discrepancies existing between results presented in this study could be also due to horizontal resolution or different measurement techniques. Unlike satellite experiments that allow global observations, a ground-based LiDAR system is able to derive aerosols characteristics at a specific location. OMPS views the Earth's limb looking backward along the orbit track of approximately 125 km with a horizontal resolution of 50 km. It is difficult for OMPS to detect with accuracy small amount of aerosol at a local point with these weak vertical and horizontal resolutions. It is for this reason that the structure of the plume observed since July is not in agreement with the ground-based LiDAR. The CALIOP figure is realized from weekly-averaged profiles within $\pm 5°$ latitude and $\pm 50°$ longitude around the Reunion site. Given that the weak horizontal resolution of CALIOP (500 km) (Vernier et al., 2011)[2], it is consistent to observe weaker values than the ground-based LiDAR.

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[1] Jaross et al (2014) : OMPS Limb Profiler instrument performance assessment, J. Geophys. Res. Atmos., 119, 4399–4412, doi:10.1002/2013JD020482.

[2] Vernier et al. (2011) : Major influence of tropical volcanic eruptions on the stratospheric aerosol layer during the last decade, Geophys. Res. Lett., 38, L12807,doi:10.1029/2011GL047563

As we discussed in Section 4, the dynamical context induced an inhomogeneity of the plume over the Reunion site. In particular during the June-July period, the Reunion site is impacted by air masses come from the Calbuco and also by others air masses (Fig. 11 and 12). This inhomogeneity of the plume could lead to incorrect identification of the volcanic aerosols by the satellites. Vernier et al. (2011) reported that it is possible for solid aerosols such as ash to be incorrectly identified and to be then removed.

We think that the discrepancies between the satellites and ground-based LiDAR at the Reunion site could be due to the resolution of the satellite observations and the inhomogeneity of the plume. This discussion was developed in the revised manuscript.

Referee 1 : Page 13: the authors should provide also the other parameter for the log-normal distribution (sigma), to give a better idea of the shape. Also, at 2 m a significant value for the distribution seems to be present in Fig. 8. This value does not fit in the lognormal distribution, and the authors should discuss at least why this value has been ignored, and if it could point out to a coarse mode that is not properly detected because 2 m is the largest class detected.

Authors : We now provide the lognormal parameters (No, Median radius and sigma). We agree with the reviewer that the value at 2 μm has not been sufficiently discussed and can be misleading for the reader. It is not clear why a secondary (coarse) mode deflecting from the unimodal distribution is apparent. It might reflect the signature of remaining ash a few weeks after the eruption. For the Pinatubo aerosol cloud a coarse mode was clearly highlighted and possibly attributed to ash particles (mentioned in our initial text p13). In the Calbuco specific case the LOAC OPC does not properly detect the very low concentrations for sizes > 2 μm and we cannot provide the whole distribution of this specific coarse mode which amplitude is not significant in comparison with the main mode. As a result we have decided to remove the 2-μm value but we add a discussion in the text (See in the revised manuscript).

Referee 1 : Page 13 and Figure 9: First of all, I would suggest limiting the range in Figure 9 to the upper troposphere and the stratosphere. The lowest values just create noise and enlarge the x-axes scale. Furthermore, as the authors somewhat point out, a single background profile cannot be used to draw any conclusion. Either the authors find more profiles to average as background, or the figure and the conclusions the authors draw from it should better highlight how limited the comparison is, or removed altogether.

Authors : The suggestion mentioned by the referee 1 was included in the revised manuscript. Figure 9 (Figure 10, in the revised manuscript) was re-plotted with y-axes scale ranging from 10 to 35 km altitude. The referee 1 is right to point out on the fact that only one profile are presented in Figure 9 for the period before the Calbuco eruption. It is for this reason that the term of background profile is not really appropriated. As we stated in the manuscript, few in situ observations are available in the tropical region to provide a reference state of the background aerosol content. An effort was realized to make in situ (LOAC) and ground-based (LiDAR) observations frequently over Reunion since 2013 in order to help to reduce this lack of observations in the tropical region. Figure 9 is also a good way to inform the community to the development of an in situ database over tropical site such as Reunion Island.

It is for the reasons mentioned previously that we decided to keep all the profiles on Figure 9 and to be more careful in our conclusions.

Referee 1 : Page 15, section 4.2: I feel this section could be largely improved. The authors should show what they have done, as described in lines 21-23, in at least one figure.

Authors : As suggested by the referee 1, this section was rewritten in the revised manuscript. The discussion on the removal processes are organized following two points: (i) dabatic stratrosphere-troposphere exchange at extratropical latitudes; (ii) sedimentation.

The line mentioned by the referee 1 was removed. Indeed, the reference cited was not appropriated.

- **Syntax comments**

Referee 1 : Page 5, line 18: I would suggest rephrasing this because it makes no sense. Maybe what the authors meant to say is "The method involved in: : :". Also the following phrases should be rewritten in a way that makes them easier to read

Authors : The referee 1 is absolutely right. This sentence was re-written in the revised manuscript.

Referee 1 : Page 5, line 25: it's "called", not "call".

Authors : It was corrected in the revised manuscript.

Referee 1 : Page 6, line 9: either a "bigger" or "smaller" is missing.

Authors : It was added in the revised manuscript

Referee 1 : Page 7, line 3: please rephrase ("relevant for the monitoring of").

Authors : It was corrected in the revised manuscript

Referee 1 : Page 8, lines 23: "corresponding" .

Authors : It was corrected in the revised manuscript

Referee 1 : Page 9, line 33: "up to" instead of "until".

Authors : It was corrected in the revised manuscript

Referee 1 : Page 10, line 10: "northern" is spelled wrong.

Authors : The sentence mentioned by the referee 1 was removed in the revised manuscript.

Referee 1 : Page 10, lines 21-24: please rephrase in proper english

Authors : This part of the manuscript was re-written

- **Figure comments :**

Referee 1 : Fig. 1: It is almost impossible to read the colorbar, and the whole figure seems a bit out of focus. Less numbers, but bigger, are needed. The map is also impossible to read. I suggest splitting the figures in three panels: a) with the map b) the cross section (with all the numbers made more readable) and c) the Brightness Temperature Difference (again, much larger).

Authors : The quality of this figure was improved in the revised manuscript. We note that this figure become figure 3 in the revised manuscript.

Referee 1 : Fig. 2: Part of the bottom of the figure is cut (Time). The caption needs to be much more detailed. Also, always call it "Altitude" over all figures (as in Fig. 1 and 3) and not "Height".

Authors : This figure was re-plotted as suggested by the referee 1. Moreover, the caption was rewritten in the revised manuscript.

Referee 1 : Fig. 4: This figure has very poor quality. Colorbar numbers are again impossible to read. Enlarge the panels and get rid of all that white space.

