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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Measurement Report: Lidar measurements of stratospheric aerosol following the Raikoke and Ulawun volcanic eruptions" *by* Geraint Vaughan et al.

**Anonymous Referee #2**

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This manuscript presents an analysis of ground-based lidar measurements of the volcanic aerosol cloud from the June 2019 Raikoke eruption, made with a Raman lidar sited at the Capel Dewi atmospheric observatory, near Aberystwyth, Wales, U.K.

The study presents a limited set of data from the Capel Dewi lidar, with initial measurements on 1st and 3rd July 2019 showing clear layers of enhanced aerosol at 12km and 14km, and then layers detected from later lidar soundings at 14-15km on 13th/14th July and then at 20-21km on 25th August.

To interpret the layers detected from Capel Dewi, the authors also analyse spaceborne lidar measurements from the CALIPSO satellite, analysing profile-transects of



backscatter and depolarisation measured by the CALIOP lidar during orbits over the UK on 14th July, with also earlier the depolarisation within CALIOP lidar transects over the UK in earlier CALIPSO orbits.

The CALIOP analysis reveals the nature of the aerosol layers detected from the ground-based Raman lidar, with the 14th July measurements showing less than 1\% depolarisation, indicating spherical particles – then identified as aqueous sulphuric acid aerosol particles typical of ash-free portions of volcanic plumes. In contrast, the early July CALIOP profiles (not shown in the paper) show 10-20\% depolarisation in the lower 12km layer, the higher layer non-depolarising.

The authors present leave open the attribution to be either volcanic ash-sulphate mix from Raikoke or biomass smoke aerosol from the strong wildfire pyroconvection in Canada during June/July, which is also known to have also injected non-spherical particles into the stratosphere.

The authors then derive aerosol extinction from the aerosol backscatter assigning values for the extinction-to-backscatter-ratio (often referred to simply as "lidar ratio") of 40-50 steradians for the volcanic aerosol and 100 steradians for the biomass smoke.

The authors have chosen to present their results in the form of a measurement report, which then allows results and conclusions to be more limited in scope than would be required for a full ACP article:

https://www.atmospheric-chemistry-and-physics.net/about/manuscript_types.html

I am not sure whether the authors are aware, but there is currently a joint ACP/AMT/GMD Special Issue on "Satellite measurements, in-situ measurements and model simulations of the 2019 Raikoke eruption". Although the title of the Special Issue does not state that the non-satellite remote sensing observations such as those presented here are within scope, it would obviously make sense to include this manuscript within that special issue, and I am sure the ACP editors would welcome its inclusion.

In my comments, I am recommending the handling Topical Editor invite the authors to consider the manuscript can be re-aligned with the Raikoke Special Issue (assuming the ACP editors for the Raikoke SI agree, which I imagine they will) and request this manuscript and review to be included as an additional manuscript for the special issue.

In the spirit of that special issue, the reporting of the ground-based lidar measurements will represent a useful addition to the Special Issue. In particular, the link between the ground-based and space-borne lidar measurements represents an interesting analysis, potentially identifying an important difference between the later non-depolarising cloud and the earlier non-spherical particle layers detected in the early July ground-based lidar soundings.

The manuscript is mostly quite well written, and certainly in scope of the Measurement Report type ACP article the authors have chosen. I would recommend however that the authors consider potentially adding Figures showing the early July CALIOP layers, and also those to interpret the later August volcanic aerosol layer observed over Wales at the higher altitude of 20km.

The manuscript as currently presented also came across as a little disjointed, with the analysis of the progression of the cloud in the initial days after the eruption (with the HYSPLIT analysis in Figures 1 and 2) and seemed quite separate from the main topic of the article to analyse the layers observed from Aberystwyth ground-based lidar.

That said, I can see that the authors chose to include that to set the scene for the later analysis. Another of my comments refers the authors to another manuscript submitted to the Raikoke special issue (de Leeuw et al., 2020) which analyses simulations of the Raikoke cloud carried out with the NAME dispersion model.

It is interesting to contrast the initial progression of the cloud predicted by HYSPLIT with that seen in the NAME dispersion model, and there is a directly comparable Figure in the de Leeuw et al. (2020) manuscript for that 25th June date shown in Figure 1. It's clear that the NAME volcanic simulations (perhaps not surprisingly) predict much



greater dispersion of the cloud, with TROPOMI measurements of the SO2 confirming there was a lobe of the Raikoke SO2 cloud dispersed to the North West of the cyclonic recirculation, stretching already to the West of the original eruption location (153W).

