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 ± 5° latitude and ± 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 SO2 is then"? What part (or layer) of the atmosphere is SO2 injected into? For example, it could be: "The injected into the stratosphere SO2 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 analternative 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

[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.

[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

[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] ;

[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 dis cussed "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.

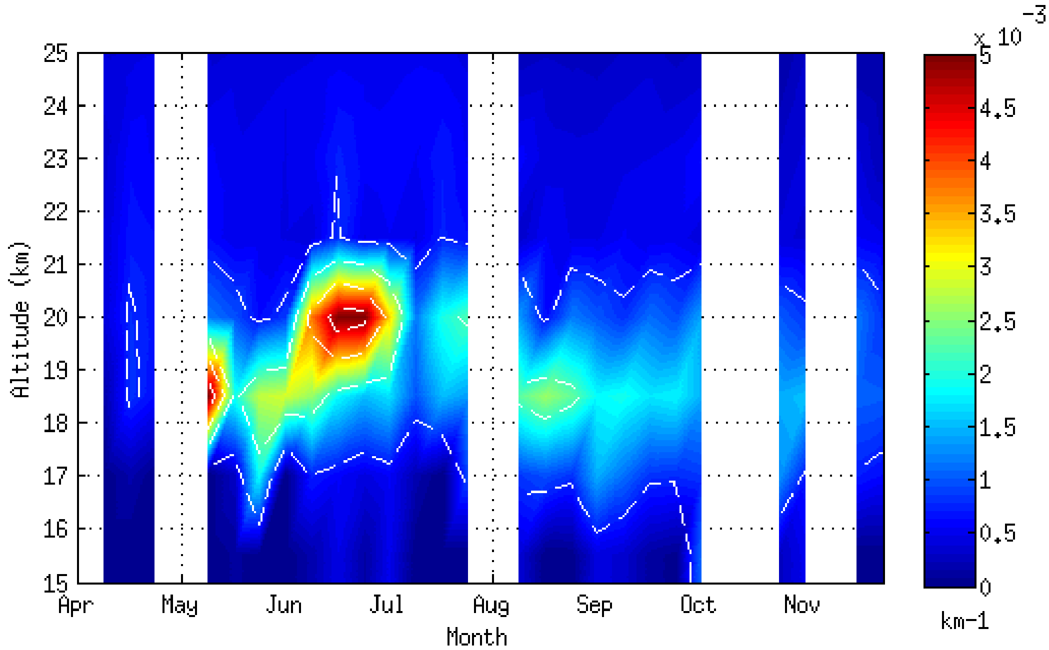

***Figure 3.1** : Time series of weekly-averaged profiles of extinction at 532 nm obtained from LiDAR with a vertical resolution of 1.5 km.*

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 :

Referee 3 : page 29 caption to Figure 2 – insert "column" between "total" and "mass". 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.

Long-range transport of stratospheric aerosols in the Southern Hemisphere following the 2015 Calbuco eruption

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Abstract

After 43 years of inactivity, the Calbuco volcano which is located in the southern part of Chile erupted on 22 April 2015. The space-time evolutions (distribution and transport) of its aerosol plume are investigated by combining satellite (CALIOP, IASI, OMPS), in situ aerosol counting (LOAC OPC) and LiDAR observations, and the MIMOSA advection model. The Calbuco aerosol plume reached the Indian Ocean one week after the eruption. Over the Reunion Island site (21°S; 55.5°E), the aerosol signal was unambiguously enhanced in comparison with "background" conditions with a volcanic aerosol layer extending from 18 km to 21 km during the May-July period. All the data reveal an increase by a factor of ~2 in the sAOD (stratospheric Aerosol Optical Depth) with respect to values observed before the eruption. The aerosol mass e-folding time is approximately 90 days which is rather close to the value (~80 days) reported

for the Sarychev eruption. Microphysical measurements obtained before, during and after the eruption reflecting the impact of the Calbuco eruption on the lower stratospheric aerosol content have been analyzed over the Reunion Island site. During the passage of the plume, the volcanic aerosol was characterized by an effective radius of 0.16 ±0.02 µm with a unimodal size distribution for particles above 0.2 µm diameter. Particle concentrations for sizes larger than 1 µm are too low to be properly detected by the LOAC OPC. The aerosol number concentration was ~20 times that observed before and one year after the eruption. According to OMPS and LiDAR observations, a tendency toward conditions before the eruption has been observed by April 2016. The volcanic aerosol plume is advected eastward in the southern hemisphere and its latitudinal extent is clearly bounded by the subtropical barrier and the polar vortex. The transient behavior of the aerosol layers observed above Reunion Island between May and July 2015 reflects an inhomogeneous spatio-temporal distribution of the plume which is controlled by the localization of these dynamical barriers.

1. Introduction

Stratospheric aerosol affect the chemical and radiation balance of the atmosphere (McCormick et al., 1995; Solomon, 1999; SPARC 2006). The importance of stratospheric aerosol on the chemistry is mostly due to its role in the ozone budget (Solomon et al., 1986; Bekki, 1997; Borrmann et al., 1997). Stratospheric aerosol provide sites for heterogeneous chemical reactions leading to stratospheric ozone depletion, significantly enhanced in periods of high aerosol loadings following major volcanic eruptions (Solomon, 1999 and references therein). In addition, periods of enhanced stratospheric aerosol loadings can lead to significant warming in the stratosphere and cooling in the troposphere (e.g. McCormick et al., 1995; Solomon et al., 2011; Arfeuille et al., 2013). As reported by Kremser et al. (2016), a better understanding of the processes governing the lifetime of stratospheric aerosol is needed to assess the impacts on climate and chemistry. Since the discovery of the permanent stratospheric aerosol layer, called Junge Layer, in 1961 (Junge, 1961), it has been established that stratospheric aerosol are mostly composed of sulfuric acid droplets with some more complex characteristics in the stratosphere where organic compounds and meteoritic dust can also contribute to its composition (Neely et al., 2011, Froyd et al., 2009). The main sources of stratospheric sulfur are Carbonyl Sulfide (OCS), Dimethyl Sulfide (DMS) and sulfur dioxide (SO_2) (SPARC, 2006), the latter being significantly enhanced after volcanic eruptions (Carn et al., 2015). The injected SO_2 into stratosphere is oxidized into H_2SO_4, which (after homogeneous nucleation and/or condensation onto existing aerosol particles) causes an increase in the content of liquid sulfate aerosol

(SPARC, 2006). Based on the control of the stratospheric aerosol burden over the last 25 years, Thomason et al. (2007) showed that volcanic effects dominate over natural and anthropogenic sources. Previous studies on stratospheric aerosol have significantly characterized its properties and variability during "background" (i.e. free of volcanic aerosol) and volcanic conditions (e.g., Stenchikov et al., 1998; Jäger and Deshler, 2002; Bauman et al., 2003; Hermann et al; 2003; Hofmann et al; 2009).

The eruption of the Pinatubo in 1991 is known to be the last major volcanic eruption injecting between 14 and 23 Tg of SO_2 significantly perturbed the global stratosphere for several years (Kinninson et al., 1994; McCormick et al., 1995; Stenchikov et al., 1998, 2002; Guo et al., 2004; Dhomse et al., 2014). As reported by Russell et al. (1996), in addition to the prodigious increase in the stratospheric aerosol loading, this event significantly affected numerous aspects of the atmospheric system including: i) a 2-year cooling of the global surface temperature of several tenths of degrees (Canty et al., 2013; Wunderlich and Mitchell, 2017); ii) a warming of the tropical stratosphere ranging from 1° to 4°C (Labitzke and McCormick et al., 1992; Young et al., 1994); (iii) a lifting of the tropical ozone layer by ~1.8 km (Pueschel et al., 1992; Grant et al., 1994). By the use of satellite and balloon-borne observations, various studies have shown that moderate volcanic eruptions (i.e., volcanic explosive index less or equal to 4) can significantly modulate stratospheric aerosol concentrations (Bourassa et al., 2010; Kravitz et al., 2010; Solomon et al., 2011; Vernier et al., 2011; Clarisse et al., 2012; Jégou et al., 2013). Based upon satellite observations, Vernier et al (2011) showed that the decadal increase in stratospheric aerosol loadings since 2002 can be attributed to a series of moderate volcanic eruptions. As reported by Kremser et al (2016), this decadal trend was also obtained from LiDAR (Hofmann et al., 2009; Trickl et al., 2013; Zuev et al., 2017) and ground-based sun-photometer observations (Ridley et al., 2014). Three moderate volcanic eruptions are ranked in the top 10 of the most influential events on the stratospheric aerosol burden including during the 2002-2012 period : (1) The Kasatochi eruption (52.2° N; 175.5° W, Alaska) in 2008 which injected 1.5-2.5 Tg of SO_2 into the upper troposphere and lower stratosphere (UTLS) (Bourassa et al., 2010; Kravitz et al., 2010; Krotkov et al., 2010); (2) The Sarychev eruption in June 2009 (48.1°N; 153.2°E, the Kuril Island) which released 0.9 Tg of SO_2 into the UTLS (Clarisse et al., 2012; Kravitz et al., 2011; Jégou et al., 2013); (3) The Nabro eruption (13.4°N; 41.7°E, Eritrea) in June 2011 which emitted 1.3 Tg of SO_2 into the UTLS (Bourassa et al., 2012; Sawamura et al; 2012). In comparison, these recurrent moderate volcanic eruptions injected 10-20 times less SO_2 than the Pinatubo eruption (Solomon et al., 2011). These eruptions can also be used to understand stratospheric dynamics as was done for the case of the Pinatubo eruption (Trepte et al., 1992).

Indeed, following a volcanic eruption, stratospheric aerosol can be used as a dynamical tracer (Bencherif et al., 2003; Fairlie et al., 2014). Based on satellite observations and a Lagrangian trajectory model, Fairlie et al. (2014) used the dispersion of the Nabro plume to study the dynamics of the Asian Monsoon Anticyclone. Hitchman et al. (1994) and SPARC (2006) suggested that the stratospheric aerosol distributions could be used to understand changes in the Brewer-Dobson Circulation. More recently, Ray et al. (2014) combined in situ balloon observations of SF_6, CO_2 with a numerical model to show that major explosive volcanic eruptions can induce large-scale changes in the stratospheric circulation via radiative perturbations, improving our understanding of stratospheric transport variability. Aerosol heating in the lower stratosphere induces a westerly wind anomaly, and enhanced tropical upwelling (Ray et al., 2014; Pitari et al., 2016a). Using the University of L'Aquila climate-chemistry model (ULAQ-CCM), Pitari et al. (2016a) analyzed the volcanic aerosol perturbation from the eruption of Mt. Pinatubo on the transport of long-lived species, N_2O and CH_4. They showed that the observed decline of long-lived greenhouse gases one year after the eruption is quantitatively consistent with enhanced stratosphere-troposphere exchange, due to a change in the Brewer–Dobson circulation. They also revealed that the volcanic aerosol radiative perturbation to stratospheric dynamics may be found by looking at the stratospheric age-of-air. According to Pitari et al (2016a), enhanced tropical upwelling tends to decrease the tropical age of air and the latitudinal age gradient after major volcanic eruptions. Although most studies discuss major tropical eruptions and their induced dynamical effects, we cannot exclude the possibility that 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).

Previous works have also revealed that stratospheric aerosol can be used to study meridional air mass transport from the tropical stratospheric reservoir (Trepte and Hitchman, 1992; Randel et al., 1993; Chen et al., 1994; Grant et al., 1996, Vernier et al., 2009). Based on satellite observations, Trepte and Hitchman (1992) have shown that transport from the tropics to mid-latitudes is favored during westerly shear phases of the quasi-biennial oscillation (QBO) rather than during the easterly shear phases. More recently, by the use of satellite observations and climate models, Hommel et al. (2015) revealed that the vertical and latitudinal extent of the stratospheric aerosol layer (between 16 and 31 km) in the tropics is modulated by the QBO. Pitari et al. (2016b) analyzed the radiative perturbations in the stratosphere induced by the last five major volcanic eruptions after 1960 (i.e, Angung, St Helens, El Chicon, Nevado del Ruiz and Pinatubo) using a climate model which included an aerosol microphysics module for aerosol formation and growth. They found an increase in stratospheric temperature associated with a significant impact on the tropical upwelling. The impact on stratospheric upwelling is found to be larger when the volcanically perturbed stratospheric aerosol is confined to the tropics, as tends to be the case for eruptions which are followed for several months with easterly shear of the QBO. They showed that the Nevado del Ruiz and Pinatubo eruptions occurred during years with dominant QBO easterly shear, which led to the confinement of the aerosols near the equator, with less poleward transport. This tropical confinement produced a larger latitudinal gradient of the perturbation heating rate and a stronger impact on the tropical upwelling (Pitari et al., 2016b). It is worth noting that the life cycle of an aerosol is affected by sedimentation. 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 sulphuric particle growth may not be significant, the accommodation of sulphur on to ultra-fine ash particles has the potential to also change the fate of a proportion of the volcanic plume.

