We would like to thank the reviewer for his helpful comments on our manuscript. In general, we respond to general comments in the preamble in the body of the review where specific comments are made. All changes in the manuscript (for all reviewers) are underlined in the new manuscript. Where things may not be obvious: New references in the text have also been added to the reference list. Also, there is a new frame in Figure 2 (a) that shows the 525 and 1020 nm aerosol extinction efficiency for sulfuric acid aerosol. Figure 11 has two added frames that depict aerosol extinction coefficient at 1020 nm for the Ambae and Nevado del Ruiz-like eruptions. There are small changes to Figures 1, 4, 5, 6, 7, and 10. We have added Julian day of eruption to Table 2 for clarity relative to the associate figures.

Interactive comment on "Evidence for the predictability of changes in the stratospheric aerosol size following volcanic eruptions of diverse magnitudes using space-based instruments" *by* Larry W. Thomason et al.

Pasquale Sellitto (Referee) pasquale.sellitto@lisa.u-pec.fr Received and published: 3 July 2020

The manuscript ''Evidence for the predictability of changes in the stratospheric aerosol size following volcanic eruptions of diverse magnitudes using space-based instru- ments, Thomason et al." discusses the relationship of multiwavelength aerosol extinc- tion observations, in the first phases of volcanic plumes dispersion following small-to- strong stratospheric eruption, and the apparent size of the particles in the plume. From my perspective, the main result of this work is the evidence of a clear relationship of the extinction ratio (shorter-to-longer wavelength, namely 525/1020 nm, hereafter referred to as ER) and the strength of the eruption in terms of the aerosol extinction at 1020 nm (hereafter referred to as AE), see Fig. 8b. This relationship is associated to the apparent particles size in the aerosol layer (Fig. 2). Even if this behaviour is probably expected, I personally think that a systematic study of this relationship is very interest- ing and important. The identification of volcanic plumes by means of the concurrent increases of the aerosol extinction (or the integrated optical depth) and modifications of the spectral variability of this extinction (or the Ångström exponent) has been exploited, in the past, in different studies – and it is a tool that I personally use a lot. Nevertheless, a systematic effort to: a) study this aspect over a long observation series, or even construct a theoretical basis, has never been attempted, to my knowledge. Thus, I think that such kind of work would deserve immediate attention and rapid publication. Unfortunately, the present manuscript, while having all elements to provide the com- munity with both points a and b mentioned above, is somewhat flawed in two aspects, that I mention in the following. I encourage the Authors to tackle these two "major" issues, as well as a number of specific issues that I also list in the following, and I'll be happy to re-evaluate the manuscript revised accordingly, once the due modifications are done.

Sincerely, Pasquale Sellitto

Major comments:

1) The conceptual model defined to connect the apparent radius of the aerosol layer and AE/ER, as defined at L271-273 and shown in Figs. 2 and 11, is not completely clear to me. The model is defined for what looks like a monodispersed aerosol layer (which is also mentioned at L102), which is fine when discussing this in theory (Fig. 2) but is a little bit odd when applying to real data (Fig. 11). In the real world, the size distribution would not be monodispersed, so I guess a more realistic size distribution should be used, which is still not very complicated using a mono-modal size distribution with varying mean radii. Another thing that gets me confused is that in the equation at L271, it looks to me that the numerator of the ratio on the right is not the perturbation delta_k but rather k (otherwise, you don't have number density n(r) but the perturbation in number density delta_n(r)). Like this, it looks like you're accounting twice for the background, when you later scale the result with respect to the background. And also, the Mie scattering efficiency is calculated with a Mie code (which one?) and is based on an assumption of the particles' composition (their refractive index): which is your assumption? If, as I imagine, the Authors have supposed a pure sulphates plume, why the ash has been neglected – it may be important in the first phases of some eruptions, e.g. Raikoke and Kelud? And again, if it is sulphates, what has been supposed for the mixing ratio of sulphuric acid in the sulphate aerosols droplets, which is a factor that can modulate the extinction of the particles, at least at longer wavelengths? By using the Mie scattering coefficient, you suppose that the absorption can be completely neglected. If ash is to be considered, this might not be completely true. All these aspects have to be clarified or maybe the model has to be slightly refined to account for the mentioned problems.