Adding the extra CALIOP Figures, and some additional discussion re: differences between the HYSPLIT predicted dispersion and that seen in the TROPOMI observations (and represented in the NAME simulations from the de Leeuw et al. manuscript) would (in my opinion) enable the manuscript to be re-submitted as a full journal article to the Raikoke Special Issue.

However if the authors for some reason wish to keep the manuscript separate from the SI, or wish to re-allocate their submission to the SI retaining this Measurement Report type of manuscript that is also fine.

In my review below, I have listed a set of minor revisions that will improve the manuscript, and, once attended to, will make it publishable then in the SI as a Measurement Report.

These include being much clearer about the wavelength for the stratospheric AOD (sAOD) values derived from the ground-based lidar. Within my minor revisions below, I'm requesting to add a sentence referring to the OMPS and SAGE-III measuremnets of the Raikoke cloud shown in the Kloss et al. (2020) paper within the Raikoke special issue. The Figure there shows the sAOD from the 670nm SAGE-III channel to compare to that derived from OMPS (at 675nm), whereas the optical depth shown in Figure 6 is for the 355nm Raman channel. Since the authors may choose to retain their manuscript as this limited "Measurement Report" I am not requiring to compare directly to these other measurements, and the difference in wavelength makes that requiring to assume some Angstrom exponent for the wavelength translation, which itself is likely uncertain. The main comment here is to make sure the wavelength shown is communicated within the Figure captions so that the reader can always bear in mind the wavelength for the sAOD values being shown.

The authors also seem not be aware that recently the CALIOP team have developed a new stratospheric aerosol typing algorithm, which is described in Kim et al. (2018) and has a formalised categorisation into 4 types of stratospheric aerosol – polar stratospheric aerosol, volcanic ash, sulfate/other and smoke. This may help to formalise the choice of lidar ratio in the comparisons presented.

I've listed 20 specific revisions below, which although mostly minor in nature, do together comprise then major corrections. However, the authors can probably make those changes quite quickly, when I suggest they then re-submit to the Raikoke Special Issue, either retaining as a Measurement Report paper, or carrying out additional comparisons to other measurements such as TropOMI SO2 (Figure 1) and SAGE-III/OMPS sAOD measurements in Figure 6 – which would then elevate the status of the paper to have sufficient results to then be a "full ACP manuscript". The authors could also consider contacting Sergey Khaykin from Hautes Provence (lidar measurements shown in the SSiRC workshop's Raikoke page) and include some comparison between the timing of the plume detected over Wales and over Southern France.

Minor revisions ————

1) Abstract line 1 – this 1st sentence should communicate more about the magnitude of the Raikoke eruption. The paper by Firstov et al. (2020) presents measurements of the infrasound wave signals from the eruptions from monitoring stations on the Kamchatka peninsula, which show the eruption had a volcanic explosivity index (VEI) of 4, i.e. it was a VEI4 eruption.

The Newhall and Self (1982) paper which presented the formalised basis for VEI estimation term VEI4 eruptions as "large magnitude eruptions" in contrast to eruptions at a VEI of 5 or larger being "very large eruptions". I recommend then the authors include to refer to the June 2019 Raikoke eruption as a "large magnitude eruption", consistent with the infrasound measurements from Firstov et al. (2020). The measurements show that there were 11 individual eruptions, with the 9th of these (from 23:00 on the 21st



June to 02:00 on the 22nd) the most explosive phase. I therefore suggest also to add "3-hour duration" to this initial sentence.

I suggest then to replace "erupted" with "began a 3-hour duration large magnitude explosive eruption", and change "On 22 June 2019" instead to "At 23 UTC on the 21st June 2019".

2) Page 1, lines 2-3 – Suggest to replace "has been used to measure" with "was deployed to measure", insert "vertical" before "extent" and either insert "enhancement to the" before "stratospheric aerosol layer" or replace with "volcanic aerosol cloud".

3) Page 1, line 5 – Replace "corrected for aerosol extinction" instead to "translated to aerosol extinction". It's not appropriate to refer to that as a correction – need to be clear that it's a conversion or translation of the measured backscatter into an aerosol extinction, based on an assumed lidar ratio (extinction to backscatter ratio), as is explained in the main part of the article. I would argue it is perhaps accurate even to refer to a "retrieval" of the aerosol extinction from the lidar's measurement of the backscatter. But suggest simply to replace "corrected for" to "translated to" or "converted to".