This paper reports on the Calbuco plume observations over Reunion Island (20.5°S; 55.5°E) and its transport in the southern tropics. The geometrical and optical properties of the Calbuco plume are inferred from the ground-based observations at Reunion Island in the framework of the MORGANE (Maïdo ObservatoRy Gas Aerosols NDACC Experiment) campaign. The aim of this study is to provide a description of the dynamical context which has favored the spread of the Calbuco plume in the southern hemisphere. The paper is organized as follows: Section 2 describes the observations and the model used for the investigation of the volcanic aerosol transport. A description of the long-range transport of the volcanic plume over the Indian Ocean is provided in Section 3; Section 4 gives a dynamical analysis of this case study; and the summary and the conclusions are given in Section 5.

2. Instrumentation and model description

2.1 Observations

2.1.1 Ground-based lidar

One part of the observations used in this study was performed during the MORGANE campaign which took place at the Maïdo observatory on Reunion Island in May 2015. The MORGANE ground-based observational systems combine LiDAR and balloon-borne payloads to study the composition and the dynamics of the UTLS in the southern hemisphere. 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). It is also possible to retrieve aerosol profiles in the 15-38 km altitude range from these measurements. This instrument has been in operation at the Maïdo observatory since early 2013. The technical details and evaluation of its performance are given by Baray et al., (2013). A brief description of this DIAL system follows. It uses a frequency-tripled Nd:YAG laser, which provides a beam at 355 nm wavelength, with a repetition rate of 30 Hz and a XeCl excimer laser which emits radiation at 308 nm at 40 Hz. The optical receiver is a telescope composed of 4 parabolic mirrors where the backscattered signal is collected by 4 optical fibers located at the focal points. The current configuration of the DIAL LiDAR system mainly detects signals in the UV regions of the spectrum (308, 332, 355 and 387 nm). The LiDAR data set used in this study consists of daily records of backscattering signal obtained from the Maido facility between 1 November 2014 and 30 November 2016 (106 profiles). It should be noted that no measurements were recorded at Reunion Island from January to April 2016 because of technical problems. The daily measurements are nighttime and time-integrated over about 3 hours in average.

We used the methodology described by Sasano (1985) to obtain the extinction and backscatter coefficient from a Rayleigh-Mie LiDAR. This methodology is similar to the approach of Klett (1981) with the advantage of providing a numerical calculation of the extinction and backscatter coefficient. Temperature and pressure profiles are needed to retrieve optical properties from this approach. For this study temperature and pressure profiles obtained from a radiosondes launched from the airport of Gillot at 11h (UTC) are used. In order to obtain a complete temperature and pressure profiles range from ground to mesosphere, we used the Arletty atmospheric model (Hauchecorne et al., 1998; Nair et al., 2012), based on European Centre for Medium Range Weather Forecast (ECMWF) data. The altitude of reference is determined for each profile. On average, the reference altitude is located between 30 and 40 km. Another parameters that we need to retrieve the optical properties is the ratio of backscatter and the extinction coefficient for aerosol, also called lidar ratio. For background stratospheric aerosol, the value found in the literature is near 60 (Trickl et al., 2013 ; Ridley et al., 2014; Sakai et al., 2016; Khaykin et al., 2017). This value is commonly used for volcanically quiescent conditions and periods of moderate eruptions (Sakai et al., 2016). The LiDAR ratio depends on the particle size distribution and the type of aerosol (Jäger and Deshler, 2002 ; Young and Vaughan, 2009). Error in the LiDAR ratio could influence significantly the uncertainty on aerosol extinction and optical depth (Sakai et al., 2016 ; Khaykin et al., 2017). Moreover, it should be noted that new approaches to derive extinction from LiDAR, which also measure depolarization, have been developed and already applied to space-borne lidar such as CALIOP (Young and Vaughan, 2009).

2.1.2 Balloon-borne OPC.

In order to analyze the evolution of the concentration and the size of the observed aerosol over the Reunion Island site, many LOAC (Light Optical Aerosols Counter) systems were launched together with balloon-ozonesondes. A detailed description of the LOAC is given by Renard et al. (2016). In brief, LOAC is a lightweight Optical Particle Counter (OPC) of 1 kg which can fly under latex weather balloons. Through the measurements of the light scattered by particles at two specific angles (Lurton et al. 2014), the LOAC provides aerosol concentrations and particle size distributions for 19 size classes ranging from 0.2 µm to 50 µm in diameter every ten seconds with a vertical resolution of nearly 50 m depending on the ascent rate of the balloon. The number concentration range is from 0.6 to a few thousand particles per cm^3 (Vignelles, 2017). Uncertainties on number concentration during the ascent under meteorological balloon are mainly due to temperature variation effects on electronics (Renard et al., 2016, Vignelles, 2017). Uncertainties on number concentrations for size bins smaller than 1 µm is estimated to be ± 30 %. For larger size bins, uncertainties on number concentration are governed by Poisson statistics and estimated to be ± 20 % and ± 60 % at particle concentrations lower than 10^{-1} and 10^{-2} cm^{-3}.

2.1.3 CALIOP

The Cloud-Aerosols Lidar with Orthogonal Polarization (CALIOP) on board The Cloud-Aerosols Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) was used to study the transport of the Calbuco plume. CALIPSO was launched to a Sun-synchronous polar orbit in 2006 (Winkler et al., 2009) with a repeat cycle of 16 days. CALIPSO is composed of an Infrared Imager Radiometer (IIR), a wide field visible camera and the CALIOP LiDAR. CALIOP is a two-wavelength polarization-sensitive LiDAR (532 and 1064 nm) which measures total attenuated backscatter vertical profiles with altitude-varying vertical (30-300 m) and horizontal (300-5000 m) resolution (Winker et al., 2009). The data used in this study are the total and perpendicular backscatter coefficient at 532 nm, available from the CALIOP level 1B V4.01

product. These data have been averaged every 1 degree in latitude for each orbit and grouped into data files containing 16 days of measurements. From there, the scattering ratio and depolarization ratio at 532 nm have been calculated (Vernier et al., 2009). Through the use of this algorithm, the full zonal means of scattering ratio between 20°S and 20°N are obtained by averaging 7200 cells, leading to a precision of ± 1.6 % (Vernier et al., 2009). The ability of CALIOP to detect small volcanic plumes in the lower tropical stratosphere has been highlighted in previous studies (Thomason et al., 2007; Vernier et al., 2009, 2011).

2.1.4 IASI

The Infrared Atmospheric Sounding Interferometer (IASI) observations were used to quantify the amount of SO_2 emitted during the Calbuco eruption. IASI is a nadir view thermal infrared sounder on board the Meteorological Operational satellite (MetOp-A and MetOp-B). The IASI observations used in this study were realized from the MetOp-A platform which launched in October 2006. The IASI global spatial coverage and footprint of 12 km make it relevant for monitoring of the key atmospheric species, in particular for the volcanic SO_2 (Clarisse et al., 2008, 2012; Clerbaux et al., 2009). The amount and altitude of emitted SO_2 were obtained from the algorithms detailed in Clarisse et al. (2012) and Clarisse et al. (2014) respectively. For each IASI observation, the altitude was estimated first, after which the column was calculated using the altitude information as an input parameter.

2.1.5 OMPS

The Ozone Mapper and Profiler Suite (OMPS) Limb Profiler (LP) is also used in the present study to analyze the optical properties of the volcanic plume over the Reunion Island site. OMPS was launched on October 2011 on board the Suomi National Polar Partnership (NPP) spacecraft. The data used in this study are the daily extinction profiles at 675 nm. A detailed description of the aerosol extinction retrieval algorithm is given by Jaross et al. (2012) and Rault and Loughman (2013). Briefly, the aerosol extinction profiles are retrieved from the scattering solar radiation. The aerosol extinction are retrieved using spectral channels with weak gaseous absorption. Rodgers' maximum likelihood technique is used to retrieve the aerosol extinction profiles independently for each wavelength s (Taha et al., 2011). We used 2 years (From November 2014 to November 2016) of satellite overpasses above the LiDAR site, within a 5°x5° in latitude and longitude grid. OMPS data have already used to be very effective at detecting and characterizing major events, such as the Chelyabinsk bolide in February 2013 (Gorkavyi et al., 2013).

2.2 MIMOSA model

The Modèle Isentropique de transport Mésoéchelle de l'Ozone Stratosphérique par Advection (MIMOSA) model (Hauchecorne et al., 2002) is a Potential Vorticity (PV) advection model running on isentropic surfaces (surface of constant potential temperaure). The advection scheme is semi-Lagrangian with a time step of 1 hour. The re-gridding onto the original orthonormal grid is performed every 6 hours. The model resolution is 0.5°x0.5°. The advection is driven by ECMWF meteorological analyses at a resolution of 0.5°x0.5°. In the case of the PV, its slow diabatic evolution is taken into account by relaxing the model PV towards the PV calculated from the ECMWF fields with a relaxation time of 10 days. Using this procedure, it is possible to run the model continuously and follow the evolution of PV filaments for several months. The accuracy of the model has been evaluated by Hauchecorne et al. (2002) and validated against airborne lidar ozone measurements using a correlation between PV and ozone, a quasi-conserved chemical tracer on timescales of a week or so within most of lower stratosphere (Heese et al., 2001; Jumelet et al., 2009). The MIMOSA model can also be used to determine the origin of air masses influencing a given site, similar to an isentropic Lagrangian trajectory model. The MIMOSA model is frequently used to detect the origin of air masses inducing laminae on ozone profiles (Hauchecorne et al., 2002; Godin et al., 2002; Portafaix et al., 2003).

2.3 DyBAL code

The Dynamical BArrier Location (DyBAL) code is an original software developed at the Laboratoire de l'Atmosphere et des Cyclones (LACy, France) to detect barriers to mixing in the subtropical stratosphere (Portafaix et al., 2003). The dynamical barriers are detected from the equivalent length of the tracer contour and the gradient of isentropic Ertel's potential vorticity (PV) in equivalent latitude coordinate as defined by Nakamura (1996). These two diagnostic tools are used by DyBAL to identify weak mixing and transport barriers. 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). The DyBAL code is applied to the PV map obtained from the MIMOSA model runs. The ability of DyBAL to detect the position and the deformation of the dynamical barriers has been highlighted in previous studies (Bencherif et al., 2007; Morel et al., 2005; Portafaix et al., 2003).

3. Long-range transport and evolution of the Calbuco volcanic plume over the Indian Ocean

3.1 Plume formation and transport

3.1.1 SO_2 plume

After 43 years of inactivity, the Calbuco erupted on 22 April 2015 and two intense explosive events were recorded during the same week. The evolution of the SO_2 total mass measured by IASI between 23 April 2015 and 31 May 2015 is reported in Figure 1. The SO_2 total mass is defined as the sum of SO_2 mass over the atmospheric column from midday to midnight over the southern hemisphere. As expected an increase of the SO_2 amounts was observed by IASI a few days following the Calbuco eruption. One day after the eruption the SO_2 total mass was 10 times higher than background levels. The SO_2 total mass increased quickly to its maximum value (0.41 Tg) on 25 April 2015 and slowly decreased to reach values close to the background values on 19 May 2015 (Fig. 2). The SO_2 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 (Jégou et al., 2013). The SO_2 total mass increased again on 28 May 2015 to reach a secondary maximum (0.13 Tg) on 30 May 2015. This new increase of the SO_2 total mass could be due to the Wolf eruption (Isabela Island, Galapagos) which occurred on 25 May 2015 (Xu et al., 2016). The amount of SO_2 emitted during the Calbuco eruption is about two times lower than the SO_2 mass emitted from the Sarychev eruption (0.9 Tg) in June 2009 (Jégou et al., 2013). It is also worth noting that the SO_2 mass injected during the Calbuco eruption is of the same order as for the Grimsvötn eruption in May 2011 (Clarisse et al., 2011). Figure 2 also depicts the maximum altitude of SO_2 over the period from 23 April 2015 to 31 May 2015. On average the maximum altitude of SO_2 is located in the lower stratosphere region around 17 km.