Authors : The quality of this figure was improved in the revised manuscript. Moreover, we reduced the white space between panels. Moreover, we divided the figure 4 in two parts: (i) One part for the CALIOP observations during May 2015 (Figure 4, in the revised manuscript);

(ii) Another part dedicated to CALIOP observations during June and August 2015 (New Figure 5, in the revised manuscript).

Referee 1 : Fig. 5: Why are some of the red points not connected to the others with a red line (Oct 2014 and Feb 2016). What do the error bars represent? The caption needs to be improved. Instead of marking with the dashed line the beginning of 2015, I would suggest making with the same line the exact day of the eruption.

Authors : The large dots represent the monthly averaged of sAOD from OMPS and LiDAR observations. The error bars associated represent the standard deviation. The date pointing out by the referee 1 refer to months where we are only one LiDAR profile. As consequence, it is impossible to calculate a standard deviation for these two months. These clarification was included in the revised manuscript.

In order to avoid to encumber the Figure, we decided to indicate the day of the Calbuco eruption by a blue arrow (See revised manuscript).

Referee 1 : Fig. 6 and 7: There is really no point in having all that white space between panels. The figures could be expanded.

Authors : A suggested by the referee 1, these figure were expanded and the white space between the panel was reduced.

Refeere 1 : Fig. 10: The continents are almost not visible at all: The contours should be thicker and more visible.

Authors : The contours of the continents are more visible in the revised manuscript.

Response to anonymous referee 2's comments

- **General points:**

Referee 2 : the Calbuco eruption in April and 2) the reaching of the record ozone hole size in October. Based on the results of the SD-WACCM* and FR-WACCM** simulations, Solomon et al. (2016) and Ivy et al. (2017) declared that the first event (eruption) led to the second one. In other words, according to Solomon et al. (2016) and Ivy et al. (2017), the Calbuco aerosol plume (including various volcanic gas emissions) penetrated the polar vortex and caused the record Antarctic ozone hole size after the eruption. On the other hand, according to the findings presented by the authors (Bègue et al., 2017), the Calbuco aerosol plume could not penetrate the polar vortex and lead to additional ozone depletion, because the plume was confined between the subtropical barrier and polar vortex. Since the results of the SD-WACCM and FR-WACCM simulations were published before, the above-mentioned contradiction between the conclusions made by two different research groups should be considered, analyzed, and discussed by the authors of the paper under consideration (Bègue et al., 2017).

*SD-WACCM is the specified dynamics Whole Atmosphere Community Climate Model
**FR-WACCM is the free-running Whole Atmosphere Community Climate Model

Authors : We thank the referee 2 for these two relevant papers. The results presented in our study are not in contradiction to the works of Solomon et al. (2016) and Ivy et al. (2017). Our study is based on isentropic analysis of the volcanic plume. In particular, we discussed on the transport of the volcanic plume exclusively at 400 K which correspond to the isentropic level where the plume is observed at Reunion. Figure 4 and 5 reveal that the meridional transport of the plume occurred between 12 and 20 km. As a consequence, the transport of the Calbuco plume at another isentropic level associate to another pathways described in our study is possible. Figure 5b reveals also the possibility to the Calbuco plume to penetrate the polar vortex at the end of August 2015. This assumption seems to be consistent to the works reported by Ivy et al. (2017) and Solomon et al. (2016). Based on SD-WACCM (Specified Dynamics-Whole Atmosphere Community Climate Model) model and balloon observations at Syowa (69°S; 34.58°E), Solomon et al. (2016) discussed on the impact of the Calbuco plume on the deepest Antarctic ozone depletion observed in October 2015. According to CALIOP observations present in our study (Fig. 4 and 5), we think that the Calbuco aerosol plume

penetrated the polar vortex more likely at isentropic level lower than 400 K. We note that the altitude which the plume penetrated the polar vortex is not reported by Solomon et al. (2016) and Ivy et al. (2017). In a way, we think that our study can come in complement to the two studies cited by the referee 2.

In a furthercoming study, we wish to analysis in details the mechanisms of transport of the Calbuco plume in the southern hemisphere. This discussion was developed in the revised manuscript.

Referee 2 : The paper cannot be published in its current form due to the poor quality of English and figures. When reading the paper, it was almost not possible to understand the meaning of some phrases and sentences. The text of the paper contains a lot of grammar mistakes and syntax errors.
The quality of all figures should also be improved. Figures 1 and 4 seem to be out of focus. The font sizes of letters and numerical symbols in Figures 1, 3, 4, 10, and 11 should be enlarged, if possible. Figures 2, 5, 6, 7, 10, and 11 should have appropriate fonts to be more readable.

Authors : We understand the point of the view of the referee 2 and also the importance of the quality of the English for an article. As a consequence, we asked a native English speaker to check and improve the quality of the paper. Thus, some parts of the manuscript was rewritten. Moreover, the quality of the Figure was also improved as suggested by the referee.

Below are my several minor comments and suggestions concerning the text content (using Sections 1 – 3.1.1 as an example). To help the authors, I also attached the highlighted discussion paper with my concerns for Abstract and Sections 1 – 3.1.1. I suppose that there is no need to reply to every comment on errors and omissions in English grammar, because the text of the paper should be substantially improved.

Authors : As proposed by the referee we do not reply to every comment on error and omissions in English grammar in this response file. However, all the suggestions pointing out by the referee 2 concerning the English grammar were included in the revised manuscript. We report on this file the response to specific scientific points mentioned by the referee 2.

- **Minor and technical points:**

Referee 2 : lines 1–3: Perhaps it would be better to write "the 2015 Calbuco eruption" instead of "the Calbuco eruption in April 2015". (No other Calbuco eruptions occurred in 2015). My suggestion for the title: "Long-range isentropic transport of stratospheric aerosols in the Southern Hemisphere following the 2015 Calbuco eruption"

Authors : We understand the point of view of the referee 2. As a consequence, this modification was included in the revised manuscript.

Referee 2 : line 26: "21_S" –> "21.1 _S" rewritten to clarify the meaning. Where do organic compounds and meteoritic dust contribute to the Jungle layer composition: in the lower stratosphere, in the upper stratosphere, or in both parts of the stratosphere?

Authors : The sentence was rewritten in the revised manuscript.

Referee 2 : line 31: "The injected SO_2 is then"? What part (or layer) of the atmosphere is SO_2 injected into? For example, it could be: "The injected into the stratosphere SO_2 is then". Anyway, please clarify the situation.

Authors : This sentence was clarified as suggested by the referee 2 in the revised manuscript

Referee 2 : line 14: "3,5 K" –> "3.5 K", the text fragment "near the aerosol peak" should be clarified. What does the aerosol peak mean?

Authors : This sentence was rewritten in the revised manuscript.

Referee 2 : line 23: Concerning the reference (Hofmann et al., 2009)... This decadal trend in stratospheric ozone loading (in the 2002-2012 period) was also determined over Garmisch-Partenkirchen (Germany) and Tomsk (Western Siberia, Russia), and can be seen from articles by Trickl et al. (2013) and Zuev et al. (2017), respectively.