We did not clearly explain the purpose of the model discussed in section 4. We are not suggesting that the simple model used there is completely realistic as it only accounts for sulfuric acid aerosol and models the perturbation as a single particle size rather than a distribution of some kind. For the latter limitation, we have limited or no information on the changes to the aerosol size distribution for these events at the latitudes and times of the space-based observations. The model is an attempt to justify our interpretation of the observations shown in figures 7 and 8 but not to exactly model any event.

The n(r) parameter is for the perturbation and this has now been noted in the text. We are indeed modelling only sulfate aerosol for which there is no absorption at the wavelengths consider in this paper (and scattering and extinction cross sections are the same). We updated the text to indicate that we are, in fact, using the extinction cross sections and the Mie code is based on Bohren and Huffman (1998).

The manuscript structure has to be improved. In the present version, the main re- sults and the overarching narrative are not completely clear. First, the Introduction fails to present the motivations of this work. Some elements of motivation are in Sect. 2 rather than in the Introduction. It is stated two times that "The primary goal of this effort was to assess data quality of data sets consisting of a single wavelength mea- surement of aerosol extinction coefficient or similar parameter particularly when a fixed aerosol size distribution is a part of the retrieval process." but I cannot see where this is discussed in the text. As stated in my introduction, I would rather say that the main motivation for this study (and it is an important motivation!) is to develop a means to identify volcanic plumes and to classify them based on the eruption strength, and I suggest mentioning this as a motivation. Section 2 does not present satisfactorily the SAGEII-III datasets and a lot of information is lacking. Care should be taken, through- out the manuscript, to introduce the Figures sequentially in the text and not to discuss them before presenting their content.

We have improved the introduction particularly in providing better motivation for the study contained in the remainder of the paper. Figures are now discussed in order of their number.

I add some specific points in the following Specific Comments.
Specific comments:
1) I don't understand the sense of the word "predictability" in the title, as you attempt no predictions

Clarified later in the text as referring to the ability to predict the impact on OSIRIS observations in the absence of SAGE-like observations.

2) L15-16: there is a lacking mention to the ER, which is the measured parameter that actually is studied and vary for different eruptions strengths (and AE)

Added

3) L16: "The relationship is measurement-based and does not rely on assump- tions about the aerosol size distribution.": strictly speaking there is an assumption of monodispersed aerosol layer

The primary results are those found in Figure 8 showing how the aerosol extinction coefficient perturbation ratio varies with aerosol extinction coefficient. These results are entirely measurement-based. The model is only used to explain (as is clarified in the text) to explain why we believe what we observe in the measurements (at least for the 8 events that follow the main curve) is consistent with homogeneously nucleation of many small aerosol.

4) L18-22: "Despite this limitation. . .particle size": these two points (the use in eval- uating global models and the improvement of mono-spectral AE observations) is not discussed in depth in the manuscript so it is strange that it is mentioned in the Abstract

We've improved this aspect of the discussion in section 4.

5) As stated at Major Comment #2, I feel that the Introduction is very synthetic and does not make a good job in motivating this work

The introduction has been expanded and better reflects the goals of this paper.

6) L24: "Eruptions of volcanoes" –> "Volcanic eruptions"

Done

7) L25: “Volcanically-derived aerosol”: Here the you talk in general of volcanically derived aerosol and, later in the text, the discussion specialises on sulphates. A line is probably lacking here on the mention of the possible variety of volcanic particles (ash and sulphates)

Done

8) L29: I think that Tang et al. 2013 do not discuss of the impact of volcanic aerosol on transport but of the decrease of ozone in the stratosphere and then, consequently, in the troposphere by stratosphere-troposphere exchanges. As one lines above you discuss about the radiative heating of volcanic aerosols, one can erroneously think that Tang et al. talk about radiative-dynamical interactions and the crossing of tropopause by plume self-lifting, which is not the case. Please correct

Changed the reference to:
Pitari, G., Cionni, I., Di Genova, G., Visioni, D., Gandolfi, I., and Mancini, E.: Impact of Stratospheric Volcanic Aerosols on Age-of-Air and Transport of Long-Lived Species, Atmosphere-Basel, 7, 149, ARTN 149

9) L31: “. . .either the measurements. . .”: please develop a bit to clarify how measure- ments have been used in global models to derive climate impact of volcanic eruptions

Clarified this statement

10) Why not more classic section titles: "2. Data and Methods" and "3. Results"?