The SO_2 measurements integrated from midday to midnight obtained from IASI are also used to describe the transport of the volcanic plume over the southern hemisphere (Fig. 2). On 23 April 2015, a part of the Calbuco plume passed close to the Uruguay coast at an altitude of 17 km and then was transported by the general circulation. The plume reached Southern Africa and East side of Madagascar on 1 May 2015 at altitude of 17-18 km and was organized following a cyclonic rolling (Fig. 2b). On 6 May, the plume is mainly located over the Indian Ocean near the east coast of South Africa and partly over Namibia and South Africa. As expected, the SO_2 plume extent and amplitude 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. The plume was embedded in a thin 15-17 km altitude atmospheric layer, extending from the Atlantic Ocean to the Indian Ocean passing over the Cape of Good Hope (Fig. 2d).

3.1.2 Spatial extent of the aerosol plume

The transport of the volcanic aerosol plume over the southern hemisphere can be followed by CALIOP observations at 532 nm. Figure 3 shows the CALIOP cross-section of the 532nm total attenuated backscatter (TAB) for the overpass over South America on 24 April. The TAB signals ranging from 1 10^{-3} to 5 10^{-3} km^{-1} sr^{-1} corresponding to weak values of brightness temperatures over the southern part of Brazil (34.22°S; 53.97°W) can be attributed to volcanic material injected up to the lower stratosphere by the Calbuco eruption. Figure 4 and 5 present the latitude-altitude cross sections of the scattering ratio observed by CALIOP for 16-day selected periods in 2015. The data displayed in Figure 4 and 5 correspond to the zonal mean averages of CALIOP scattering ratio during 16-day periods. The data are calculated within 1° latitudinal zonal bands and have about 9% precision (Vernier et al., 2009). The scattering ratio values observed during the 16-30 April period (before the eruption) in the Southern hemisphere, particularly in the lower stratosphere, were in average at 1.05 (not shown). Between one and three weeks after the eruption (1-16 May period), CALIOP observations reveal that SR increased up to 1.12 in the southern lower stratosphere (Fig. 4a). The amplitude of the plume during the first weeks following the eruption was higher than the background aerosol levels at mid-latitudes but was still below the scattering ratio values observed in the tropics. The first weeks following the eruption correspond to the period when the SO_2 is still being converted (Fig. 1). The elevated backscatter in the tropics could be attributed to possible remnants of the Kelud (7.5°S; 112.2°E; erupted in February 2014) volcanic aerosol superimposed to the equatorial background aerosol layer (Kristiansen et al., 2015). About one month after the eruption (16-31 May period) the Calbuco plume was much more pronounced with scattering ratio values (ranging from 1.16 to 1.18) largely above values observed before the eruption from CALIOP and greater than aerosol amounts confined in the tropical reservoir (Fig. 4b). The second half of May (16-31 May) corresponds to the period where the SO_2 has been oxidized to aerosol. The plume extended up to about 20 km in altitude and spread over a wide range of latitudes, nearly reaching 60°S and intruding into low latitudes near 5°S. About one month later (16-30 June), the plume top had moved upward by several hundred meters and the layer was thicker (Fig. 5a). The southern hemisphere between 10°S and polar latitudes was full of volcanic aerosol with scattering ratio values much higher than elsewhere in the whole stratosphere. About four months after the eruption (16-31 August) the volcanic aerosol layer was even thicker with scattering ratio remaining high (Fig. 5b). Figure 5d also reveals a deepening of the volcanic aerosol layer and an enhancement of the equatorial backscatter in the Upper Troposphere. In the 21-28 km altitude range, detrainment of aerosol from the equatorial reservoir depends upon the phase of the quasi-biennial oscillation (QBO) and on the intensity of planetary wave activity (Trepte and Hitchman, 1992). Through the use of numerical model, Pitari et al (2016b) discussed on the impact of the QBO phase on the meridional transport of the aerosols plume to mid- and high latitudes. They revealed that the volcanic aerosols is confined to the tropics when the volcanic eruption occurred during the easterly shear of the QBO. When QBO easterlies descend in the tropics, propagation of planetary waves is inhibited from entering this region, thus limiting the extent to which these waves may detrain aerosol laterally from the tropical reservoir (Trepte et al., 1993). This corresponds to the situation in April-May, 2015 (Figure 4a, b). However, during the westerly phase of the QBO, mixing across the subtropics is favored, especially in winter (Trepte et al., 1993). The meridional spread in aerosols to southern midlatitudes shown above 21 km in Figure 5a and 5d is consistent with the phase reversal of the QBO from easterlies to westerlies observed from mid-2015.

3.2 Evolution of the aerosols plume over the Reunion site

3.2.1 Ground-based and satellite observations

Figure 6 depicts the evolution of the stratospheric AOD (sAOD) at 532 nm calculated between 17 and 30 km from the Reunion ground-based LiDAR and OMPS observations over Reunion Island from November 2014 to November 2016. The wavelength conversions to 532 nm were performed using the Angström exponents where detailed description of the methodology is given by Khaykin et al (2017). The Angström exponents for the 355-532 nm and 532-675 nm pairs were adopted from Jäger and Deshler (2002) and Khaykin et al (2017) set to -1.3 and -1.8 respectively. sAOD were calculated from LiDAR and OMPS observations at 532 nm using Angstrom exponent mentioned previously. As expected, an increase in the aerosol loading was observed over the Reunion site a few weeks after the Calbuco eruption. OMPS data show a doubling in the sAOD record in comparison with values observed at the end of 2014 and at the beginning of 2015 (Fig. 6). sAOD reached its maximum values (0.014 for OMPS) at the beginning of June 2015, decreased afterward to 0.01 on August 2015 and went back to pre-eruption values (0.004-0.006) in April 2016. The LiDAR record peaks at the same period, but sAOD values are 1.2 times weaker than those observed by OMPS during the June-December period. The lidar sAOD observations show less difference with values obtained prior to the eruption (0.008). Discrepancies between OMPS and LiDAR were significantly reduced by April 2016, (cf. relative differences of 25% over the January-December 2015 period and 10% over the April-November 2016 period). From both datasets an aerosol e-folding of approximately 90 days can be derived, which is rather close to the value (~80 days) reported for the Sarychev eruption (Jégou et al., 2013).

Figure 7a illustrates the weekly-averaged extinction profiles at 532 nm derived from LiDAR measurements over Reunion Island. This figure reveals a sharp increase of the extinction between 18 and 19 km in May 2015 and reaching its maximum value (greater than 4 $x10^{-3}$ km^{-1}) in June. The vertical extent of the plume had increased significantly over the May-July period with a volcanic aerosol layer spanning from 18 to 21 km. At the beginning of June, the plume was structured in two layers with the first one centered at 18.5 km and the second one at 20 km (Fig.7a). We note also a brief decrease in the local extinction, ranging from 1.5 to 3 km^{-1}, around mid-May. The variability observed in the weekly-averaged extinction profiles and in the vertical extent of the aerosol signal over the May-July period reflects the presence of transient aerosol layers above Reunion Island and indicates that the plume is not homogeneously distributed at this stage. The altitude of the volcanic aerosol plume and the extinction values decreased from mid-August onwards. The plume is hence centered around 18 km in September, and extinction values in September are around two times less than those observed in June. This decrease of the extinction values is accompanied by a decrease of the altitude of the plume and could be due to the sedimentation processes (Fig. 7a). Hamil et al (1997) revealed that sedimentation can play a significant role in loss of stratospheric aerosol to the troposphere. Overall, the temporal evolution of the weekly-averaged extinction presents similar general features as the LiDAR observations, with maximum values in June and a subsequent gradual decrease of the aerosol signal. Nevertheless, in the OMPS data the plume is smeared out over a wider vertical range than in the lidar record (Fig. 7b). The vertical and horizontal structures of the plume are not reproduced in the OMPS data. In particular, the decrease in the plume altitude in mid-August is not observed by OMPS. More generally, extinction values observed by OMPS in the 15-17 km altitude range are higher than those observed by the LiDAR. The evolution of the scattering ratio at 532 nm obtained from the LiDAR and CALIOP space-borne observations during the April-December 2015 period over the Reunion Island site are presented on Figure 8. The scattering ratios from CALIOP have been averaged within ± 5°in latitude and ± 50° in longitude (extending from Africa to Australia) around Reunion Island (Fig. 8b). CALIOP observations confirm the presence of the volcanic aerosol plume over the Reunion

Island site at the beginning of May 2015 with maximum scattering ratio values (greater than 1.9) on mid-May 2015. Overall, the aerosol variability is smoother in CALIOP observations than in the LiDAR record, which shows more fluctuations in the altitude of the volcanic plume. In contrast to the LiDAR and OMPS observations, CALIOP data do not show an increase in the vertical extent of the plume and maximum scattering ratio values at the beginning of June 2015. According to CALIOP, the scattering ratio begins to decrease in mid-June followed by a slight decrease of the altitude of the plume from the end of July (Fig 8b). From July onwards, the CALIOP aerosol scattering ratios decrease gradually with similar values as observed by the LiDAR. The decrease of the aerosol scattering ratio is associated with a descent in the altitude of the plume, which could be due to sedimentation, and is consistent with the observations from OMPS.

The reasons for these discrepancies between ground-based and satellite observations may be multiple but effects due to different spatial samplings cannot be excluded. 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). 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. Given that the weak horizontal resolution of CALIOP (500 km) (Vernier et al., 2011), it is consistent to observe weaker values than the ground-based LiDAR. As we will discuss more details in Section 4, the dynamical context can induce an inhomogeneity of the plume. As a consequence, 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 ash ash to be incorrectly identified as “ice nuclei” and to be then removed.

3.2.2 In-situ observations

Four LOAC OPCs were launched over the Reunion Island site on 26 November 2014, 19 May 2015, 19 August 2015, and 2 November 2016 respectively. sAOD 532 nm were calculated following Mie Theory from fits to the observed size distributions at each level. LOAC observations also reveal an increase by a factor of 2 in the sAOD on 19 May 2015 (1.35×10^{-2}). sAOD decreases to 8.4×10^{-3} on 19 August 2015 followed by a return to pre-eruption levels by November 2016 (Fig. 6). The overall evolution of sAOD derived from LOAC compares fairly well to OMPS and to lidar observations, accounting for error bars (Fig. 6). The 10-second sampling rate of the LOAC instrument and the ascent velocity of the balloon determine the vertical resolution. The difference observed between the daily-integrated LiDAR and the in-situ data may be due to the 1-minute averaging of the in-situ data, which tends to smooth structure attributed to the volcanic aerosols.

Figure 9 illustrates the number (dN/dln(D)) and volume (DV/dln(D)) concentrations obtained from the LOAC OPC observations over the Reunion Island site on 19 May 2015 at 1746 UTC. LOAC OPC observations reveal a size distribution with decreasing concentrations for particle sizes larger than 0.2 µm (Fig. 9). The value of 0.2 µm represents the lower bound of the LOAC size range, so the data may miss possible secondary modes for smaller particles (Wilson et al., 2008). Particle concentrations for sizes larger than 1 µm too low to properly detected by the LOAC OPC but undoubtedly indicate a coarse mode. However there is no clear evidence for a bimodal distribution, as observed during the first weeks after the eruption of Mt. Pinatubo, an intense second mode possibly consisting of volcanic ash (e.g. Russell et al., 1996). The shape of the size distribution obtained during the Calbuco event is similar to that obtained by Kravitz et al. (2011) for the Sarychev eruption. As suggested by Kravitz et al. (2011), we can also assume that the Calbuco eruption did not eject enough material to create a bimodal structure over Reunion Island.

