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 #2

Received and published: 8 July 2020

General Comments

This paper presents a simple study of the way the 525 nm to 1020 nm-extinction coef- ficient ratio, a proxy for the particle size, evolves in function of the 1020 nm-extinction coefficient during the early period following the volcanic eruption. This study is based on ten test cases of volcanic eruptions that affected the stratosphere and were mea- sured by eitther SAGE II, or SAGE III on ISS. The author's aim is to show that the joined evolution of these two parameters follows in many cases a simple relationship, regardless of the particle size distribution or the details of the circumstances of the eruptions.

This study definitely lacks precision in my opinion. The whole analysis is based on approximations, quite subjective considerations and fast conclusions that remove con- vincing ground to the argumentation, yet based on a necessary limited sample of erup- tions. Furthermore, since one of the two key dataset (i.e. SAGE III/ISS) shows a bias at one of the considered wavelengths, they interpolate these values using a simple law, while the situation after a major eruption like the ones studied here is expected to be complex. Building on approximate data with coarse approximations and arguments gives results that are sometimes questionable, especially in view of some applications the authors have in mind.

Concerning the text, the authors' formulation is in some cases so general that they make the described concept no more relevant, e.g.: "the stratospheric aerosol optical depth" [at 525 nm or 1020 nm], mentioned as if it was a fixed number. The authors should also be more accurate in their naming of the quantities they consider (e.g.: "[aerosols] extinction coefficient perturbation ratio", "perturbation aerosol extinction co- efficient ratio", "perturbation extinction ratio", "perturbation ratio"). Overall, sloppiness brings a lot of confusion in the text and undermines the argumentation.

Concerning the case of the Mt Pinatubo, it is particularly surprising that this case is considered and discussed without any mention of the fact that SAGE II could not ap- propriately measure the extinction coefficient during several months after the eruption due to the extreme opacity of the volcanic cloud. This is however a major drawback for the present study.

Consequently, using this analysis and the relationship inferred by the authors to vali- date complex models as suggested by the authors seems far premature and overrated.

These points (above) are dealt with as specific points below. We have corrected the usage of perturbation terminology. Since much of the finding we show below are based on observations and, in fact, could be inferred from SAGE II observations alone (whose mission ended in 2005), we argue that the basic observationally-based findings as shown in Figures 7,8, and 9 are overdue.

Specific comments

L. 15, p. 1: The integral over its whole domain of definition, of a distribution function is equal to 1. The change in multiple orders of magnitude should concern the aerosol size number density, not the aerosol size distribution.

Corrected to clarify that the 'several orders of magnitude' refers to extinction coefficient rather than size distribution

L. 24-25, p. 1: For such general consideration, I would suggest citing Robock, Rev. Geophys., doi: 10.1029/1998RG000054, 2000.

Done

L. 1-2, p. 2: To illustrate the efforts to model the climate impact, I suggest citing some work by Timmreck et al., for instance, Timmreck et al., Geophys. Res. Lett, doi: 10.1002/2015GL067431, 2016.

Done

L. 55-58, p. 2: This sentence is correct, but gives still a biased view of the reality: the SAGE II mission was exceptional in several senses, one of them being that it spanned a period with a particularly high amount of very large volcanic eruption. If it had been launched 10 years later, the situation would have been very different. The authors should be attentive to give a correct view of the reality.

We have clarified this statement.

L. 56, p. 2; L. 79, p. 3; L. 360, p. 13; L. 437-438, p. 26; caption Figure 1, p. 15: Kovilakam et al. is not published so far, and mentioning it as if it was the case is not ethical and should not be done. If this paper is not accepted in due time, please refer to another paper, e.g. Thomason et al., 2018.

This paper is in the final stages of the publication process. We have updated the reference to the Discussions paper but we should be able to update to the final reference before this paper is completed.

L. 63, p. 2-L. 65, p. 3; L. 72-75, p. 3: This is not true and should be corrected. The ESA Envisat mission provided three experiments with high interest for aerosol stud- ies: the SCIAMACHY spectrometer measuring in the UV-visible-near-IR range (von Savigny et al., doi:10.5194/amt-8-5223-2015, 2015; Noel et al., Atmos. Meas. Tech, doi:10.5194/amt-2020-113, in review, 2020), the IR limb sounder MIPAS (Griessbach et al., Atmos. Meas. Tech., doi:10.5194/amt-9-4399-2016, 2016), and the UV-visible- near IR stellar occultation instrument GOMOS (Bingen et al., Remote Sensing Env., doi: 10.1016/j.rse.2017.06.002, 2017). Also, the ACE-MAESTRO mission provided aerosol extinction from solar occultation measurements (McElroy et al., Appl. Opt., doi10.1364/AO.46.004341,2007).

While in retrospect, this sentence could be deleted completely, we have included the references to the ENVISAT instruments.

