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.

Anonymous Referee #3

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General comments

The paper describes the relationship between the perturbation in the aerosol extinction coefficient and the extinction ratio or particle size in the early months following small to midsize volcanic eruptions. The authors acknowledge the limitation of the analysis, which were restricted to early months following volcanic eruptions while tracking the main layer, mainly because of the presence of significant amount of ash. I believe that the analysis presented here are incomplete and the paper would benefit from expanding the analysis to a longer period (not only the early months) and wider range of altitudes, rather than the peak of the aerosol layer. In addition, the paper needs clear and defined objectives (see my comment below).

Specific comments

Page 3, L64-66: ‘space-based missions are mostly limited to single wavelength mea- surements associated with instruments such as . . .’ The statement is only valid for the datasets used in GloSSAC climatology. CALIPO routinely provides aerosol measure- ments at two wavelength 532 and 1064 nm, while OSIRIS can provide measurements at 750 and 1530 nm, which were used to derive stratospheric aerosol particle size in- formation (Rieger et al., 2014). In addition, MAESTRO (McElroy et al., 2007), GOMOS (Vanhellemont, et al., 2016), and SCIAMACHY (Taha et al., 2010; Malinina et al., 2019) can also provide aerosol measurements at multiple wavelengths. While the quality of the measurements is debatable, its existence is not.

We’ve included some additional sources of stratospheric aerosol measurements and clarified the data available.

Page 3, L73-74: ‘It is also, until the start of the SAGE III mission, a period where the long-term stratospheric record is less robust due to the lack of global multiwavelength measurements of aerosol extinction coefficient.’ Again, the Authors are describing the GloSSAC dataset rather than the global stratospheric records. See previous point.

This point is similarly clarified.

Page 3, L76-81: ‘Thus, the original aim of this work was to understand how volcanic events manifest themselves in SAGE II/III observations with the goal of 1) inferring the uncertainty in single wavelength space-based data sets that use a fixed aerosol size distribution as a part of their retrieval algorithm such as the OSIRIS and CALIOP and 2) infer how well the wavelength dependence can be estimated for these single wavelength measurements.’ The authors failed to address both objectives and I don’t see how the paper’s findings, in its current form, can be of any use to these instruments because of the limited analysis shown her. I suggest either expanding the scope of the work to address those objectives or revising it to more realistic objectives.

We’ve revised the goals of the paper (and moved the discussion of them primarily to the introduction) as the need to characterize the way in which small-to-moderate volcanic eruptions manifest themselves in SAGE-like observations and that the goal for applications to OSIRIS and other data sets is a longer term objective.

Table 1: ‘Volcanic eruptions and smoke events that significantly impact stratospheric aerosol levels in the Version 2.0 of the GloSSAC data set’ Figure 1 shows very low aerosol and no impact of any of the volcanoes listed between 1998-2004, which implies that none of these eruptions reached the stratosphere. I suggest revising Table 1 by removing all volcanic eruptions that is not seen by GloSSAC. In addition, add the eruption altitude, similar to Table 2.

Since the Table refers only to Figure 1, the altitude of the event other than being stratospheric isn’t relevant.

Figure 3: Unlike the rest of the analysis shown in this paper, the figure is for GloS- SAC zonal mean aerosol stratospheric optical depth and ratio rather than extinction coefficient and

ratio for the peak aerosol layer. I suspect that the extinction ratio is inaccurate, given that SAGE II measurements were missing following the early months of the eruption, and the dataset was mostly reliant on single wavelength Lidar mea- surements. For the sake of consistency, I suggest using SAGE II measurements of the aerosol extinction and extinction ratio, similar to Figures 4-7.

GloSSAC data is used for a relatively minor and a broadly accepted idea that the wavelength dependence of aerosol extinction/optical depth becomes very small (near 1) very quickly following the Mt. Pinatubo eruption. The data the reviewer requests is partially in Figure 7b. We really don't see any reason to change this figure. The process for filling missing data in GloSSAC is described in (Thomason et al., 2018;Kovilakam et al., 2020).

Figure 4a: Can you use different color for the extinction values at 20.5 km?

All data is at 20.5 km.

Figure 4b: Can you plot all data shown in Figure 4a while using different color for extinction ratio at 20.5 km?

All data is at 20.5 km.

Figures 4, 6, and 7: The Days label is confusing. Can you use month/year or something similar?

We have explained the coordinate system in the caption more fully.

Figure 5: change 'Same data as shown in Figure 3' to Same data as shown in Figure 4a or 4b. In addition, can you specify if the data shown are for 20.5 km or all altitudes? If so, can you use different color for 20.5 km.

Done

Page 6, L189 ‘the maximum extinction coefficient at 525 nm does not necessarily occur at the same altitude or time as the maximum in 1020 nm extinction coefficient’. This is all the more reason to track the plume at different altitudes/zones rather than the maximum extinction value.

See response below.