The effective radius derived from the LOAC OPC on 19 May is 0.17 ±0.02 µm indicating that the particles observed several weeks after the Calbuco eruption are quite small. Interestingly, Jégou et al. (2013) reported that the effective radius obtained during the Sarychev event ranged from 0.15 to 0.20 µm more than one month after the eruption, in agreement with the results of O’Neill et al. (2012). Therefore, both eruptions are comparable in terms of size distribution shape and effective radius. Russell et al. (1996) reported that in the month following the

Pinatubo eruption the mean effective radius did not differ greatly from pre-eruption values (i.e. 0.17 ± 0.07 µm in their study), possibly because a large number of particles with sizes both smaller and larger than 0.17 µm were injected (the latter consisting most likely of volcanic ash). Russell et al (1996) discussed how particle growth processes (condensation and coagulation), may be compensated by particle loss, which tends to decrease mean effective radius. This could explain the weak evolution on the effective radius during the month following the Pinatubo eruption.

The integrated number of particles obtained over the full 19 size classes from 0.2 to 2 µm in diameter is presented in Figure 10. A local aerosol concentration enhancement is detected in the lower stratosphere (16.8-19 km) over Reunion Island on 19 May 2015 at 1746 UTC. A maximum concentration of about 150 particles per cm^3 (total number of particles: 730 ± 130 particles (±1σ)) is observed by the aerosol counter for particle sizes larger than 0.2 µm. Few in situ observations are available in the tropical region such as Reunion Island to provide a reference state of the background aerosol content. In comparison to LOAC flight on 26 November 2014 at 1442 UTC (five months before the eruption), the aerosol number concentration observed on 19 May 2015 is ~20 times higher. Three months later, another LOAC OPC was launched over the Reunion Island site but the in situ profile only partially shows the volcanic aerosol layer because of a telemetry loss. The aerosol number concentration obtained on 19 August 2015 at 1300 UTC over Reunion Island may reveal a tendency to return to concentration values observed before the eruption (Fig. 10), with a number concentration of 40 particles per cm^3 in the lower stratosphere. This tendency is confirmed with the LOAC flight conducted on 2 November 2016 at 2030 UTC with a total number concentration close to 20 particles per cm^3 in the lower stratosphere (Fig. 10).

The residence time of the aerosol particles in the stratosphere depends on the balance between the growth processes and the removal processes which are likely to be controlled by the dynamical context. In the following section, we will discuss the influence of the dynamical activity on the variability of the volcanic aerosol over the southern hemisphere.

4. Dynamical modulation of the aerosol plume

4.1 Long-range transport

In order to analyze the isentropic transport, the high resolution MIMOSA model has been used to produce a continuous evolution of PV fields for the period from 1 April 2015 to 31 August 2015. Four advected PV maps, derived for the 400 K isentropic level from the MIMOSA model, together with dynamical barrier locations derived from the DyBaL code are superimposed in Figures 11 and 12. The localization of the volcanic aerosol plume obtained from OMPS observations at 400 K ± 5 K isentropic level is also superimposed (Fig. 11 and 12). On 24 April 2015, a significant wave activity is observed, leading to a fairly mixed surf zone in the 20°S-60°S latitude band (not shown). The Calbuco plume is situated inside the surf zone and the plume was mixed equatorward. On 27 April 2015, the subtropical and mid-latitude barrier are detected following the Nakamura's formalism (described in Section 2.3) around 15°S (red line, Fig. 11) and 40˚S (blue line) in latitude respectively, limiting the geographical extent of the plume (Fig. 11a). Figure 11a shows clearly that the air masses containing aerosols cannot move beyond the south of Brazil because of the presence of the subtropical barrier. On 01 May 2015, the air masses were confined between the two dynamical barriers located in average at 25˚S (red line, Fig. 11b) and 40˚S (blue line, Fig.11b) in latitude respectively. The air masses were advected eastward between South Africa and Madagascar following the wave shape of the barrier, consistent with the OMPS observations near South Africa (Fig. 11b). The subtropical barrier previously located at 25˚S (red line, Fig.11b) moved northward crossing South Africa. The air masses containing aerosol previously situated in the south side of Madagascar were transported northward and eastward following the displacement of the barrier and reached the Reunion Island site.

On 19 May 2015 (Fig. 12a), the volcanic aerosol plume was confined between the two dynamical barriers and advected eastward. At this stage, the presence of the subtropical barrier and the polar vortex seems to constrain the Calbuco plume inducing its transport eastward. Between end of May and beginning of June, the subtropical barrier has dissipated while the edge of the polar vortex was around ~40˚S (blue line, Fig. 12b). The OMPS observations reveal that the most part of the plume was located over the southern African and the Indian Ocean region in June (Fig. 12b). On the following months of July and August, the polar vortex is clearly identified at 60˚S (blue line, Fig. 12b) which is a classical pattern for the austral winter.

This present study discuss only on the transport of the volcanic aerosols plume at 400 K isentropic level (isentropic level where the Calbuco plume is detected 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 above is possible. Figure 5b reveals also the possibility to the Calbuco aerosols 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. They reveal that the integrated additional Antarctic ozone column losses averaged over the polar cap are between 5 and 13 DU following the Calbuco eruption. Through the use of FR-WACCM (free-running Whole Atmosphere Community Climate Model), Ivy et al. (2017) shown that the forced response to the eruption of Calbuco was an increase in the size of the ozone hole by 4.5 10^6 km^2.

4.2 Removal processes

As discussed above, in the lower stratosphere distributions of the aerosol are modulated (or mostly driven) by isentropic transport. However, particle removal processes should be considered. As reported by Kremser et al (2016), aerosol in the vicinity of the tropopause can be transported into the troposphere by variety of mechanisms. Hamill et al. (1997) reported that stratosphere-troposphere exchange (STE) on isentropic surfaces due to Rossby wave activity can be considered a significant dynamical process for removal of stratospheric aerosol. Depending on the strength of the Brewer-Dobson circulation, stratospheric materials such as aerosols can be rapidly transported from the tropics to high latitudes (Dhomse et al., 2006, 2014). Based on semi-Lagrangian and analyzed winds from ECMWF, Chen et al. (1995) investigated the extratropical STE on isentropic surfaces that intersect the tropopause. Above 340 K they found that STE exhibits a strong annual cycle where very little STE takes place in the winter hemisphere, but significant STE occurs in the summer hemisphere, particularly in the northern summer. The weak STE in the winter hemisphere is mainly due to the barrier effect of the strong PV gradient at the tropopause (Chen et al., 1995). Through the use of 10 years of LiDAR observations at Pasadena (34°N, 118˚W; California), Menzies and Tratt. (1995) found a clear link between the aerosol optical properties in UT-LS and the active extratropical STE processes occurring the winter and early spring. The calculated stratospheric mass extrusion rate is consistent with a 45-day lifetime of lower stratospheric aerosol during this part of the year, which implies that extratropical STE is a significant sink for stratospheric aerosol (Menzies and Tratt, 1995). Given the potential of a STE event to impact the stratospheric aerosol loading, we cannot exclude its contribution (even though small) on the stratospheric aerosols loading at Reunion Island.

Dhomse et al. (2014) using the CCM model (UM-UKCA) discussed the influence of STE events on the budget of the stratospheric aerosol. In particular, they suggested that a general overestimation of STE in global composition climate models could lead to overestimated removal of aerosols from stratosphere into the troposphere. Through the use of ULAQ-CCM model, Pitari et al (2016b) shown that the efficiency of STE depends on large-scale transport following the down-welling branch of the Brewer-Dobson circulation together with gravitational settling. The long-range transport of a volcanic plume is less likely to be isentropic, due to sedimentation of ash or other large particles (with any accommodated sulphur) within the plume. The modulation of the plume over Reunion Island could be caused by particle removal processes such sedimentation (considered as the primary loss mechanism of stratospheric aerosol) or by dilution of the stratospheric plume (Hamill et al, 1997; Rasch et al., 2008). Sedimentation is an effective removal mechanism for particles that survive long enough in the stratosphere to grow to larger sizes (Hamill et al., 1997).

We note the potential role of removal processes on the initial dispersion of volcanic aerosols, in particular co-emitted ultrafine ash particles, but do not explore this effect here. Highlighting removal processes from Figures 7 and 8 is somewhat complicated by the transience in plume altitudes especially in the LiDAR local data. The potential role of removal processes on the evolution of the plume requires further investigation and will form the basis for a forthcoming study.

5. Summary and conclusion

The long-range transport of the volcanic aerosol produced following the Calbuco eruption has been examined. The analysis focuses on the dynamical context which led to the spread of the aerosol plume over Indian Ocean between April 2015 and November 2016. The transport of the volcanic aerosols to the Indian Ocean was investigated by combining satellite (CALIOP, IASI, OMPS), and ground-based experiments: Optical Particle Counter (LOAC) and lidar, in addition to numerical tools: the DyBal code and the high-resolution MIMOSA model.

The amount of SO_2 injected into the atmosphere during the Calbuco eruption has been quantified using IASI observations. SO_2 mass emitted by the Calbuco eruption was about two times lower than for the moderate northern hemisphere eruption of Sarychev in June 2009, but had a similar SO_2 e-folding time (Jégou et al., 2013; Kravitz et al., 2011). It is found from CALIOP observations that the Calbuco aerosol layer was observable in the lower stratosphere, between 18 and 21 km, and spread exclusively in the southern hemisphere. OMPS observations reveal that the Calbuco plume reached the Indian Ocean two weeks after the eruption. It is shown from ground-based observations deployed at Reunion Island that SAOD increased by a factor of ~2 by the beginning of May 2015 and decreased afterward, returning to pre-eruption values by November 2016. The aerosol e-folding time is estimated to be ~90 days, close to the ~80 days reported for the Sarychev eruption (Jégou et al., 2013). Though the various datasets agree in terms of aerosol signal intensity, we report significant differences for the plume height and its variability, possibly as a result of different observations geometries, resolutions and spatial scales inherent to each instrument.

In situ measurements by the LOAC OPC have pointed out the impact of the Calbuco eruption on the lower stratospheric aerosol content over the Reunion Island site. Aerosol number concentrations were 20 times higher than values observed before and one year after the eruption. On May 2015, the volcanic aerosol was characterized by an effective radius of 0.16 ±0.02 µm and a unimodal lognormal size distribution above 250 nm diameter. These microphysical characteristics are in agreement with previous studies focusing on the Sarychev eruption (Kravitz et al., 2011; Jégou et al., 2013).

Through the use of the MIMOSA model and the DyBAL code, it was clearly identified that the eastward transport of the volcanic aerosols occurred mainly in form of planetary-scale tongues. In particular, the combination of MIMOSA and DyBal simulations revealed that the transport of the volcanic aerosol plume eastward was confined between subtropical barrier and mid-latitude (polar vortex) dynamical barriers, within which most of the zonal transport took place. Our results support the assumption that the processes explaining the structure of the plume over the southern hemisphere had mainly a dynamical origin. Thus, the fluctuation of the subtropical barrier induced transient aerosol layers above Reunion Island and an inhomogeneous distribution of the plume between May and July 2015. The present study also supports the hypothesis that the modulation of a volcanic plume results from a contribution of both dynamical and microphysical processes. Fully understanding the contribution of the microphysical processes to the evolution of the volcanic plume over the southern hemisphere requires further investigation. This will be examined in a forthcoming study.

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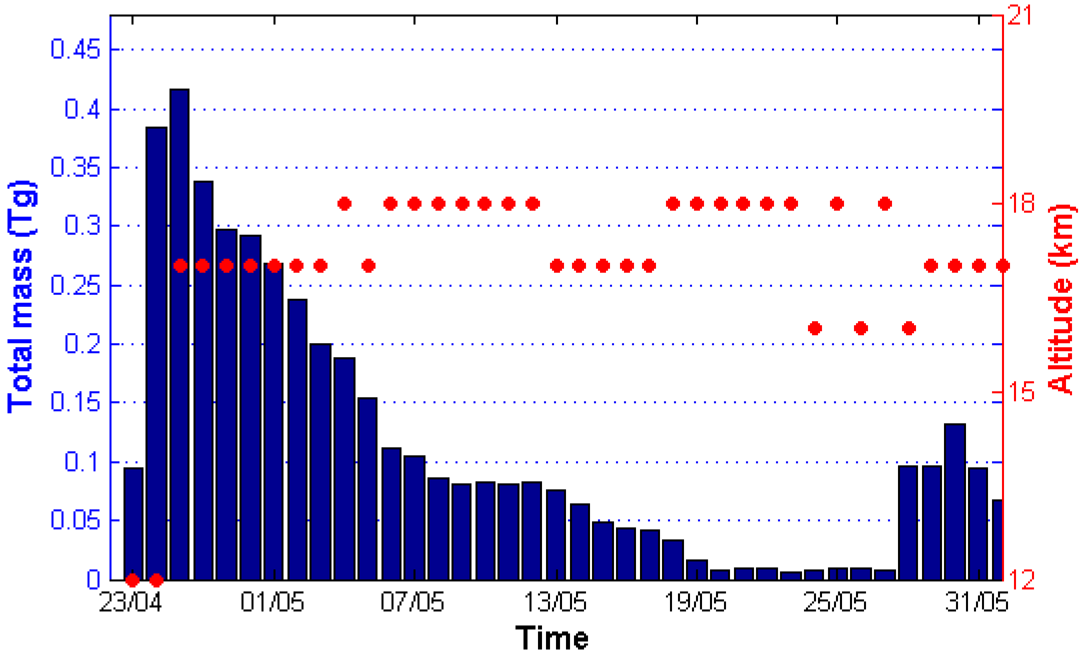


Figure 1 : Evolution of the SO_2 total column mass (in blue) and the altitude of the maximum SO_2 mass (red dots) obtained from IASI from 23 April 2015 to 31 May 2015 over the southern hemisphere. The altitude of the maximum SO_2 mass was obtained from the algorithms detailed in Clarisse et al. (2014).