4) Page 1, line 5 – Change "comparison with aerosol-free profiles" instead to "subtracting the molecular backscatter profile (i.e. that from the gas phase)". The radiosondes enable to calculate the molecular backscatter, which then enables to isolate the aerosol backscatter signal from the total backscatter measured by the lidar.

5) Page 1, lines 6-8 – This sentence about the biomass burning smoke from the wildfires in Canada needs to be clearer the dates on which the Capel Dewi lidar observations are suggested to be wildfire smoke. The sentence after that states that the 14km layer in the 3rd July profile (Figure 3b) may have been volcanic ash. And the text in the main article states those layers in the 1st July and 3rd July lidar soundings (Figures 3a and 3b) could be caused either by the smoke aerosol from the Canadian wildfires or by volcanic ash from Raikoke. The text refers to CALIOP measurements from 2nd July and 4th July, but these are not shown in the manuscript, so it's not possible for

me as a reader to be able to make a judgment on which attribution is more likely. As I mentioned in my general comments above, I strongly recommend the authors add an extra Figure to show these CALIOP measurements – similar to Figure 5, but here perhaps it is possible to include these within Figure 3 as additional panels. This will really help the article to be able to understand the potential alternative attribution for these layers as either biomass smoke or volcanic ash (which would have within that air mass externally and internally-mixed volcanic sulphuric acid also.

Please clarify this in the Abstract text, being clear that the depolarisation from the CALIOP profile transects over the UK are being used to attribute the aerosol type measured from Capel Dewi. And please also re-draw Figure 3 adding the CALIOP transects from 2nd July and 4th July, and sharpen up the associated text discussing these early soundings.

6) Page 1, lines 8-9 – the language here is a little too informal here where it says "reached around 0.05 by early August" – the text needs to be precise in the Abstract here. Suggest to re-word the sentence with some additional context such as "A sustained period of clearly enhanced stratospheric Aerosol Optical Depth (sAOD) began in early August, with maximum sAOD at around 0.05 in mid-August, remaining above 0.02 until early November (around Julian day 310)."

7) Page 1, lines 9-10 – Replace "location of peak backscatter" with "altitude of peak backscatter" and insert "(between 14 and 18km)" after "considerably" to clearly summarise the variation seen in Figure 6.

8) Page 1, line 21 – Add a sentence here (after the text "previous 2 months.") referring to the OMPS and SAGE-III satellite measurements presented by Kloss et al. (2020), and re: the maximum values of the 675nm stratospheric AOD shown in the Figure 7 of that paper.

9) Page 2, line 25 – give some indication of the frequency at which the lidar measurements were made from Capel Dewi, and how many nights in each month of this period



the lidar soundings were made.

10) Page 2, line 41 – change "simulate an aerosol-free lidar profile" to "construct a molecular backscatter profile (for the gas phase)"

11) Page 2, line 42 – the authors refer to an "onion peeling algorithm" which I was not aware was being used in the retrieval of the backscatter ratio. My understanding is that once the molecular backscatter profile was determined, the aerosol backscatter was then simply determined from subtracting that molecular backscatter from the total backscatter measured. I am aware that one can account for the attenuation during the two-way transmission through the atmosphere, i.e. convert the "attenuated backscatter" to a "clean" aerosol backscatter (e.g. as explained in Young et al., 2015; and Antuna Marrero et al., 2020) which then accounts for the attenuation from aerosol and gas phase species in the two-way transmittance. Please add a sentence here to explain what is meant by the "onion-peeling algorithm.

12) Page 2, line 47 – The authors have stated "the choice of lidar ratio is to some extent arbitrary, since a wide range of values are given in the literature for aerosols of volcanic origin". But it is not correct that the choice is arbitrary, and also it does not follow from the remainder of that sentence. The choice of lidar ratio clearly affects the value of the aerosol extinction derived from the measured backscatter – and just because the values are variable or uncertain does not mean that the choice is necessarily arbitrary. I was very surprised at this sentence given that the lead author's 1994 GRL paper shows a very interesting Figure illustrating how the lidar ratio varies for a log-normal size distribution of sulphuric acid aerosol particles as one varies the geometric mean and geometric standard deviation for the mode (see Figure 6 of Vaughan et al., 1994). That Figure demonstrates how the size distribution alone can explain why the lidar ratio is variable between those values of 40 and 50. The presence of ash either externally mixed and/or internally mixed with the sulphuric acid aerosol will further affect the variation of the lidar ratio within a volcanic aerosol cloud. Please change that sentence from "arbitrary" to explaining "For volcanic aerosol clouds from very large magnitude