Authors : We thank the referee 2 for these relevant references. We added these references in the revised manuscript.

Referee 2 : lines 32–33: "contributed to counterbalance the global warming"? It is not clear to what extent these recurrent "minor" volcanic eruptions (in comparison to the Pinatubo eruption) contributed to counterbalance the global warming.

Authors : This sentence was rewritten in the revised manuscript.

Referee 2 : line 9: "meridional transport"? What is the transport (aerosol transport or air mass transport in total)? May be "the meridional air mass transport" could be more correct?

Authors : This sentence was clarified in the revised manuscript.

Referee 2 : lines 19–21: This sentence should be rewritten to explain more clearly the aim of the study.

Authors : This sentence was clarified in the revised manuscript.

Referee 2 : lines 1–4: I am confused about the meaning of this sentence. "Lidar systems" and "measurements" are intercompared in the sentence. Otherwise speaking, "measurements" cannot be among "lidar systems". My suggestion for this sentence: "Among measurement data from four lidar systems operated during this campaign, we used data from the Differential Absorption Lidar (DIAL) system built for stratospheric ozone monitoring (Baray et al., 2013)."

Authors : This sentence was clarified as suggested by the referee 2.

Referee 2 : line 18: "method"? What is the method about? It is not clear. There was no description of any methods above. Please clarify it.

Authors : This part of the manuscript was rewritten in the revised manuscript.

Referee 2 : lines 18–19: My suggestion for this sentence: "The aerosol measurement method described by Klett (1981) involves obtaining the aerosol extinction and backscatter coefficient from Rayleigh-Mie lidar measurements."

Authors : This part of the manuscript was also rewritten in the revised manuscript.

Referee 2 : line 21: "Several parameters are needed:"? What are the parameters needed for (or to)? Please clarify it. It should be written: "Several parameters are needed for" or "Several parameters are needed to".

Authors : This sentence was clarified in the revised manuscript.

Referee 2 : lines 22–23: "The profile is completed by the Arletty model"?? What is this profile? Is this profile of temperature or pressure, or both of them? The verb "completed" should be substituted by an appropriate verb. Please clarify the meaning anyway. What

is this (Arletty) model about? The model description and corresponding reference are required.

Authors : This section was improved in the revised manuscript.

Referee 2 : line 24: "The second parameter"? Is this parameter really the second??? The fact is that TWO parameters (temperature and pressure) are already mentioned in the previous sentence. The same remark is for the "third" parameter (altitude) on line 27.

Authors : This section was improved in the revised manuscript.

Referee 2 : line 25: It should be "also called the lidar ratio" instead of "also call the lidar coefficient". Please rewrite the sentence in accordance with the following definition at the website: http://glossary.ametsoc.org/wiki/Lidar_ratio It would be better to write "The ratio value depends on" instead of "It depends of".

Authors : This section was rewritten as suggested by the referee 2.

Referee 2 : line 26: Perhaps, it would be better to write "Under the background stratospheric aerosol conditions," instead of "In the case of background stratospheric aerosol,". "in the literature"? Some references are required here.

Authors : This sentence was rewritten and we added references as suggested by the referee 2.

Referee 2 : line 6: (Vignelles 2017). This is an incorrect reference.

Authors : It was corrected in the revised manuscript

Referee 2 : line 7: What kind of uncertainties is meant here and further? Please clarify it.
Authors : Authors talked about the uncertainties on the determination of the concentration from LOAC device. In particular, we talked about technical context where the measurements ae realized which could lead to uncertainties on the concentration estimation.

Referee 2 : lines 10–11: Perhaps it would be better to write "are governed by Poisson statistics and estimated" instead of "is dominated by Poisson law statistics estimated".

Authors : It was corrected as suggested by the referee 2

Referee 2 : line 23: Is it a CALIPSO orbit?

Authors : The referee 2 is right. We talked about a CALIPSO orbit.

Referee 2 : line 26: "full zonal mean"? According to Vernier et al. (2009) it should be the word "means" (not mean). What are the full zonal means? Are these means of: the scattering ratio, the depolarization ratio, or both of them? Please clarify it.

Authors : This typo error was corrected in the revised manuscript. This calculation was applied to the scattering ratio (this information was reported in the revised manuscript.).

Referee 2 : lines 1–2: "the Meteorological Operational satellite (MetOp-A and MetOp-B) launched in October 2006"? One IASI is on board two satellites, isn't it? Which meteorological operational satellite do you mean? MetOp-A and MetOp-B were launched in 2006 and 2012, respectively. Please clarify it. See, for exapmle: Divakarla, M., et al. (2014), The CrIMSS EDR Algorithm: Characterization, Optimization, and Validation, J. Geophys.Res. Atmos., 119, 4953–4977, doi:10.1002/2013JD020438.

Authors: **The** referee 2 is right and this point was clarified in the revised manuscript.

Referee 2 : line 15: What is the spectrum? Please clarify it.

Authors: The RT model used for the LP (Limb Profiler) was initially developed by Herman et al., (1994[3], 1995)[4], and has been tuned and optimized for limb studies by Rault (2005)[5]. The radiance term used in this paper refer to the scattering solar radiation used to infer information on the ozone concentration vertical profiles and also aerosol extinction profiles (Taha et al., 2011)[6]. The spectrum term used in the initial manuscript refer to spectral band extends from UV to visible wavelength. The aerosol extinction and aerosol size distribution are retrieved

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[3] Herman, B. M., Ben-David, A., and Thome, K. J.: Numerical technique for solving the radiative transfer equation for a spherical shell atmosphere, Appl. Optics, 33, 1760–1770, 1994.

[4] Herman, B. M., Caudill, T. R., Flittner, D. E., Thome, K. J., and Ben-David, A.: Comparison of the Gauss-Seidel spherical polarized radiative transfer code with other radiative transfer codes, Appl. Optics, 34, 4563–4572, 1995.

[5] Rault, D. F.: Ozone profile retrieval from Stratospheric Aerosol and Gas Experiment (SAGE III) limb scatter measurements, J. Geophys.Res., 110, D09309, doi:10.1029/2004JD004970, 2005.

[6] Taha, G., Rault, D. F., Loughman, R. P., Bourassa, A. E., & Savigny, C. V. (2011). SCIAMACHY stratospheric aerosol extinction profile retrieval using the OMPS/LP algorithm. *Atmospheric Measurement Techniques*, *4*(3), 547-556.

using spectral channels with weak gaseous absorption. The description of OMPS was rewritten in the revised manuscript.

Referee 2 : lines 2–3: This sentence must be rewritten to be understandable. My suggestion for this sentence: "The SO2 e-folding time was estimated to be about 11 days that is in agreement with the time value reported for the 2009 Sarychev volcanic eruption"

Authors : This sentence was rewritten as suggested by the referee 2.

Referee 2 : line 12: It would be better to write "injected sulfur" or "stratospheric aerosol loading" instead of "produced aerosol loading". "Figure 2 also depicts the maximum altitude of the SO2 plume"?? This description of Figure 2 and the Figure 2 caption contradict one another. Because the maximum altitude of the SO2 PLUME and the maximum altitude of the SO2 MASS are different matters. Please clarify it.