Fine.

11) L45: “well-known SAGEII”: it is well-known but it might be less known for a part of the readership of ACP. Thus, please suppress “well-known” and mention the years and months of operations of SAGEII, from launch to end of mission.

Removed.

12) In Tab. 1 there are more eruptions than what studied in Sect. 3 and 4 and there are fires as well. While later in the text it is said that fires are not in the scopes of this work (while it would have been interesting to see where the points in Fig 8b locate for Canadian and Australian fires. . .), it is not clear to me why many eruptions possibly present in the datasets have not been included in this study: Sarychev, Kasatochi and Nabro are largely considered “major moderate eruptions” for their impact on the stratosphere but are neglected in this study.

The fire events tend to align more with Raikoke than with what we interpret as sulfuric acid dominant eruptions. We felt including them distracted from the main goals of the paper since there is little reason to expect them to behave the same as a volcanic eruption. Some events in Table 1 occur during periods where SAGE measurements do not exist and thus are not included in the analysis.

13) L55: Please briefly introduce this GloSSAC dataset. Also please mention in the corresponding reference that the relative manuscript is presently under discussion/review and add a link for the discussion paper

Added a bit of material here. The paper has been accepted for publication. The reference was updated to the discussion but we plan to update to the final reference before this paper is complete.

14) L59: “SAGEII record”: here is very clear that mentioning the start-end of SAGEII operations is important

Updated to include month..

15) L61: “*. . .*subtly modulate climate*. . .*”: probably it can be mentioned that the ag- gregated impact of these "small-to-moderate" eruptions is significant (see also Ri- dley, D. A., 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)

Added

16) L63: as for SAGEII, the precise period of operations of SAGEIII/ISS should be clearly written

The mission is on-going so data is available from June 2017 through the present.

17) Fig. 1: the periods of operations (start/end of missions) of SAGEII and III should be probably indicated in this figure, e.g. with vertical dotted lines. That would help a lot in the understanding of the different discussions of Sect. 2

Added

18) L67: “Raikoke*. . .*2020): why not arranging these eruptions in chronological order?

Reordered.

19) L69: in the following paper about Canadian fires 2017, a similar method as the one discussed in this manuscript is used to

identify and separate a fire plume from an anthropogenic plume (see their Fig. 2b): Kloss et al. Transport of the 2017 Canadian wildfire plume to the tropics via the Asian monsoon circulation, Atmos. Chem. Phys., 19, 13547–13567, https://doi.org/10.5194/acp-19-13547-2019, 2019.

Added.

20) L69: please note this pre-print manuscript on the Australian fires 2019-20: *https://arxiv.org/abs/2006.07284*

To our understanding, this is not considerable referenceable by Copernicus. However, we have added a similar reference.

21) L71-72: ". . .a qualitative difference. . .": please mention a difference with respect to what? (That would be clearer if the periods of operations of SAGEII and III are indicated in Fig. 1)

We've rewritten this section to make the qualitative difference clearer.

22) L73-80: These motivations should be moved to the Introduction

This discussion is now in the introduction

23) L81-93: also more appropriate in the Introduction?

We think this discussion needs to be located here since it deals with issues related to the instrument and the analysis.

24) L95: what do you mean with "robust"?

Updated to 'high accuracy and precision'

25) Are footnotes allowed in ACP format?

Removed

26) L101-102: see Major Comment #2

Mie calculations are based on the code provided in Bohren and Huffman (1998). We've also changed the text to indicate that we are discussing single particles (at this stage) not a size distribution.