eruptions (e.g. 1991 Pinatubo) the lidar ratio for the cloud has been shown to be highly variable, for example due to variations in the size distribution of the sulphuric acid aerosol (e.g. Vaughan et al. 1994)". That sentence can follow after the sentence ending "volcanic origin.", replacing "is to some extent arbitrary" with "is known to be highly variable" and deleting "choice of" and changing "since a wide range..." to "with a wide range...". Then the sentence "For example, Mattis et al. (2010)..." can be revised to "For moderate magnitude eruptions, Mattis et al. (2010)..." so that it follows on from the sentence about very large magnitude eruptions.

13) Page 3, line 54 – The authors have stated "when the volcanic aerosol was bounded from above and below", and I think this must be referring to the method for calculating vertically integrated properties such as the stratospheric AOD. But it's far from clear what is meant there. Please re-word to clarify this.

14) Page 3, line 55 – the authors have written "an appropriate value of lidar ratio could be found by requiring that the backscatter return to the molecular profile below the layer." But that sentence does not make sense at all. Please revise the wording to explain what is meant.

15) Page 3, line 64 – the authors have stated "the ash and sulphur dioxide plume initially moved westward" – but I think the authors must mean "eastwards". Perhaps they are confusing this to "westerly" (i.e. from the west). Best to use eastwards though as that's less confusing to non-meteorological readers.

16) Page 3 line 66 – change "figure 1" to "Figure 1".

17) Page 3 line 67 – after "Kamchatka and Alaska." add a sentence explaining more about the HYSPLIT model. It needs to be communicated to the reader that this is not really a volcanic aerosol model – but just an isentropic air mass trajectory model. It should be made clear to the reader that the trajectories are indicative only, and not expected to be so accurate after several days. This is an interesting case that provides an opportunity to compare the volcanic plume dispersion shown in Figure 1 with equiv-



alent simulations from the NAME dispersion model (used operationally for the London VAAC predictions) shown in Figure 6 of the manuscript by de Leeuw et al. (2020) submitted to the Raikoke special issue, currently in review for publication in ACP.

It is clear that the NAME dispersion model predicts quite different dispersion of the SO2, with a lobe of the volcanic cloud is transported to the west, which is not seen in the HYSPLIT predicted transport of the plume, the NAME predictions in good agreement with the TropOMI satellite measurements of SO2.

Please add a sentence acknowledging the simpler nature of the HYSPLIT predictions, and referring to this finding from the NAME dispersion model simulations that actual simulate the transport of the SO2 and ash, with vertical profile of emission reflecting an emission vertical profile from the VolRes community (see de Leeuw et al., 2020). And refer to the TropOMI observations showing the SO2 was dispersed also to the west of the Raikoke volcano as shown in Figure 6 of the de Leeuw et al. (2020) paper.

18) Page 4, lines 77-80 – The authors explain that in addition to the Raikoke volcanic aerosol cloud's dispersion, there is also biomass burning smoke from wildfires in Canada, which complicated attribution of the layers detected from Capel Dewi. The authors may not be aware however, that recently the CALIOP team have developed a new stratospheric aerosol typing algorithm, which is described in Kim et al. (2018) and has a formalised categorisation into 4 types of stratospheric aerosol – polar stratospheric aerosol, volcanic ash, sulfate/other and smoke. This may help to formalise the categorisation being explored here. Please add a sentence referring to this stratospheric aerosol type algorithm presented in Kim et al. (2018).

19) Page 5, line 97 – replace "lay" with "was detected".

20) Page 10, Figure 6 – Please redraw this Figure changing the symbols to be error bars showing the range in sAOD values derived for lidar ratio of 40 and 50. The two limit values will be relatively close together but in that sense represent a range based on assuming the layers are composed of volcanic aerosol. Then there should be two

values – one showing a range between the 40 and 50 lidar ratio derived sAOD (i.e. assuming volcanic) with the earlier points also having additional symbol for the 100 lidar ratio derived sAOD value (i.e. assuming biomass smoke).

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