Authors : The referee 2 is right. As a consequence, this point was clarified in the revised manuscript.

Referee 2 : lines 20–21: "the plume is mainly located over the Atlantic Ocean near the east coast of South Africa"? How is it possible? It definitely should be "the west coast" instead of "the east coast".

Authors : The authors would talk about Indian Ocean and not Atlantic Ocean. This point was corrected in the revised manuscript.

Response to anonymous referee 3's comments

- **Specific points:**

Referee 3 : Firstly, I consider use of the word "isentropic" within the phrase "long range isentropic transport" in the title, and at other points in the manuscript, to be inappropriate. The topic of the paper is to assess the long-range transport of the plume – but although the long-range transport of the constituents within an airmass might generally be expected to be isentropic, for a volcanic plume this is very often not the case, due to sedimentation of ash particles (with also any accomodated sulphur) or from growth of the particles within the plume (if the plume is long-lived enough and has sufficient growth).

The vertical profile measurements suggest some elements of the plume extend down to several kilometers below the main altitude of the SO2. Whether this is indicative of some separation of the plume (related to the ash) is not clear from this analysis. Nevertheless, this issue of volcanic plumes in general not necessarily being isentropic in my opinion means it would best to avoid the word "isentropic" within the phrase "Long-range transport" (unless the analysis specifically shows this to be the case). For this reason, the first non-minor revision I ask is for the authors to remove the word "isentropic" from the title.

Authors : We understand the point of view of the referee 3, as a consequence, the term of « isentropic » was removed in the revised manuscript.

Referee 3 : The authors state with certainty that the aerosol particle size distribution is unimodal, but the OPC only measures particles which are larger than 250nm, with the behaviour of particles smaller than that size simply not monitored. And yet the particles measured by the OPC are really only measuring those particles in this "shoulder" of an accumulation mode, which may only be reflecting the size distribution of one particular subclass of particles. Murphy et al. (2014) identify three main particle types in the stratosphere (sulphuric, meteoric-sulphuric and organic-rich), and one could potentially consider an alternative classification based on origin (tropical-homogeneously-nucleated, polar-homogeneously-nucleated and meteoric-smoke-heterogeneously-nucleated), which would surely have different size modes reflecting their distinct sources and different experience of interacting with other constituents or processes during their lifetime. Indeed

Wilson et al., (2008) present the many years of in-situ stratospheric particle size distribution measurements by the FCAS instrument, which measure down to _30nm radius, and explain (Wilson et al., 2008) that ..."number size distributions extending below 100 nm may require more modes for accurate characterization". Although I appreciate this classification has not yet been established, I would recommend the authors avoid using the term "unimodal" since it seems quite possible the sub-200nm may have multi-modal size distribution, analagous to that observed in the troposphere (e.g. Whitby et al., 1978).

It would be fine to provide clarification that there is only one mode in the particular size range observed by the LOAC OPC, but the authors need to make that clear in the revised version of the manuscript.

Authors : "We agree with the referee 3 that balloon-borne OPCs cannot provide easily particle concentrations for sizes (diameters) below ~0.2 or even ~0.3 μm. The OPC from University of Wyoming is able to provide values at 0.02 μm (Deshler et al., 2003)[7] though size bins are definitely lacking between 0.02 and ~0.3 μm and possible other distribution modes cannot be highlighted in this range. However with such as size sampling distribution shapes defined as mainly "unimodal" were inferred for sizes smaller than ~0.5 μm for reported volcanic eruptions observed in the past (e.g. Russell et al., 1996[8]; Deshler et al., 2003; Kravitz et al., 2011[9]). The intense second mode inherent to the Pinatubo aerosol was apparent for larger sizes and was no present in the Sarychev moderate eruption for instance. As a result, the term "unimodal" can be considered as an approximation (or in the worst case, as a sort of "language abuse" commonly used in the literature dealing with balloon-borne observations by OPCs).

Integrated parameters (such as Reff, surface Area density, extinction) derived from log-normal unimodal shapes fitted on in situ data have shown good agreement with satellite data in volcanic

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[7] Deshler, et al. (2003). Thirty years of in situ stratospheric aerosol size distribution measurements from Laramie, Wyoming (41 N), using balloon-borne instruments. *Journal of Geophysical Research: Atmospheres*, *108*(D5).

[8] Russell et al. (1996). Global to microscale evolution of the Pinatubo volcanic aerosol derived from diverse measurements and analyses. *Journal of Geophysical Research: Atmospheres*, *101*(D13), 18745-18763.

[9] Kravitz, et al (2011) : Simulation and observations of stratospheric aerosols from the 2009 Sarychev volcanic eruption, J. Geophys.Res., 116, D18211, doi:10.1029/2010JD015501,

conditions (e.g. SPARC, 2006[10]). Perhaps the reason why this approximation has survived throughout stratospheric aerosol literature.

A sort of "language abuse" in such but also modelling work (see SPARC 2006 stratospheric aerosol Assessment). Data from the LOAC OPC used in our manuscript provide partial size distributions for sizes greater than 0.2 μm. This means that based on previously published work we can expect.

Anyway, following the reviewer's comment we have toned down our interpretation of the size distribution of the Calbuco aerosol and rewritten the corresponding subsection 3.2.2 in the revised manuscript:

- **Minor points:**

Referee 3 : Title, page 1, line 1: As explained above please remove the word "isentropic" from the title.

Authors : It was corrected in the revised manuscript

Referee 3 : Abstract, page 1, line 25: Please replace "1" with "one".

Authors : It was corrected in the revised manuscript

Referee 3 : Abstract, page 1, line 28: Please replace "SAOD" with "sAOD" because the AOD is already an established acronym for "aerosol optical depth" and it's easy for the reader to recognise the metric with the S in lower case. For this reason also change the word "Stratospheric" to have lower-case "s" on line 29. Please change also other instances of "SAOD" to "sAOD".

Authors : We thank the referee 3 for this suggestion. The correction was added to the revised manuscript.

Referee 3 : Abstract, page 2, line 1: Is this 90-day e-folding timescale for aerosol mass? Please clarify. Can any statement be made about whether this e-folding scale is faster initially than later?

Authors : The referee 3 is right. This point was clarified in the revised manuscript.

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[10] SPARC 2006 Assessment of stratospheric aerosol properties (ASAP). Technical report WCRP-124/WMO/TD-No. 1295/SPARC report no. 4, SPARC, Toronto, Ontario, CA, pp. 322.

Referee 3 : Abstract, page 2, line 5: Further to my comments above, please avoid the word "unimodal" as the LOAC OPC really is only characterising the "accumulation mode shoulder" of the particle size distribution, there could be other modes before. Suggest to reword replacing "an unimodal lognormal size distribution" with "the accumulation mode shoulder of the particle size distribution (above 250nm dry-diameter) log-normal in shape." Can you give a number for the geometric standard deviation here?