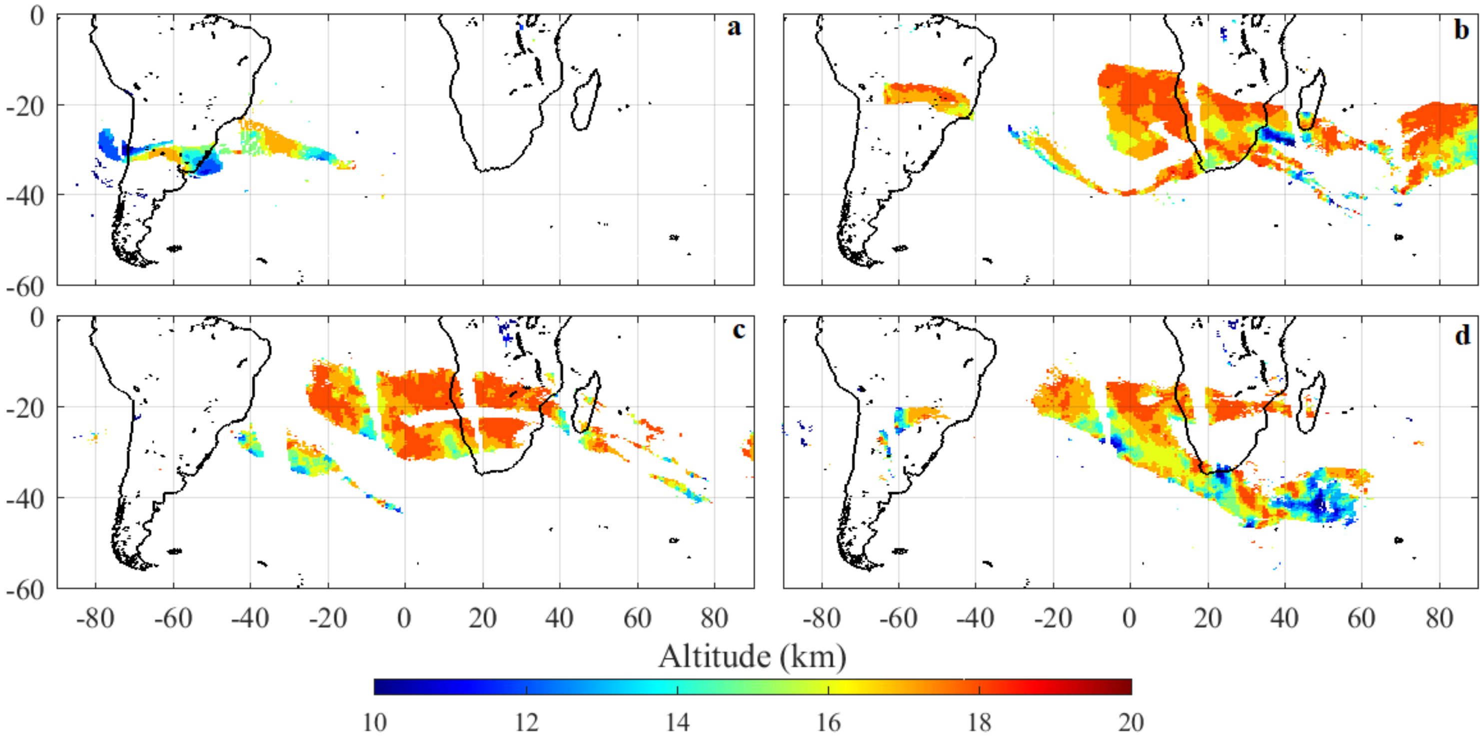


Figure 2 : Injection Height (km) and transport of SO_2 obtained from IASI observations during (a) 24/04, (b) 01/05, (c) 06/05 and (d) 11/05

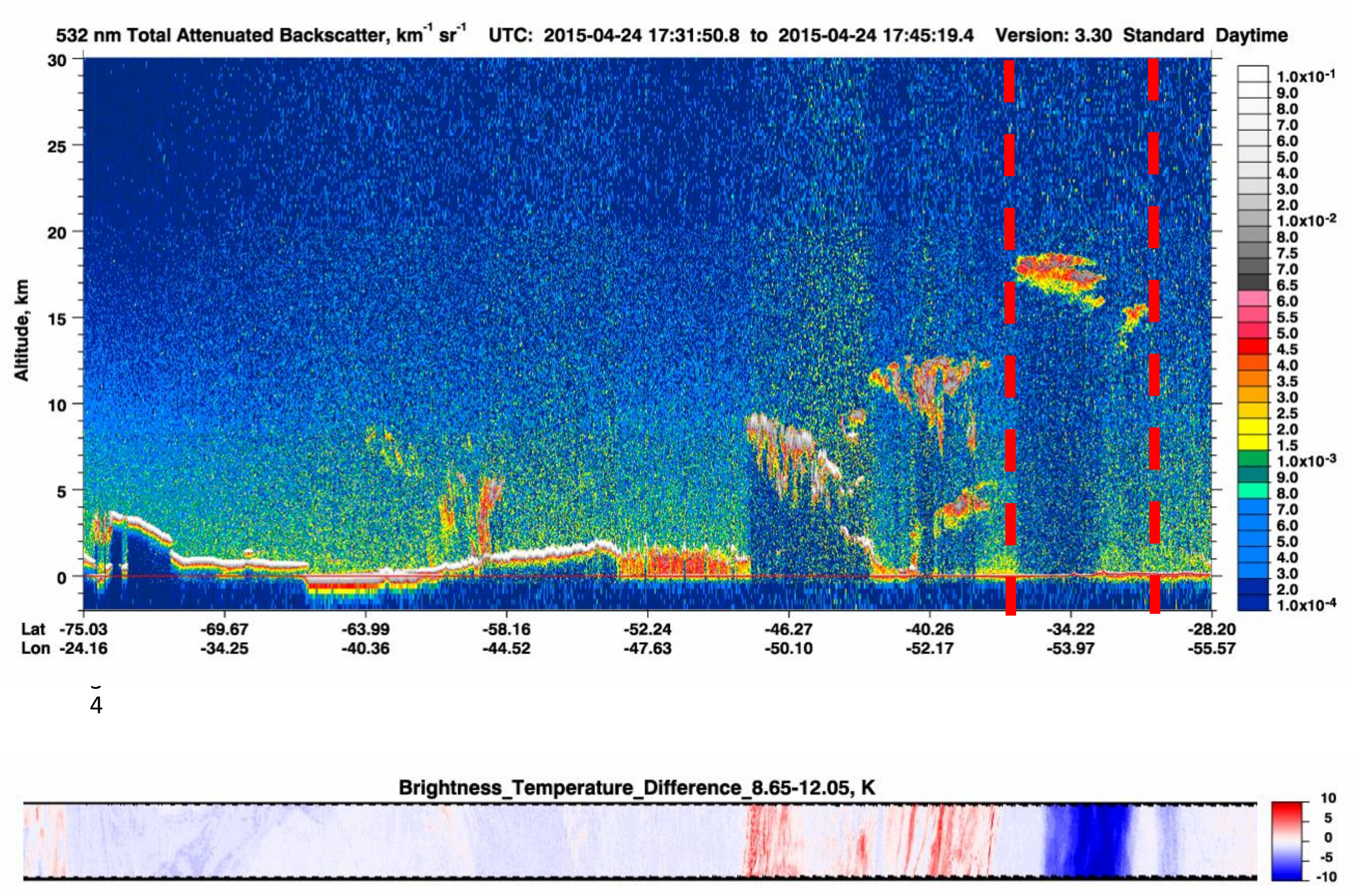
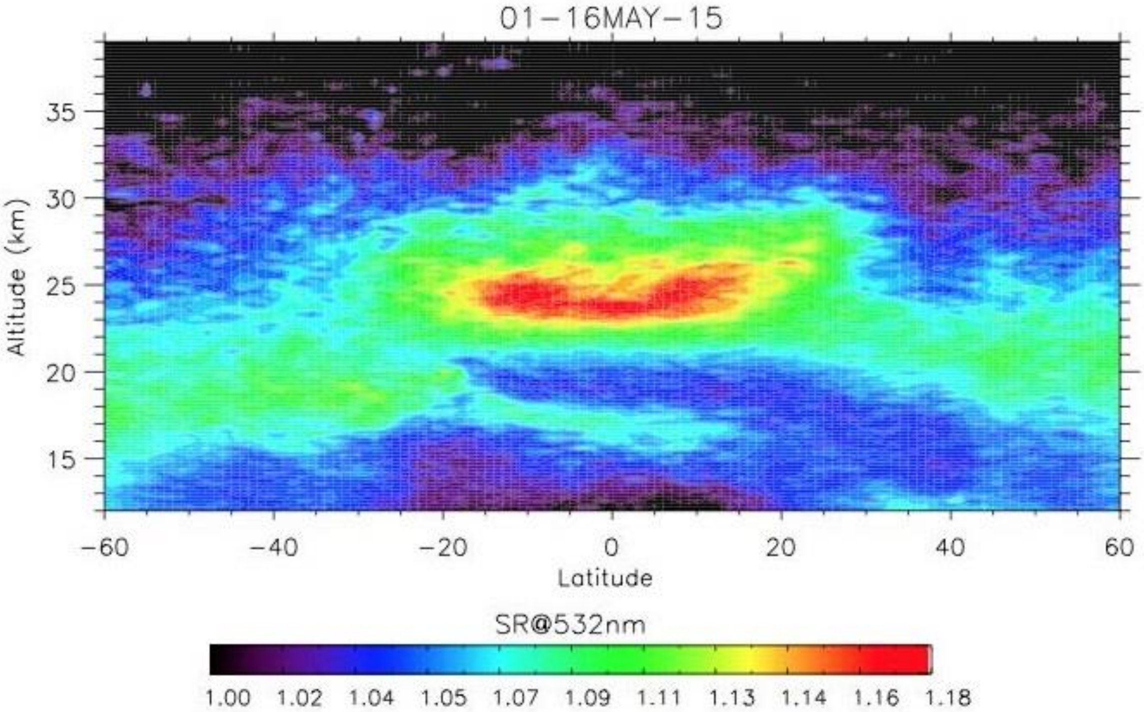


Figure 3 : CALIOP cross-section of 532 nm attenuated backscatter and Brightness temperature difference for the overpass at 1730-1745 on 24 April 2015 over the South America from 28˚S to 75˚S latitude range. The two red dash lines delimited the geographical region where the Calbuco plume is observed by CALIOP on 24 April 2015.

a)



b)