Figure 7: I suggest over plotting points symbol to show the number of points used in each figure, adding a second vertical line for Ambae and Ulawun denoting the second eruption date, and using the same x-axis scale for all figures.

Done.

Figure 9 and page 7, L220 ‘It is clear here that the maximum in the extinction ratio lies below the main peak in extinction coefficient in the tropics and, notably stretches to higher southern latitudes and the maximum values actually occurs near 30° S de- spite more inhomogeneous conditions at this latitude than in the tropics’ and page 8 L240: ‘Both eruptions show increased aerosol extinction coefficient ratios away from the main aerosol peak suggesting, at least in part, behavior more consistent with most eruptions.’ This is interesting observation that raises more concerns about the analysis shown in Figures 6 and 7. Is it possible that tracking the maximum extinction value is not the best approach as it might bias the outcome, especially where the aerosol extinc- tion is very large? Very large extinction values can be caused by the presence of Ash particles, or it is an artifact in SAGE measurement when the volcanic plume is localized and spatially inhomogeneous. In addition, the result results shown here can be easily biased by

SAGE limited coverage. Perhaps repeating Figure 7 using zonal means at different altitudes and extending period of the analysis can produce a more consistent relationship between the aerosol extinction and extinction ratio perturbations.

We agree that tracking the maximum in extinction raises the likelihood that the observations include ash particles. However, material is transported out of the latitude band where the event occurred, it is about as likely to include ash as sulfuric acid aerosol. In addition, as the observations become further from the point of injection, there is a greater likelihood that a SAGE II profile with a volcanic signature at a particular altitude consists of an inhomogeneous mix of air unaffected by the eruption with the volcanic plume through either mixing or the plume only occupying a part of the measurement volume. This makes the interpretation of the an extinction measurement pair more problematic much in the way that SAGE observations of water clouds are better interpreted as aerosol/cloud mixed extinction coefficient values rather than purely 'cloud' extinction coefficient (Thomason and Vernier, 2013). While this is always a problem, we have focused on the densest part of the plume as a means to mitigate this effect. We have included mention of this issue in the paragraph originally beginning at L81 (It should be clear…).

Page 8, L246: 'peak extinction level as essentially all the data follows the mean rela- tionship in Figure 7g.' The sentence is unclear. This correct for Ulawn, however, the paragraph is discussing Raikoke eruption. Please revise the sentence.

We have clarified this statement.

Page 8, lin3 253: ‘It is also possible that a pyrocumulus event, that occurred in Alberta, Canada just prior to the Raikoke eruption, plays a role in the evolution of extinction fol- lowing this event.’. In addition to Alberta, there was a second PyroCb event in Siberia, Russia in July 2019, that reached the stratosphere and was also seen by SAGE III/ISS (https://directory.eoportal.org/web/eoportal/satellite-missions/i/iss-sage-3, Figure 7). It is more likely that the smoke aerosol interfered with the Raikoke analysis. If the two different aerosol layers were separated (which is most likely), then repeating the analy- sis at different altitudes instead of tracking the peak extinction can explain the behavior seen in Figure 7h.

We have found the separation of the pyrocumulus events and the volcanic events in the lower stratosphere to be particularly complicated. The sulfuric acid aerosol layer from the Raikoke eruption was impacted by smoke injected from the Canadian and Siberian wildfires. The degree to which the two mixed is uncertain, but the data clearly show some mixing. Despite a secondary cloud (most likely smoke) breaking from the main Raikoke layer and moving south, our research indicates that smoke likely remained within the main Raikoke peak until it was fully dispersed. Therefore, repeating the analysis does not deconvolve these two components. We have a manuscript in preparation dealing with some of the unusual aspects of this eruption which is a really interesting event or set of events given the smoke plumes.

Section 4: The first paragraph leading to the aerosol perturbation model relies on unsupported speculations (by the authors own admission), and it can benefit from the addition of few references that support those assumptions.

References added.

I also find the whole discussion regarding the model calculation and figure 11 confusing and I can’t relate the figure to what was presented in the previous section. The aerosol extinction ratio perturbations for Ambae are almost double those for del Ruiz eruption and figure 11b in particular shows large extinction ratios (>5) not seen by any of the cases shown in this paper. Close inspection of both figures indicate that the extinction ratio perturbation is very sensitive to the baseline ratio, and if Manam or Ulawn baseline values were used, the extinction ratio values would've been even higher.

That the differences for the same perturbations manifest themselves differently depending on the pre-eruption background is one of the points behind the model and is consistent with what is shown in Figure 8. The other key reason for including is to provide a rationale for our interpretation of observations as suggesting the homogeneous nucleation of many small particles followed by coagulation. The increase in extinction ratio as the aerosol perturbation becomes relevant followed by a relaxation to smaller values. We have substantially rewritten the introduction to this section to explain the goals and rationale of this section. Some extinction ratio values exceed those observed herein are associated with hypothetic 'volcanic' events substantially greater those observed in the events in Figure 11.

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

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