Authors : We now provide the lognormal parameters (No, Median radius and geometric standard deviation). Moreover, we understand the point of view of the referee 3. As consequence, this part of the abstract was re-written in clarifying the size range of this lognormal distribution (See revised manuscript).

Referee 3 : Abstract, page 2, lines 6-7: State briefly which measurements you mean here re: that the "background" conditions have been reached by this time. Which measurement established this, and is this compared to conditions before the eruption?

Authors : In order to reduce confusion this sentence was rephrased in the revised manuscript.

Referee 3 : Abstract, page 2, lines 11-12: It is explained that "the inhomogeneous geographical distribution of the plume is controlled by the latitudinal motion of these dynamical barriers". I see what you mean about the effects from this controlling behaviour of the dynamics, but suggest to use the word "spatio-temporal" rather than "geographical". Also, I don't quite follow what is meant by "latitudinal motion of these dynamical barriers" – please can you explain this and re-word that part of the sentence accordingly.

Authors : The correction was added in the revised manuscript. The dynamical barriers are depending to the PV gradient and the equivalent length. In particular, the position of the dynamical barrier is characterized by a local maximum of the PV gradient and a local minimum of the equivalent length (Nakamura, 1996[11]; Portafaix et al., 2003[12]). As a consequence, the

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[11] Nakamura (1996)., Two-dimensional mixing, edge formation, and permeability diagnosed in an area coordinate. *Journal of the atmospheric sciences*, *53*(11), 1524-1537

[12] Portafaix et al (2003) : A.. Fine-scale study of a thick stratospheric ozone lamina at the edge of the southern subtropical barrier. *Journal of Geophysical Research: Atmospheres*, *108*(D6)

dynamical barriers are not located at the same place during the time. As illustrated through our study, the dynamical barriers is located around at 15°S latitude on 27 April and around at 25°S latitude on 01 May. This evolution on the position of the dynamical barriers was formulated through the sentence pointing by the referee 3. In order to reduce the confusion, we rewritten this sentence in the revised manuscript.

Referee 3 : Introduction, page 2, line 16: Please re-word "meanly due to their role in ozone budget" to something like "principally due to their role in the ozone budget".
Authors : The sentence was corrected in the revised manuscript

Referee 3 : Introduction, page 3, lines 7-8: Suggest to reword "eruption which injected up to 20 Tg of SO2" to "injecting between 14 and 23 Tg of SO2 (Guo et al., 2004)" and replace "perturbed" with "perturbing".
Authors : We thank the referee 3 for this suggestion which was included in the revised manuscript.

Referee 3 : Introduction, page 3, line 14: The authors present a range for the tropical stratospheric warming as "(3,5 K) near the aerosol peak". Please re-word to put the range in words and explain whether what baseline this anomaly is comparing to (or cite the reference for the values given for the range)"
Authors : This sentence was re-written in the revised manuscript.

Referee 3 : Introduction, page 3, line 17: The authors clarify their use of the term "moderate eruption" as those which are "10-20 times weaker than Pinatubo eruption". Is this in terms of the amount of SO2 emitted? Is there a reference that has established that "magnitude" relative to Pinatubo to classify moderate eruptions?
Authors : The referee 3 is right to point about the term of « moderate eruption ». The volcanic eruption is classified following the Volcanic Explose Index (VEI) (Kravitz et al., 2010[13] ;

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[13] Kravitz et al (2010): Negligible climatic effects from the 2008 Okmok and Kasatochi volcanic eruptions,J. Geophys. Res., 115, D00L05, oi:10.1029/2009JD013525.

Vernier et al., 2011[14]). A major eruption like the Pinatubo eruption is affected to a VEI larger than 5 (Vernier et al., 2011). The moderate volcanic eruption are associated to VEI less or equal to 4 (Kravitz et al., 2010). In comparison to three moderate volcanic eruptions are ranked in the top 10 of the most influential events on the stratospheric aerosol, it could be possible to infer that the moderate eruption could be characterized by amount of SO_2 injected 10-20 times weaker than Pinatubo eruption. In order to clarify the term of the moderate eruptions we applied the VEI to define them in the revised manuscript.

Referee 3 : Introduction, page 3, line 25-32: On line 32 it is clarified that 10-20 times less than Pinatubo is referring to mass of sulphur emitted, but as the authors have stated, Kasatochi emitted between 1.5 and 2.5 Tg of SO2 which is less than 10 times Pinatubo's 14-23 Tg, so by that classification it would be considered larger than "moderate". Please revise the "10-20 times larger" classification for moderate – can a different classification be given?

Authors : As we mentioned previously, the term of the moderate eruptions was defined in the revised manuscript through the use of the VEI.

Referee 3 : Introduction, page 3 lines 33 and page 4 lines 1-2: The previous sentence discussed "minor" or "moderate" eruptions (I prefer the term "moderate", and best to be consistent with this terminology) but this sentence is then referring to major eruptions – please re-word to clarify this distinction.

Authors : The referee 3 is right. As a consequence, it was corrected in the revised manuscript.

Referee 3 : Introduction page 4, after line 15. Further to the comment about the aerosol plume not necessarily being transported isentropically, suggest also to add one or two sentences something like this, "The fact that sulphuric particles grow larger following major eruptions (e.g. Russell et al., 1996; Bauman et al., 2003) means they can sediment appreciably during transport within the stratosphere, causing the plume transport to diverge from the expected isentropic trajectory. Even in moderate eruptions, where

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[14] Vernier et al (2011): Major influence of tropical volcanic eruptions on the stratospheric aerosol layer during the last decade, Geophys. Res. Lett., 38, L12807,doi:10.1029/2011GL047563, 2011.

sulphuric particle growth may not be significant, the accomodation of sulphur onto ultra-fine ash particles has the potential to also change the fate of a proportion of the volcanic plume."

Authors : We understand the point of view of the referee 3. The life cycle of an aerosol is affected by microphysical processes such as sedimentation which does not favor to isentropic transport. Thus, we added the sentence suggested by the referee 3 in the revised manuscript.

Referee 3 : Introduction, page 5, lines 8 and 9: insert commas between "laser" and "which", "wavelength" and "with" and delete "a" between "emits" and "radiation".

Authors : It was corrected in the revised manuscript

16) page 5, line 19, replace "lidar has been described first by" with "lidar, first described by"

Authors : It was corrected in the revised manuscript

17) page 5, line 21, replace "the" between "and" and "pressure"

Authors : It was corrected in the revised manuscript

Referee 3 : page 5, lines 25-25, Replace "also call lidar coefficient. It depends" with "also called lidar coefficient, which depends...". The authors cite an extinction-to-backscatter ratio of 60 for background stratospheric aerosol, but do not provide a reference for that value.

There should also be added mention of how this ratio varies as the particle size distribution is perturbed (e.g. see Vaughan et al. ,2004). Please also mention here approaches to utilize more complex algorithms to derive extinction from lidars which also measure depolarization, for example as developed by Young and Vaughan, (2009) to derive extinction from CALIOP space-borne lidar.