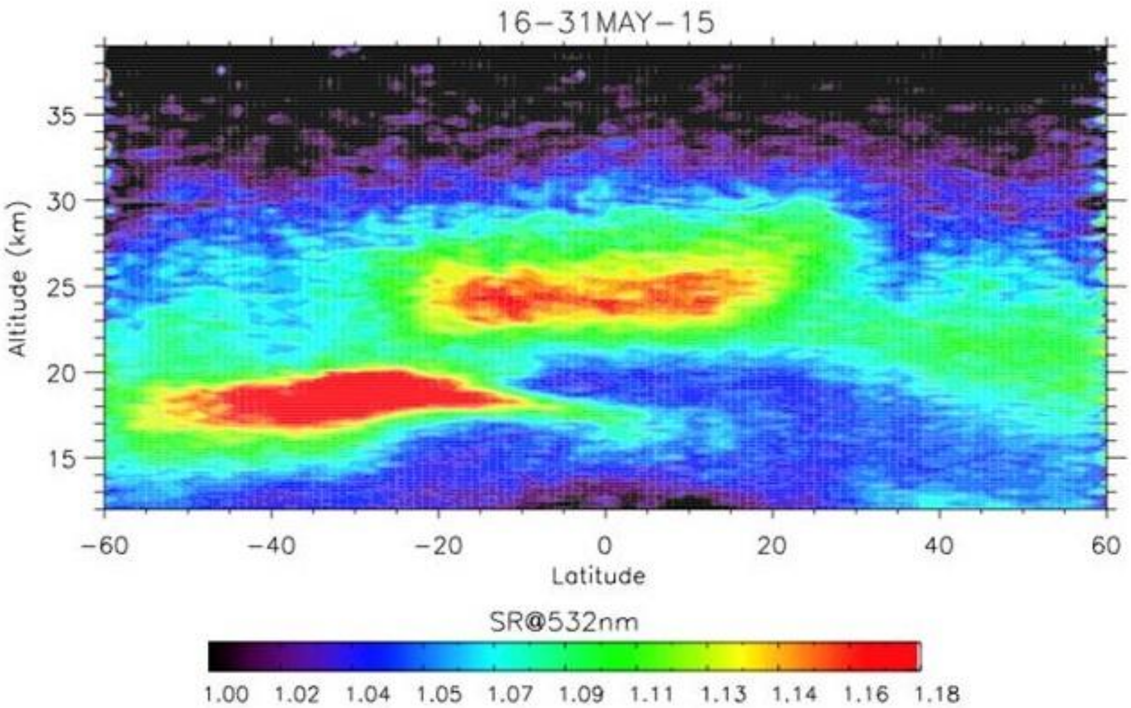
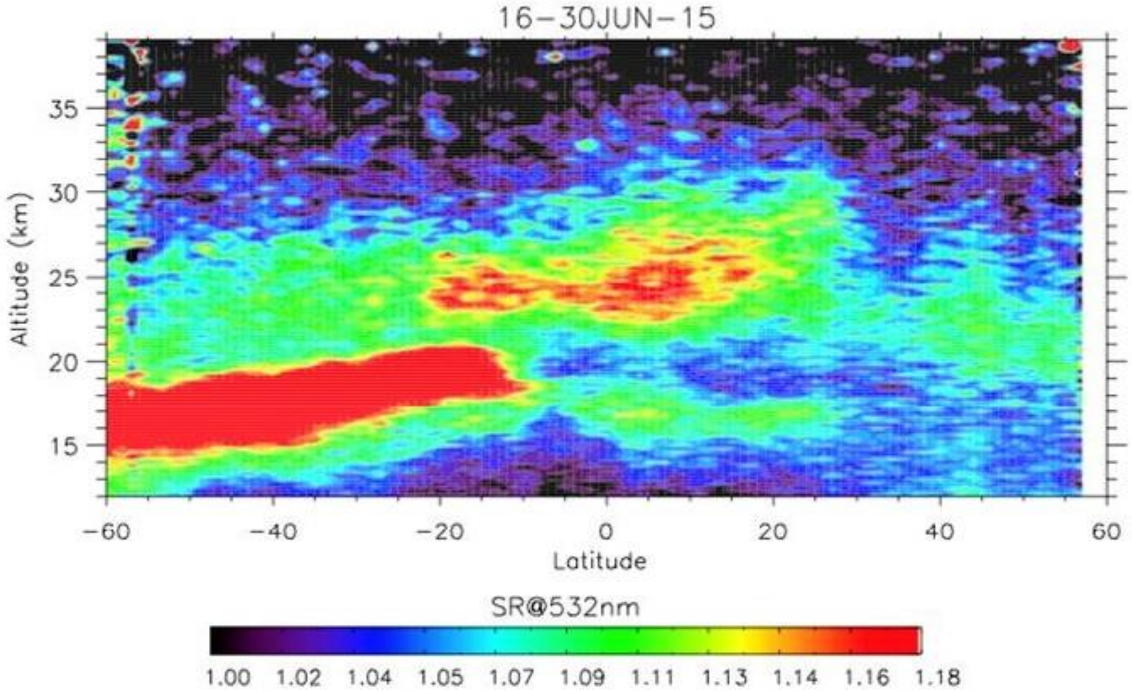


Figure 4 : Half monthly mean of the zonal scattering ratio at 532 nm in (a) 1-15 May (1-2 weeks after eruption), (b) 16-31 May (3-4 weeks after eruption).

a)



b)

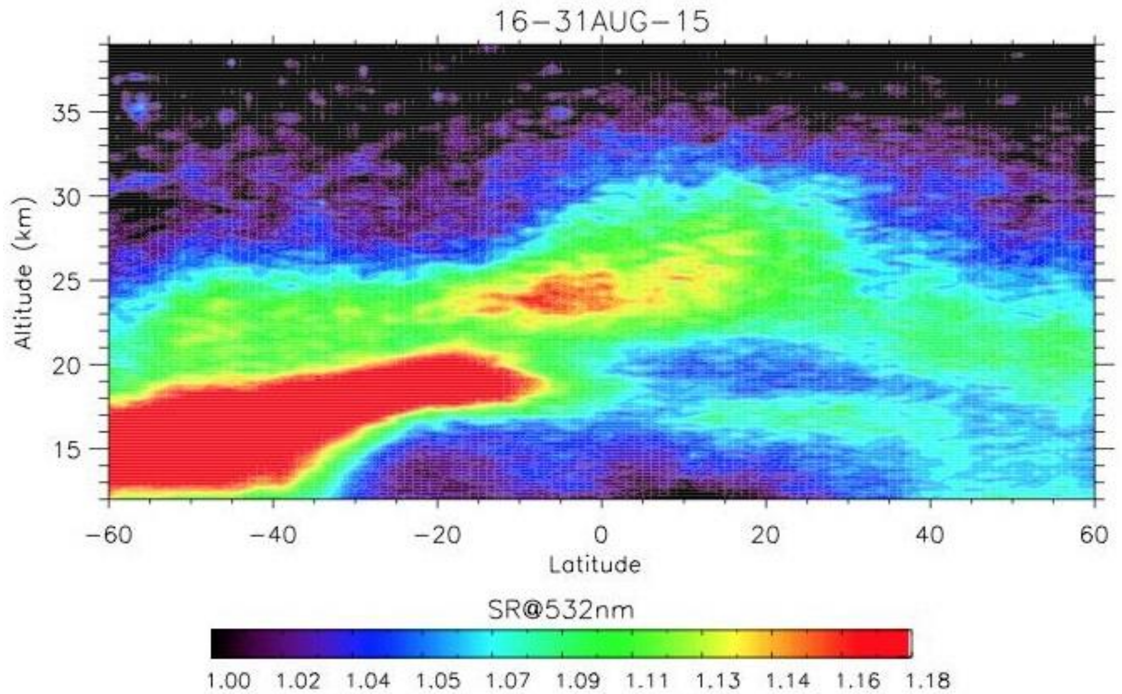


Figure 5 : Half monthly mean of the zonal scattering ratio at 532 nm in (a) 16-30 June (2 months after eruption) and (b) 16-31 August (4 months after eruption).

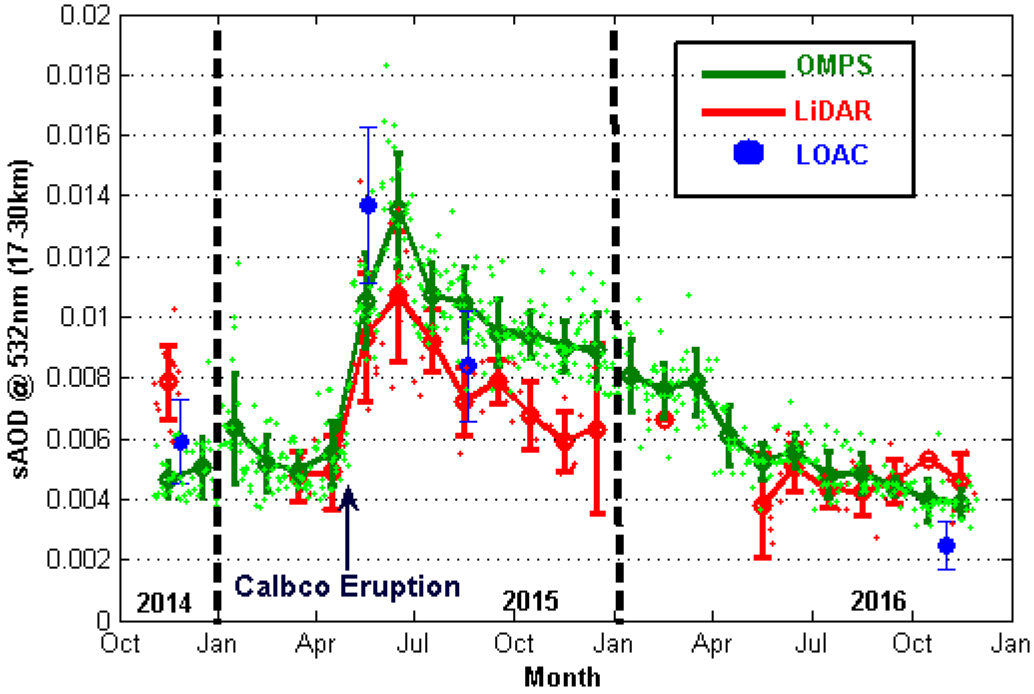
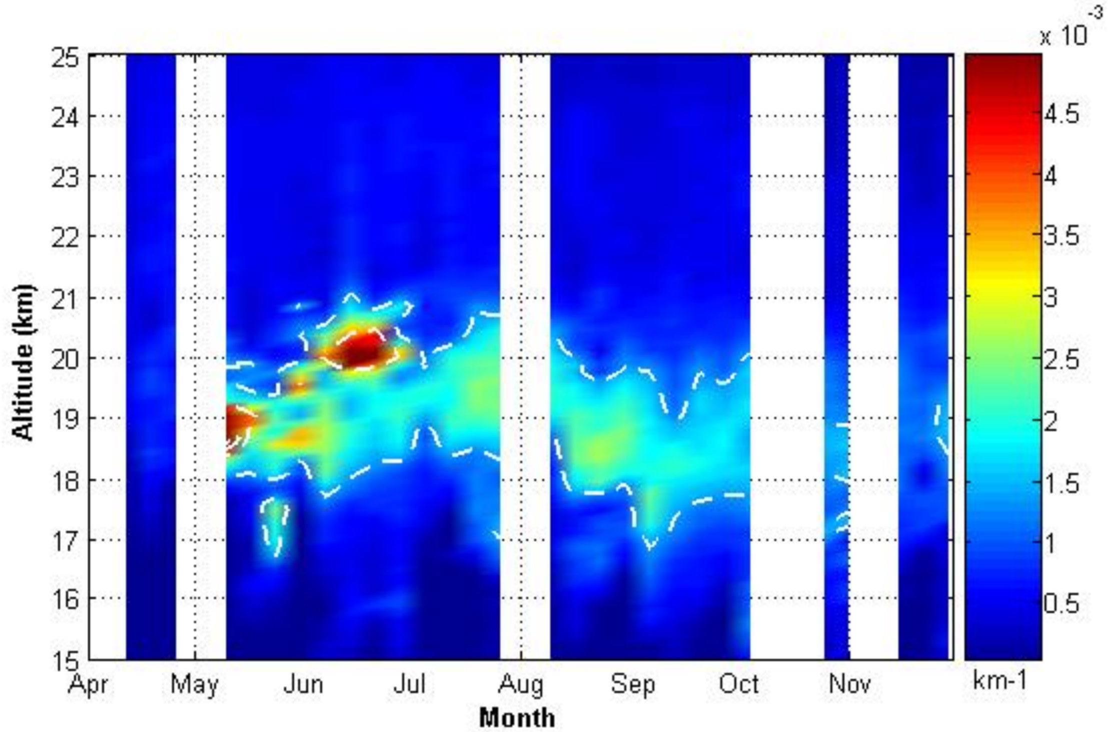