27) L124-125: “These begin. . .Table 2)”: Is this line to be moved earlier (e.g. L120)?

Moved.

28) L129: “. . .O4 absorption. . .version.”: Is there a reference for this underestimation? And also, a few words should be included to clarify why using the interpolation between 448 and 756 nm limits (or avoids) this underestimation: O4 has no absorption at these wavelengths and has a significant absorption at 521 nm?

The O4 error has a subtle (positive) impact on the ozone retrieval below 20 km where there is significant overlap in the spectral regions used to retrieve ozone and where O4 absorbs. The small error in ozone has a larger impact on aerosol where ozone absorbs strongly (521, 602 and 676 nm) but other aerosol measurement wavelengths are unaffected.

29) L131: “. . .(602 nm. . . 521 nm)”: why? (see previous comment)

See above

30) L138: “. . .November 13. . .”: please mention the year

Added

31) 142: “The opposite of what. . .” → “This is the opposite of what...”

Updated

32) L143: “The extinction ratio becomes. . .”: here talking about Nevado del Ruiz (not Pinatubo)?

Ruiz, clarified.

33) Why not aggregating Fig 4 and Fig 5 in a unique figure with 3 panels?

We preferred it this way.

34) Fig. 4: Why not restricting the yaxis scale to something like 2 to 4? There are no values <2 or >4.

Done

35) Fig. 4 caption: What do you mean with "the scatter"? I would just say "the time series of 1020... and 525 to 1020..."

Changed

36) Fig. 5: why not evidencing the pre-eruptive points (the cluster for smaller values of the 1020-nm AE) with a different colour or different symbol? Can it be possible to identify the points in the earlier stage of eruption, as well (so to corroborate the hypothesis of a different cluster of values, i.e. with ash)?

Done

37) L148-149: “The distinction. . .recognizable”: this can put more in evidence in Fig. 5 (see previous comment

Figure enhanced as requested.

38) L150: Has a sulphuric acid hypothesis been proposed earlier? A precise point where ash is discarded is not present in the previous text

This is now introduced more clearly in the introduction

39) L151-152: “Generally,. . .events”: Can this be shown more clearly?

We’ve clarified this discussion along with the discussion in comment 40.

40) L153-156: “This was particularly. . .higher latitudes events”: all this part is not very clear to me

See above.

41) L193-194: “At some point. . .reasonable”: this makes reference to Fig. 2? Please clarify

Added reference to Figure 2.

42) L197-201: “This relationship. . .space-based instruments”: how this can be done (inferring uncertainty in mono-spectral observations and evaluating aerosol modules in GCMs)? I feel that this should be discussed much more in depth

We have clarified this discussion to point out that since models with detailed aerosol microphysical models possess knowledge of the composition and size distribution of aerosol in space, it is a straightforward calculation to produce extinction at any wavelength.

43) L202-: in general, in Section 3 and 4 discussions of volcanic events considering existing information in the literature are lacking. For Ambae, for example, in the paper already cited, Kloss et al., 2020 (by the way , please correct the reference as this paper is now published in JGR and no more in preprint), the plumes are detected using SAGEIII observations and simultaneous increases of the AE and the partial column Ångström exponent (Fig. 8 of Kloss et al.). This can be easily put in connection with Fig. 7f.

Added.

44) L226: "*. . .*Ruang*. . .*": please add year of eruption

Done

45) L230-232: "The Kelud*. . .*um)": here is an example where your work can be put in context with existing literature. In the following paper, it has been shown that ash was present for a long time in Kelud plume: Vernier, J.-P., Fairlie, T. D., Deshler, T., Natarajan, M., Knepp, T., Foster, K., Wienhold, F. G., Bedka, K. M., Thomason, L., and Trepte, C. (2016), In situ and space-based observations of the Kelud volcanic plume: The persistence of ash in the lower stratosphere, J. Geophys. Res. Atmos., 121, 11,104– 11,118, doi:10.1002/2016JD025344.