Authors : The sentence mentioned by the referee 3 was corrected in the revised manuscript. Furthermore, we included some references concerning the choice of the value of the LiAR coefficient at 60. Values range from 50 to 60 are commonly assumed for volcanically quiescent

conditions and periods of moderate eruptions (Trickl et al., 2013[15] ; Ridley et al., 2014[16] ; Sakai et al., 2016[17] ; Khaykin et al., 2017[18]). The referee 3 is right to mention that the LiDAR coefficient is sensible to particle size distribution. Thus, the value of this LiDAR coefficient in background aerosol condition is different to value in volcanically perturbed conditions. The error in the LiDAR coefficient has a larger impact on aerosol extinction and optical depth (Khaykin et al., 2017). This discussion was included in the revised manuscript.

Referee 3 : page 6, line 12: The authors have given uncertainty estimates for each size bin, but then in Figure 8 have plotted the observed size distribution within error bars. Please add those to indicate the overall uncertainty, as explained there. Also, please replace "part per cm3" with "cm-3" (with superscript "-3"), move the "particles" to before "concentrations" as "particle concentrations" and move "respectively" to the end of the sentence.

Authors : The modification asked by the referee 3 was added in the revised manuscript

Referee 3 : page 6, line 22: add "," after "nm" and before "available". Also the citation Winker et al. (2010) is given but only the 2009 paper is given in the references – I assume the 2009 reference was intended. Please correct.

Authors : The referee 3 is correct. It was corrected in the revised manuscript.

Referee 3 : page 6, line 32: replace "looking" with "view".

Authors : It was corrected in the revised manuscript

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[15] Trickl et al (2013) : 35yr of stratospheric aerosol measurements at Garmisch-Partenkirchen: from Fuego to Eyjafjallajökull, and beyond, Atmos. Chem. Phys., 13, 5205–5225, doi:10.5194/acp-13-5205-2013

[16] Ridley et al (2014).:Total volcanic stratospheric aerosol optical depths and implications for global climate change, Geophys. Res. Lett., 41, 7763–7769, doi:10.1002/2014GL061541

[17] Sakai et al (2016).: Long-term variation of stratospheric aerosols observed with lidars over Tsukuba, Japan, from 1982 and Lauder,New Zealand, from 1992 to 2015, J.Geophys.Res.Atmos., 121,10283–10293, doi:10.1002/2016JD025132, 2016.

[18] Khaykin et al (2017). Variability and evolution of the midlatitude stratospheric aerosol budget from 22 years of ground-based lidar and satellite observations. *Atmospheric Chemistry and Physics*, *17*(3), 1829-1845.

Referee 3 : page 8, lines 21-25: The two sentences beginning "Figure 1" and "The ATB" are describing the aerosol signal observed by CALIOP and therefore do not belong in this "3.1.1 SO2 plume" section – suggest to move to the start of section "3.1.2 Spatial extent of the aerosol plume". Also that title for the 3.1.2 should potentially have "and temporal evolution" after "Spatial extent".

Authors : The referee 3 is right and this modification was added in the revised manuscript.

Referee 3 : page 9, lines 6-8: please give the actual values here (with uncertainty range if possible) for the SO2 emitted in the two eruptions being explained.

Authors : These elements were added in the revised manuscript.

Referee 3 : page 9, lines 10-12: this sentence is not quite worded correctly, but sounds like it is saying the ratio between SO2 emitted and maximum sulphate aerosol loading is different for Calbuco than for other similar eruptions. Please can you re-word to explain what is meant here.

Authors : This sentence was rewritten in the revised manuscript. Moreover, the manuscript was corrected by a native English speaker in order to check the quality of the English of the revised manuscript.

Referee 3 : page 9, line 23: The authors specify condensation of H2SO4 into the liquid binary aerosol, but some small proportion of the H2SO4 is also converted into aerosol via new particle formation, suggest to replace "condensed" with "converted".

Authors : It was corrected in the revised manuscript

Referee 3 : page 9, line 24: Insert commas after "15-17km" and "Atlantic Ocean".

Authors : It was inserted in the revised manuscript

Referee 3 : page 9, line 28: Presumably this refers to "2-week composite" type product here from CALIOP – has this already been explained? With the move of the two sentences in comment 22) could also add a sentence explaining these 2-week near-global crosssection composites?

Authors : We added a sentence in the revised manuscript in order to explain the calculation realized for the Figure 4 and 5.

Referee 3 : page 10, lines 1-3 – mention that this is the period when the SO2 is still being converted (refer to Figure 2). Then when you say "This could be attributed" state exactly what you mean: "This elevated backscatter in the tropics...". Have there been other studies that tracked the Kelud eruption can be cited here re: the longevity of the Kelud plume?

Authors : These modifications were added in the revised manuscript. Moreover, we cited the work of Kristiansen et al (2015)[19] on the stratospheric volcanic ash emissions during the Kelut eruption.

Referee 3 : page 10, line 7 – state how the background levels are established here. And that this 2nd two-weeks will be after the SO2 has been oxidised to aerosol.

Authors : We explained in the revised manuscript how the background level was defined. Moreover, as suggested by the referee 3, we mentioned that the 16-31 May period correspond to the period where the SO_2 has been oxidized to aerosol.

Referee 3 : page 10, line 10 – "norther than" –> "north of".
Authors : It was corrected in the revised manuscript.

Referee 3 : page 10, lines 14-15 – suggest to expand this sentence also mentioning the deepening of the layer. Also it looks like the equatorial backscatter is also enhanced in the UT – suggest to mentioned this here too.

Authors : The two points mentioned by the referee 3 were added in the revised manuscript.

Referee 3 : page 10, lines 31-32 – Need to explain how the Angstrom exponents are used here – has the wavelength been converted to 532nm from some other frequency so that it can be compared equivalently? For the lidar there is the issue of the conversion to extinction from backscatter – what is assumed here in deriving the lidar extinction (see comment 18)?

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[19] Kristiansen et al (2015) : Stratospheric volcanic ash emissions from the 13 February 2014 Kelut eruption, *Geophys. Res. Lett.*, *42*,588–596, doi:10.1002/2014GL062307

Authors : The methodology used to convert the wavelength to 532 nm is explain in details by Khaykin et al (2017). The wavelength conversion of extinction coefficient α can be performed according to the following equation :

$$\alpha^{\lambda_2} = \alpha^{\lambda_1} \left(\frac{\lambda_2}{\lambda_1}\right)^{K_e} \quad (Eq.1)$$

Where K_e is the Angström exponents. The Angström exponents for the 355-532 nm pair were adapted from Jäger and Deshler (2002)[20] and set to -1.3. The Angström exponents used for OMPS (from 675 to 532 nm) is based on the work of Khaykin et al (2017) and set to -1.8. The LiDAR extinction is derived from the Klett inversion (Klett, 1981) which is the common method used by the community. One source of uncertainty of this approach is based on the value of the LiDAR coefficient. As reported above, we adopted a value of 60 which is commonly assumed for volcanically quiescent conditions and periods of moderate eruptions. The discussion on the Angström exponents was added to the revised manuscript.