Figure 6 : Evolution of sAOD calculated between 17 and 30 km at 532 nm from LiDAR (red), LOAC OPC (blue) and OMPS (green) observations between November 2014 to November 2016 over the Reunion site. The small dots represent the daily sAOD and the large dots represent the monthly averaged sAOD obtained from OMPS and LiDAR observation. The large blue dots represent the sAOD calculated from LOAC OPC observations over Reunion during the 26 November 2014, the 19 May 2015, the 19 August 2015 and the 2 November 2016. The error bars associated to LiDAR and OMPS observations represent the standard deviation. The error bars associated to LOAC OPC represent the uncertainties values. The date of the Calbuco eruption is indicated by a blue arrow.

a)



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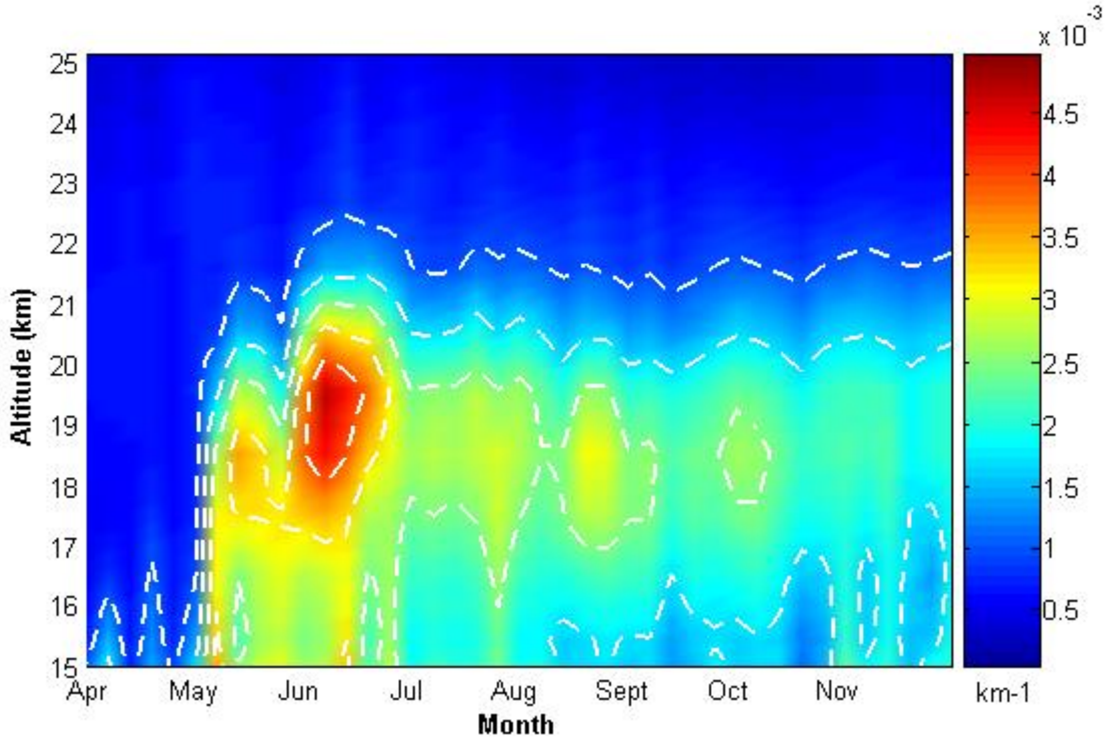
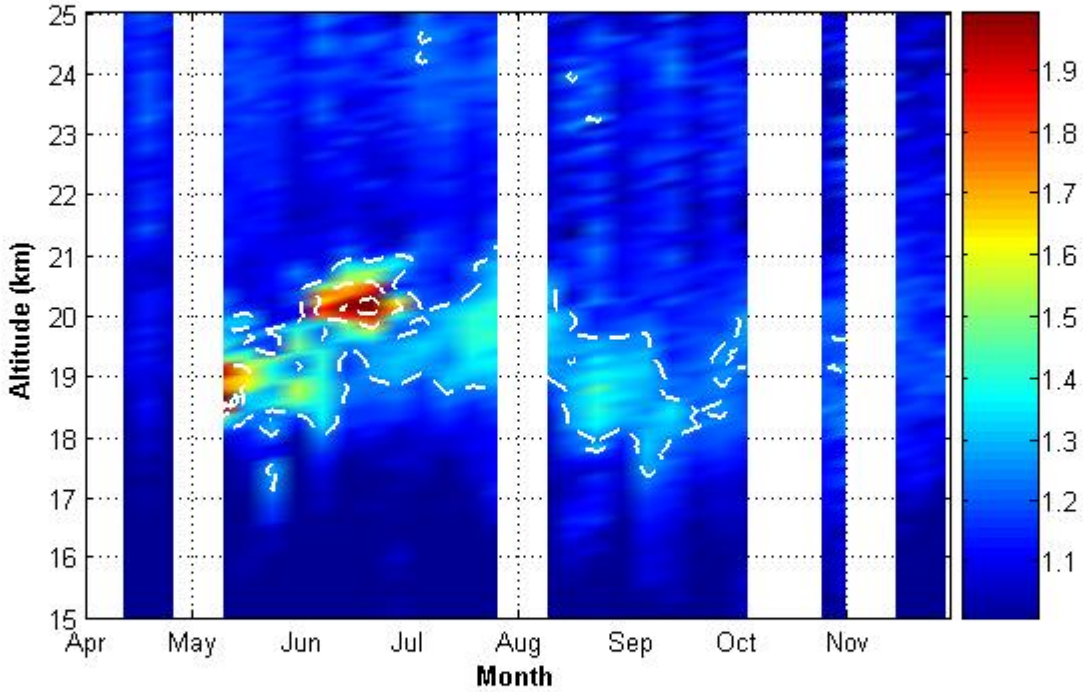


Figure 7 : Time series of weekly-averaged profiles of extinction at 532 nm obtained from (a) lidar and (b) OMPS observations over Reunion between April 2015 and December 2015.

a)



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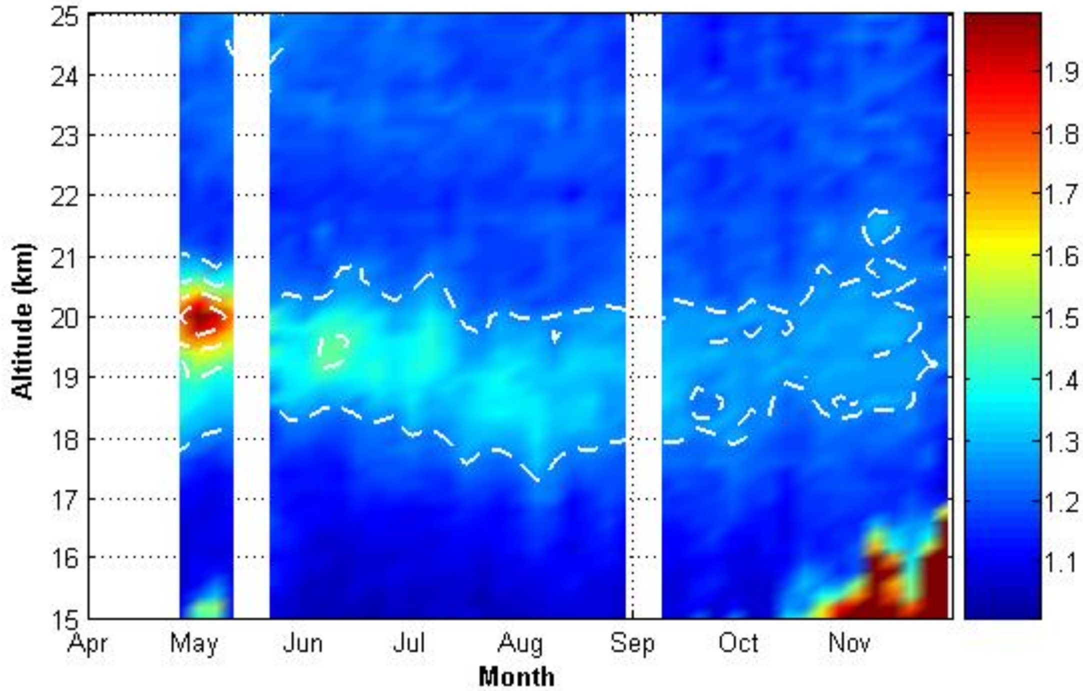


Figure 8 : Time series of weekly-averaged profiles of scattering ratio at 532 nm obtained from ground-based (a) and space-borne (CALIOP, b) LiDAR observations over Reunion Island between April 2015 and December 2015. The scattering ratios from CALIOP have been averaged within ± 5° in latitude and ± 50° in longitude (extending from Africa to Australia) around Reunion Island.

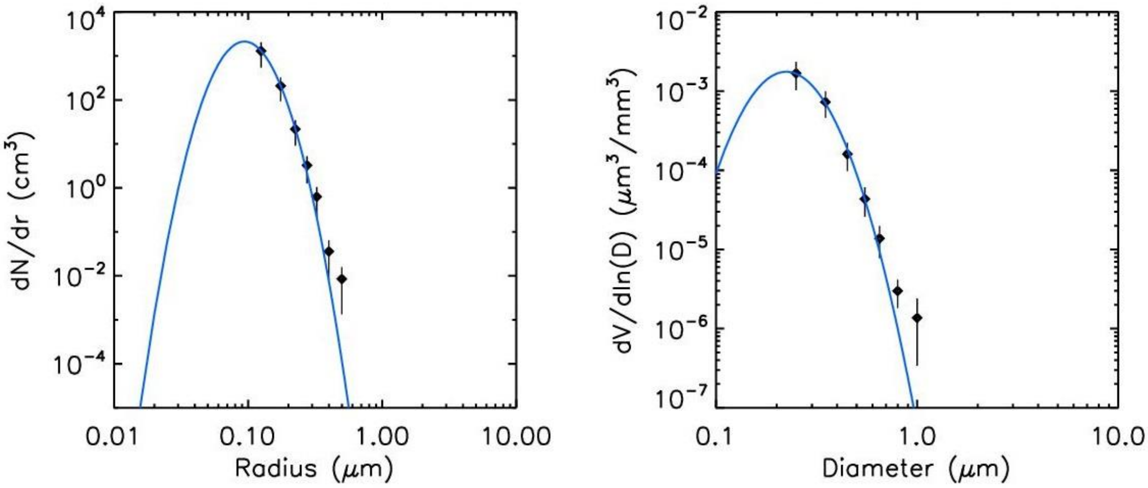


Figure 9 : Number (dN/dln(D)) and Volume concentration (dV/dln(D)) obtained from LOAC OPC observations on 19 May 2015 at 1746 UTC over the Reunion site.

