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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-range isentropic transport of stratospheric aerosols over Southern Hemisphere following the Calbuco eruption in April 2015" *by* Nelson Bègue et al.

Anonymous Referee #3

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Review of "Long-range isentropic transport of stratospheric aerosols over Southern Hemisphere following the Calbuco eruption in April 2015" by Nelson Begue, Damien Vignelles, Gwenael Berthet et al.,

This manuscript prsents an interesting analysis to assess the long-range transport of the volcanic aerosol plume from the April 2015 Calbuco eruption (Chile). The authors bring together ground-based lidar and balloon-borne LOAC particle measurements from a field campaign from Maido (Reunion Island) also with several satellite datasets through the period, combining analysis of the SO2 from IASI also with the vertical aerosol extinction profile observed from OMPS and CALIOP.

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Discussion paper

The topic of the paper is certainly suitable for publication in Atmospheric Chemistry and Physics, and the synthesis of observational datasets, together also with analysis of the plume's detection alongside isentropic air-mass transport through the post-eruption period, will represent a valuable contribution to the journal.

The written style of the paper is generally very good, and my opinion is that mostly only minor revisions are required before the paper can proceed to publication. There are however two principal revisions required which would likely be considered more substantial than minor, both of which relate to two particular terminologies used in the paper.

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.

The other principle point which might potentially be considered (by some) to be more than minor, is in relation to the phrase "unimodal", which is stated within the discussion of the size distribution observed by the Optical Particle Counter (OPC) on the LOAC

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balloon soundings. 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 $\sim$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.

The length of my comment above should not be misunderstood as being critical, I am just trying to explain the rationale behind my recommendation that the sentences with the terms "isentropic" and "unimodal" are reworded accordingly. It is perfectly understandable that the authors phrased the sentences like this, but I assert that current

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knowledge now suggests that is often not the case at all.

The paper presents an excellent analysis of observations and the list of suggested minor comments below are intended to improve the manuscript before it proceed to publications in the main Atmospheric Chemistry and Physics journal.

Suggested minor revisions ————————-

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

2) Abstract, page 1, line 25: Please replace "1" with "one".

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".

4) 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?

5) 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?

6) 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?

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7) 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.

8) 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".

9) 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".

10) 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)"

11) 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?

12) 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?

13) Introduction, page 3 lines 33 and page 4 lines 1-2: The previous sentence dis-

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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.

14) 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 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."

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

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

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

18) 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.

19) page 6, line 12: The authors have given uncertainty estimates for each size bin, but

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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.

20) 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.

21) page 6, line 32: replace "looking" with "view".

22) 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".

23) 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.

24) 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.

25) 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".

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

27) 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

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in comment 22) could also add a sentence explaining these 2-week near-global cross-section composites?

28) 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?

29) 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.

30) page 10, line 10 – "norther than" –> "north of".

31) 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.

32) 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)?

33) 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?

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

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

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36) 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?

37) 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.

38) 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?

39) 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.

40) 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.

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

42) 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.

43) 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".

44) page 12, line 23: Delete "using" and put "or may also be due to" (or reword re:

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comment 43 above).

45) 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?

46) page 12, lines 25-27 – This sentence is unclear – please re-write.

47) page 12, lines 29 – Replace "DV" with "dV".

48) 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.

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

50) 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.

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

52) 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.

53) page 14 lines 20-21 – "cannot move beyond the south of Brazil" – suggest to re-word this – is it just that the trajectory for the airmasses takes the plume this way – I

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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?

54) 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"

55) page 15 line 17 – replace "stratosphere into the middle and high latitude" with "stratosphere into the troposphere".

56) 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."

57) 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.".

58) page 16 lines 25 – insert comma before "possibly".

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

60) 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?

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

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(km)"

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

63) 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".

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

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