Referee 3 : page 11, lines 6-15 – There needs to be some discussion here on the differences in vertical resolution between the ground-based lidar and the satellite profiler different sensors. What is the vertical resolution of the OMPS profiler and its horizontal footprint?

Authors : This part of discussion was improved in the revised manuscript. We discussed on the difference in vertical resolution between LiDAR and OMPS which could be one possible cause at the origin of discrepancies. Indeed, the vertical resolution of the ground-based LiDAR and OMPS are 0.15 km and 1.5 km respectively (Jaross et al., 2014)[21]. Moreover, the discrepancies existing between results presented in this study could be also due to different measurement techniques or horizontal resolution observation. Unlike satellite experiments that allow global observations, a ground-based LiDAR system is able to derive aerosols characteristics at a specific location. OMPS views the Earth's limb looking backward along the orbit track of approximately 125 km with a horizontal resolution of 50 km. These points were discussed in the revised manuscript.

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[20] Jäger, H., and T. Deshler (2002) : Lidar backscatter to extinction, mass and area conversions for stratospheric aerosols based on midlatitude balloonborne size distribution measurements,Geophys. Res. Lett., 29(19), 1929, doi:10.1029/2002GL015609

[21] Jaross et al (2014) : OMPS Limb Profiler instrument performance assessment, J. Geophys. Res. Atmos., 119, 4399–4412, doi:10.1002/2013JD020482.

Referee 3 : page 11, line 22 – replace "quick" with "brief" – state the altitude range over which this minimum is seen.

Authors : It was corrected in the revised manuscript

Referee 3 : page 11, line 27: Compare here to the stronger June period when the extinction is maximum at 19.5 km, what is the reason for the descending signal?

Authors : One sentence was added in the revised manuscript for the comparison to the stronger values observed in June. Given that this descending signal is associated to a decrease of height of the plume, we assume that the main reason could be due to the sedimentation process. We note that the influence of the removal processes on the evolution of the plume were already discussed in the section 4.2. Nevertheless, we added a sentence in the section 3.2.1 to introduce the discussion on the possible impact of the removal processes on the evolution of the plume.

Referee 3 : page 11, lines 30-31 – is it possible to add a third plot that degrades the vertical resolution of the lidar to match the vertical resolution or averaging kernel of the satellite instrument?

Authors : We thank the referee 3 for this interesting suggestion. Thus, we degraded the vertical resolution of the LiDAR to match the vertical resolution of OMPS (See Figure below). The comparison in term of altitude and shape of the plume stay unchanged.
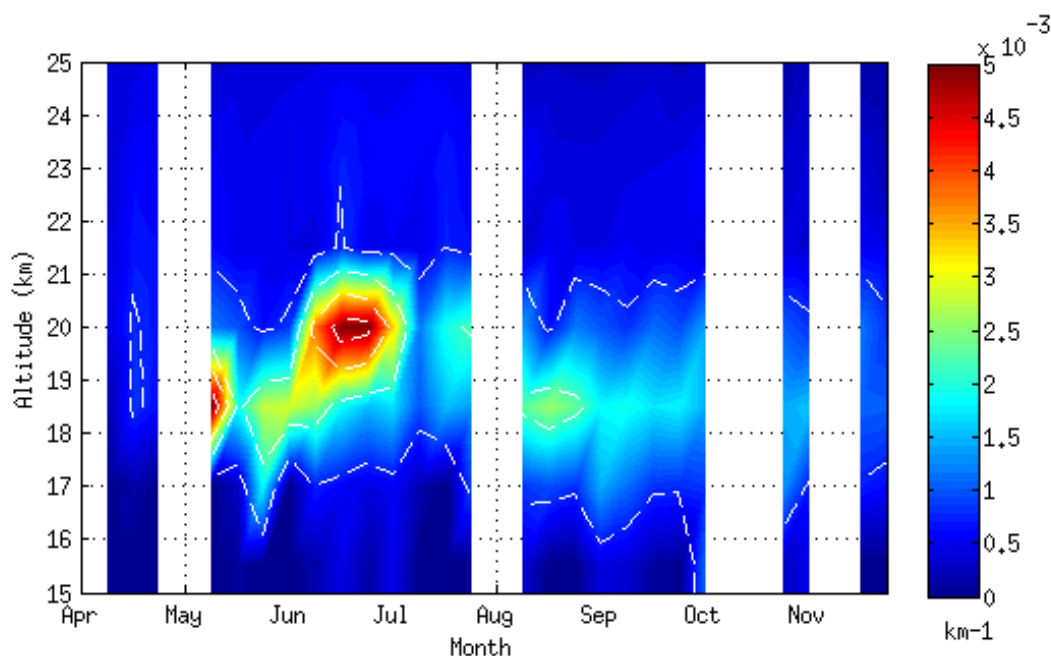
Figure 3.1 *: Time series of weekly-averaged profiles of extinction at 532 nm obtained from LiDAR with a vertical resolution of 1.5 km.*

As we discussed previously, the differences between the satellites and ground-based observations could be also due to the inhomogeneity of the plume. We think that this figure provides nothing new to the discussion on the difference between the satellites and ground-based observations. It is for this reason we decided not to include this figure in the revised manuscript.

Referee 3 : page 11, lines 32-33: This sentence is because of the vertical resolution of the OMPS – move this to the discussion of the resolution differences.

Authors : This sentence was moved to the discussion of the resolution difference as suggested by the referee 3.

Referee 3 : page 11, line 33: A key difference looks like that the OMPS profiler sees much higher aerosol extinction in the 15-17km region? Suggest to add mention of this – is there a potential reason for this?

Authors : The referee 3 is right. This information was added in the revised manuscript

Referee 3 : page 12, line 4: The text says 50 degrees longitude – is this a typo?. On this I suggest moving Figure 1 to here or adding the sampling region for the comparison with the observations onto that Figure.

Authors : We confirm the referee 3 that it is not a typo. We understand the point of view of the referee 3. In order to avoid to encumber the figure and to focuses mainly on the comparison on the CALIOP and LiDAR profiles, we added a description of the domain in the caption and the text.

Referee 3 : page 12, line 14: I am surprised the authors have not mentioned the clear signal of the descent in the altitude of peak extinction from _20km in May to _18.5km in August – this can be seen in both datasets and there needs to be some discussion of this here.

Authors : The point highlighted by the referee 3 was added in revised manuscript.

Referee 3 : page 12, line 16: Please state the 4 dates here of the LOAC OPC soundings.

Authors : As suggested by the referee 3, the 4 dates were stated in the revised manuscript.