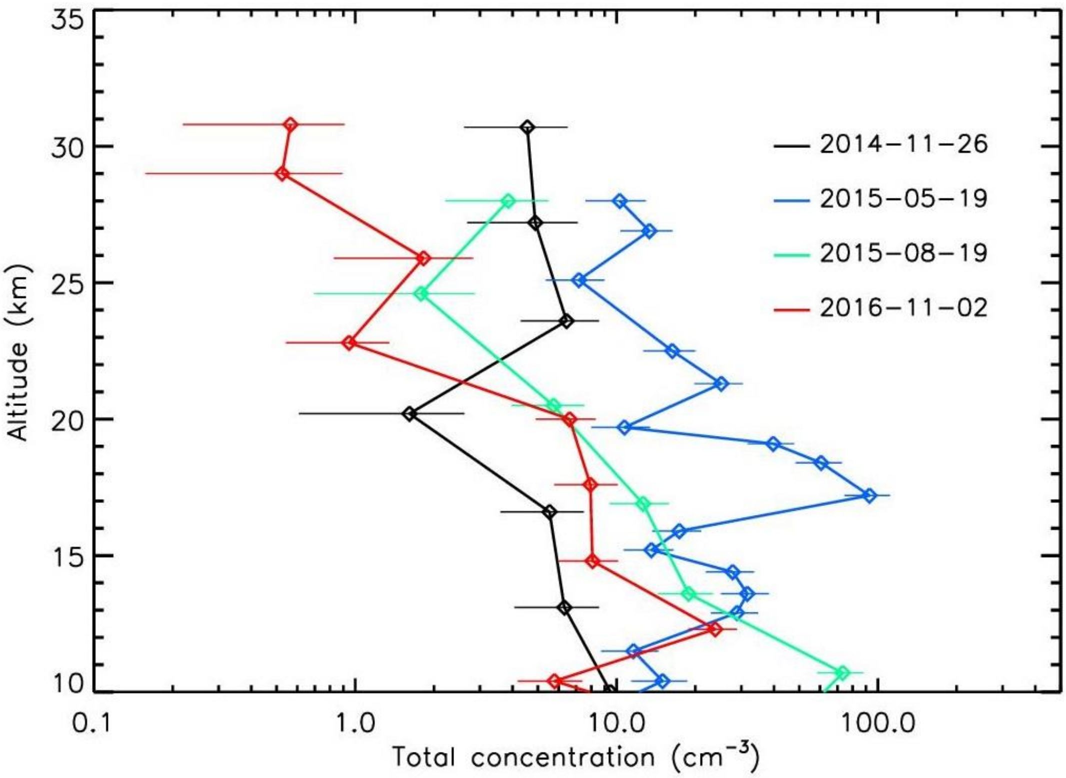
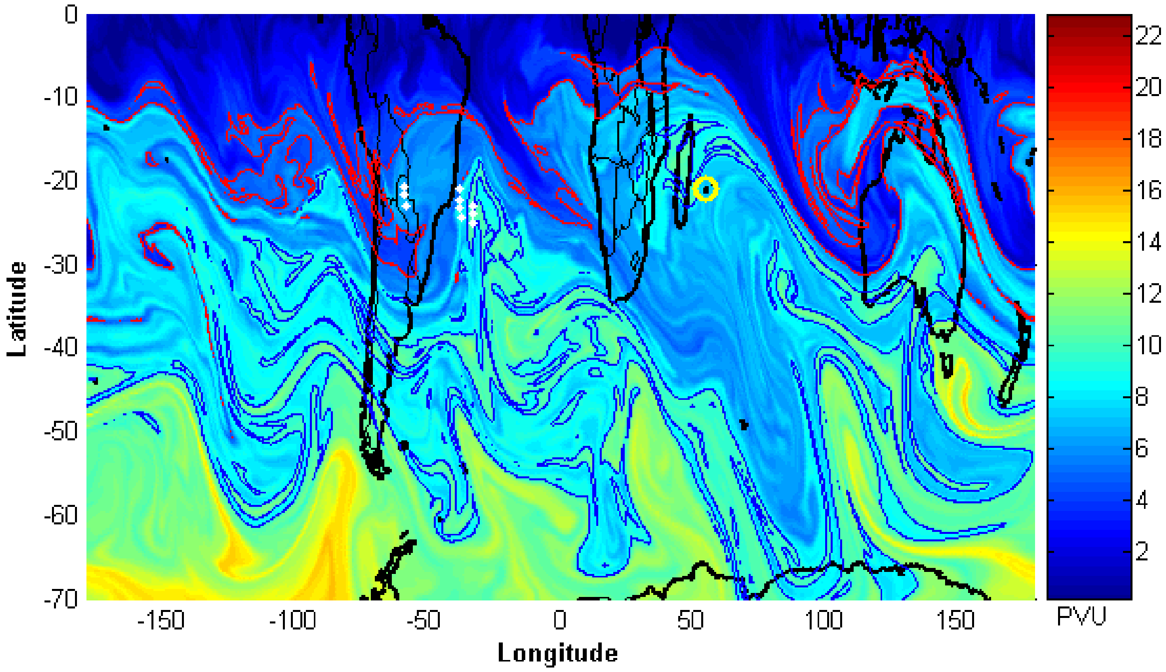


Figure 10 : Total number concentration of aerosols (0.2-50µm) profiles obtained from LOAC OPC observations over Reunion during the 26 November 2014 (black line), the 19 May 2015 (blue line), the 19 August 2015 (green line) and the 2 November 2016 (red line). The aerosols layer is delimited by two horizontal black lines.

a)



b)

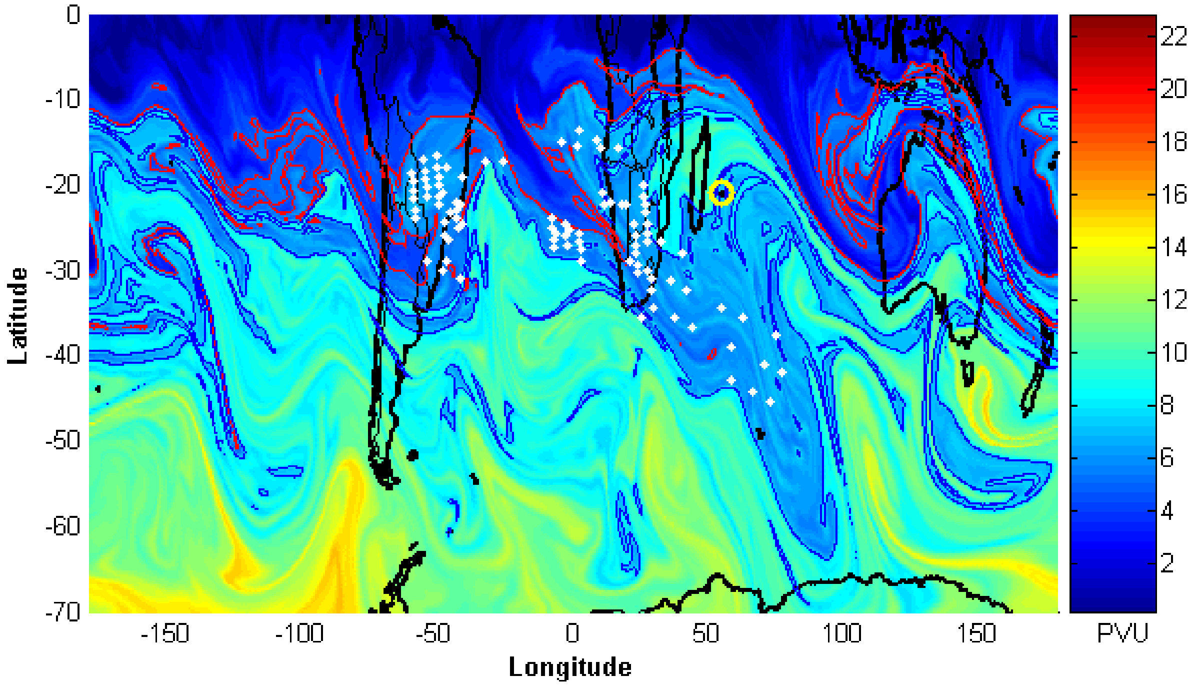
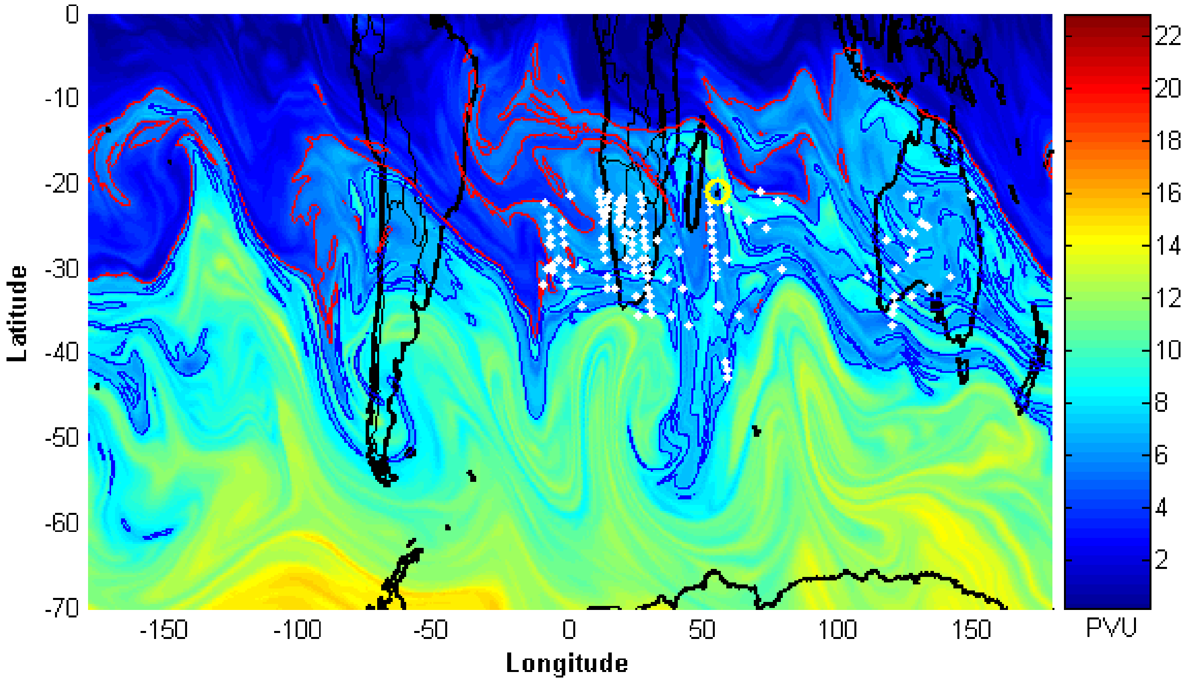


Figure 11 : Advected PV map at the 400 K level obtained from the MIMOSA model (a) on 27 April 2015 and (b) on 01 May 2015. The positions of the subtropical barrier (red line) and a south dynamical barrier (blue line) are detected from the DyBAL code. The white dots represent the localization of the aerosol plume at 400 K ± 5 K obtained from OMPS observations, while the yellow circles indicate the Reunion site.

a)



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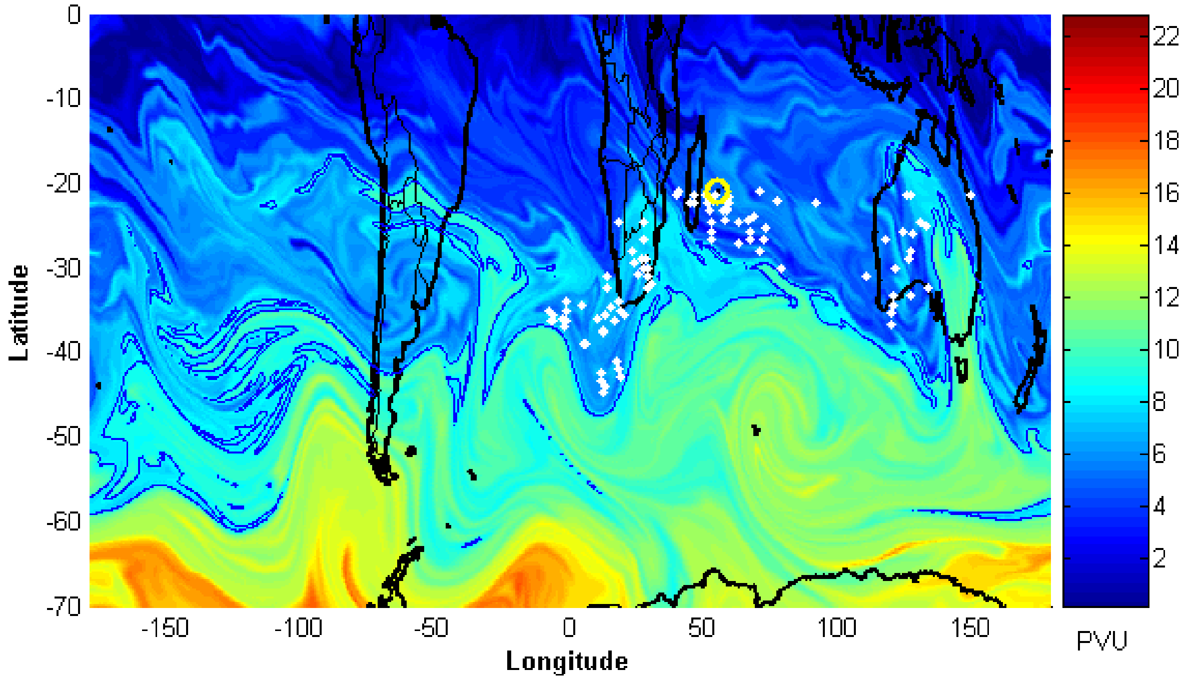

Figure 12: Same as Figure 11 but for (a) 19 May 2015 and (b) 03 June 2015.