Sorry this paper is on the 2014 eruption of Kelud whereas we are discussing the 1990 eruption for which no comparable data exists.

46) Fig. 7: it would be useful to have the indications (red dashed lines) for the two eruptions of Ambae and Ulawun

Done

47) Fig. 7 caption: the mention to the volcanoes names is probably redundant, as the volcanoes are also mentioned in the panels. Also "*. . .*vertical dashed lines." –> "*. . .*vertical red dashed lines."

Done

48) Fig. 8b: Why not quantifying this trend (linear regression and correlation parame- ter)?

We thought about this but decided to not do so at this time because there are sufficient questions in our mind regarding the details of this relationship (particularly linearity in log-extinction coefficient/extinction ratio space).

49) L242: "*. . .*(possible ash)*. . .*": but also sulphate-coated ash or large sulphates in the accumulation mode possible, how do you

exclude these?

Also possibilities as well as perhaps some ice crystals. Pure sulfuric acid droplets seem unlikely unless they are directly injected as droplets but with the lack of composition information, ash is a surmise. Clarified.

50) L248-250: "For instance, for Raikoke...and ratio.": this makes reference to the (complicated) issue of the mixing state of the aerosol population, including the possibility that ash is sulphate-coated and/or these particles may freeze. Probably a discussion about that is needed here.
We have included a brief discussion of alternative compositions

51) L250-251: "It is also possible...this event.": interesting, can you please develop this point a bit?

There was a pyroCB about a month before the Raikoke eruption that SAGE II observed at about 12 km. After the eruption the two events became indistinguishable rather rapidly so how they interacted, if at all, is an interesting topic for consideration. For instance, SAGE II and CALIPSO observed a blob of aerosol that originated at high latitudes slowly rose through the stratosphere and ends up near 25N. This behavior is not typical of volcanic material (to our experience) but is fairly common for smoke events like a pyroCB (reference to this blob is included). Whether the smoke material managed to pass through the volcanic layer in some way or if the blob is some sort of mixed aerosol material including sulfuric acid coated smoke particles, is difficult to assess at this time. At this point, we know that it is possible that they two events interacted and this is the subject of current research and a forth coming publication.

52) L257: "These are initially...Figure 11)": How is it visible in Fig. 11. And also, why Fig. 11 is discussed before its content is defined (in the following lines)?

Updated to reference Figure 2a.

L257: "...but coagulate...": if talking about coagulation, why not of heterogeneous nucleation/condensation over pre-existing particles (sulphate aerosol or ash)? At this point, it looks clear to me that a discussion on the mixing state and aerosol micro- physics is quite needed

In this case, we are discussing what we infer to be a process that produces the observations we report. Condensation onto existing particles or the rapid scavenging of these new small particles would always decrease the extinction ratio as particles would become systematically larger. We cannot exclude that this process happens at some level but that it is not consistent with the observations reported herein.

53) Equations at L270-273 and inherent discussion: see Major Comment #1

This discussion has been clarified as indicated in the response to Major Comment #1

54) Please add equation numbering

Added

55) The priority of argumentations in the Conclusions is not clear to me. The main results of this work are probably the evidence of the dependence of the ER from the eruption intensity and AE (Fig 8b) but this is not even mentioned in the Conclusions

We have clarified the goals and outcomes from this study both in the introduction and in the conclusions including referencing the key findings shown in Figure 8b.

56) L327-328: “The primary goal. . .process”: as mentioned above (Major Comment #2 and Minor Comment #4) this is not discussed in the text, so it is strange to see this in the Conclusions

We have clarified the difference between a long term goal (or motivation) of improving OSIRIS-like observations and the goal of this paper which is to establish how small-to-moderate volcanic events manifest themselves in SAGE-like observations.

57) L330-331: “It is clear. . .therein”: this is actually not very clear to me: in the text there is no assumption on the aerosol chemical composition.

We’ve clarified that the model used in Section 4 is based on sulfuric acid aerosol. Here we’ve added material that notes that the model is homogeneous nucleation of very small particles that subsequently coagulate.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-480, 2020.