Referee 3 : page 12, line 17: Suggest to move the "532nm" to be before "were calculated" and add "from the fits to the observed size distributions at each level". One might expect the in-situ measured size distribution is to be considered the reference against which to

compare the satellite and lidar values? Is that reasonable to consider that or are the plume inhomogeneity and sampling differences too big to make that simplistic assessment.

Authors : The suggestion of the referee 3 was included in the revised manuscript. The referee 3 is right to mention that the comparison between these different devices is not a simplistic assessment. We could be expected that the in-situ measured size distribution should be consider as the reference. Given that the poor sampling (4 sounding for the study period) and the plume inhomogeneity, it is not reasonable to consider that. It is for this reason that the LOAC measurement are not presented as the reference in the manuscript. The comparison is pointing on the LiDAR and satellite observations.

Referee 3 : page 12, line 22: Is "discrepancies" the right word here – as above please be clear whether to compare against the reference in-situ AOD? Or are the inhomogeneity and differences in sampling mean it is not so simple. Please clarify in the text. Also delete " terms of" and "mainly observed in May".

Authors : We understand the point of view of the referee 3 and we mentioned previously the comparison is pointing mainly between LiDAR and satellite observations. As a consequence, this part of the sentence was removed in the revised manuscript.

Referee 3 : page 12, line 23: Delete "using" and put "or may also be due to" (or reword re: comment 43 above).

Authors : As mentioned previously, this part of the sentence was removed in the revised manuscript.

Referee 3 : page 12, line 25 – Please explain here – what is the limit for the vertical resolution that the integration time and ascent rate limits it to?
Authors : This sentence was rewritten in the revised manuscript.

Referee 3 : page 12, lines 25-27 – This sentence is unclear – please re-write.

Authors : This sentence was rewritten in the revised manuscript.

Referee 3 : page 12, lines 29 – Replace "DV" with "dV".
Authors : It was corrected in the revised manuscript

Referee 3 : page 12, lines 30-31 – Further to my comment about unimodal size distribution at the start, please reword the categorisation here. The LOAC OPC only measures the "shoulder" of the size distribution so it does not constrain whether there is more than one mode for particles below 250 nm diameter – need to state this.

Authors : The lower bound of the LOAC size range is of 0.2 μm which does not provide a full description of the size distribution and of possible secondary modes for smaller particles (Wilson et al., 2008). The comment of the referee 3 was taking into account in the revised manuscript.

Referee 3 : page 13, line 1 – please insert a clarifying phrase that you mean bimodal in particles above 250nm diameter.

Authors : A sentence was added in the revised manuscript.

Referee 3 : page 13, lines 12-13 – this is interesting – are you saying you mean that there might be some compensation between the additional coarse (ash?) particles and additional ultra-fine particles e.g. from nucleation? Please re-word to clarify what you mean here.

Authors : This part of the manuscript was rewritten as suggested by the referee 3.

Referee 3 : page 13, line 14 – suggest to insert "the full" before "19 size classes" so it's clear this is a total number of particles.

Authors : It was corrected in the revised manuscript.

Referee 3 : page 14, lines 8 and 9 – As per my first general comment at the start, please remove the word isentropic here as this may not be the case due to sedimentation. I realise that the model is providing isentropic trajectories but then suggest to move the word "isentropic" in line 9 to be instead before "MIMOSA model". By inserting "of the plume" before "the high resolution" that then reads fine I think – please also provide brief descriptor for the model such as "isentropic Lagrangian trajectory model" or similar.

Authors : The modification suggested by the referee 3 was included in the revised manuscript. Moreover, we added a brief description of MIMOSA like as an isentropic Lagrangian trajectory in section 2.2.

Referee 3 : page 14 lines 20-21 – "cannot move beyond the south of Brazil" – suggest to reword this – is it just that the trajectory for the airmasses takes the plume this way – I see what you mean but I think better to phrase it differently. Also it is only 5 days since the eruption at this time (in panel a) – or was this meant as panel b?

Authors : The sentence pointing by the referee 3 was rewritten in the revised manuscript.

Referee 3 : page 15 lines 14 – perhaps to rephrase as "discussed" rather than "revealed" as this was clearly established already (e.g. Deshler, 2008) and replace "they showed" with "they suggested". Also replace "overestimation of the strength of a STE event" with "a general overestimation of stratosphere-troposphere exchange with global composition climate models"

Authors : It was corrected in the revised manuscript

Referee 3 : page 15 line 17 – replace "stratosphere into the middle and high latitude" with "stratosphere into the troposphere".

Authors : It was corrected in the revised manuscript

Referee 3 : page 15 line 15 into page 16 lines 1-2. Suggest to re-write this as something like "We note the potential role of sedimentation on the initial dispersion of volcanic aerosols, in particular the effects from with co-emitted ultrafine ash particles, but do not explore this effect here."

Authors : As suggested by the referee 3 this part of the manuscript was rewritten.

Referee 3 : page 16 lines 13-15 – state the actual values for the SO2 emitted and replace "amounts" with "mass" – the sentence can also be shortened by moving "northern hemisphere" before "Sarychev" and deleting "in the". Suggest also to delete "we report the same" replacing with "with similar SO2" and delete "i.e.".

Authors : These modifications were included in the revised manuscript

Referee 3 : page 16 lines 25 – insert comma before "possibly".

Authors : It was included in the revised manuscript

Referee 3 : page 16 line 31 – reword re: unimodal or at the least need to add clarifying "above 200nm diameter"

Authors : It was clarified in the revised manuscript

Figures :

 Also – it would help to indicate the period where the SO2 is being depleted until about the 7th May when it seems to barely be depleted at all. This needs to be mentioned in the next – can the change in total mass be explained in some way with some hypothesis? Is chemistry/oxidant-limitation involved?

Authors : The modification suggested by the referee 3 was included in the revised manuscript. The discussion on the evolution of the SO_2 and the involvement of the chemistry/oxidation limitation was already presented in the manuscript in the Section 3.1. By combining IASI to CALIOP observation, we think that the second half of May (11-31 May) corresponds to the period where the SO_2 has been oxidized to aerosol. The SO_2 plume began to diminish on 11 May 2015 by the oxidation of SO_2 to gaseous sulphuric acid which further converted into H_2SO_4-H_2O liquid aerosol.

Referee 3 : page 30 caption to Figure 3 – replace "Height injection(in km)" with "Injection height (km)"

Authors : The referee 3 is right. It was corrected in the revised manuscript.

Referee 3 : page 31 Figure 4 – in the caption add text in brackets after each date-range something like "(1-3 weeks after eruption)" or similar.

Authors : It was corrected as suggested by the referee 3

Referee 3 : page 34 Figure 7 – in the caption replace "from (a) lidar and (b) CALIOP" with something like "from ground-based (a) and space-borne (CALIOP, b) lidar" before "observations". Insert "Island" after "Reunion".

Authors : It was corrected as suggested by the referee 3

Referee 3 : page 35 Figure 8 – add error bars for each size channel (with the relative uncertainty values given in the text).

Authors : This modification was added in the revised manuscript.