L.103-104, p. 4: In the case of the major eruptions that the authors consider, the situ- ation is definitely more complicated than in this simple monodispersed aerosol model.

In this case, we have changed the reference to simple aerosol radius rather than reference size distribution.

L. 106-108, p. 4, Figure 3: “The stratospheric aerosol optical depth” is vague. Please specify. Figure 3: SAGE II was not able to measure correctly the aerosol extinction during the months following the Mt Pinatubo due to the saturation of the atmosphere in aerosols. How do the authors infer the “stratospheric aerosol optical depth” during this period?

We have changed the text to reference the GloSSAC v2.0 data set. Descriptions of how missing data are accounted for are explained in the Kovilakam paper and the v1.0 paper (Thomason et al., 2018) in detail.

L. 125-126, p. 4: The authors should be specific: Which value of the 525 nm and 1020 nm aerosol extinction coefficient do they choose to infer “the impact of these eruptions”? The conversion of sulfur gases to sulfuric acid is a process that requires several weeks, and the presence of ashes may significantly influence the aerosol pop- ulation in the early phase after the eruption, as mentioned before by the authors.

We have clarified this statement.

L. 128-129, p. 5: This is a serious setback for this study! A “simple Angstrom coef- ficient” interpolation states that the aerosol population has a simple structure, which is probably not the case in the post-eruption period where different aerosol modes (thin particles, ashes, aged aerosols) coexist. The interval [448 nm, 756 nm] is quite large and inferring the extinctin coefficient value at 521 nm is uncertain. Furthermore, while implementing an interpolation, why don’t the authors interpolate at 525 nm, the wavelength effectively used in the paper?

We agree that the bias is certainly inconvenient for this study, however we do not believe that this interpolation introduces significant uncertainty into the analysis. The new text:

We have replaced the 521 nm data product with an interpolation between 448 and 756 nm that employs a simple Angstrom coefficient scheme (602 nm and 676 nm measurements have biases similar to like those at 521 nm). This is possible since the stratospheric aerosol extinction coefficient is always observed to be smoothly varying with wavelength and approximately linear in log-log space. The presence of the 521 nm bias is inferred using this methodology and this approach was used in the validation paper for SAGE III/Meteor 3M aerosol data (Thomason et al., 2010). The differences between the inferred 521 nm extinction coefficients and the reported values in the lower stratosphere (tropopause to 250 km) average about 6% and are usually less than 10%. Above 20 km the differences are usually on the order of 1 to 2% with the estimate usually less than the observation. This difference is probably a reflection of the limitation of the accuracy of the interpolation and consistent with past uses of the same approach (Thomason et al., 2010). In any case, the effects of small to moderate volcanic eruptions on aerosol extinction coefficient as a function of wavelength described below are consistent whether 448 or 521 nm aerosol extinction coefficient in used in the SAGE III analysis. We interpolate the 521 nm values solely for comparison purposes with SAGE II data and this process has minimal impact on the conclusion drawn below.

L. 132-133, p. 5: How do the authors assess the difference between the interpolated value and a good approximate of the true value, if the SAGE III/ISS are biased?

See above text.

Figure 4: Please indicate the Julian day corresponding to 1 January 1985. Is it Day 200?

This figure is in days since 1 January 1985 which would be day 1 which is now noted in the caption.

L. 148-149, p. 5: Without indication of which points correspond to volcanically pertubed observations, and which ones to unperturbed periods, it is not possible to assess the pertinence of this sentence.

We have updated the figure to color the pre-eruption observations red.

L. 149-150, p. 5: What is a “low extinction coefficient ratio”?

We have updated this text to refer to extinction coefficient ratios close to and occasionally less than observed prior to the eruption (<2.3 or so)

L. 150, p. 5: Was this hypothesis checked in some way?

SAGE-like observations contain little or no information on composition. We have updated the text to indicate that the composition is unknown.

L. 152-155, p. 5: If the authors clearly excluded any considerations about fire and pyrocumulus events, what do they mention them here? Or conversely, why did they reject such events in L. 69-70, p.3, if they use them here?

This is simply a comparison of low vs high latitude events and their differences in zonal variability. The nature of the events is, in this case, immaterial.

L. 158, p. 5: Since the time between two visits is about a few weeks to months, the observed evolution potentially starts at very different stages of plume development, possibly characterized by very different aerosol population features (importance of the ash fraction, development of the aerosol microphysics, mono/multimodal character of the size distribution, etc.).

We agree that this is a complicating factor as we discussed L81-93, p3

L. 160, p.5: How do the authors select “the maximum value”? As shown in Figure 4, such situation is rich in outliers. Do the authors consider the highest outlier? Which is the relevance of such a choice, in a situation where the instrument measures locally, at a single time occuring after “a few weeks to months”, more or less close to the eruption?

The data is well behaved and low noise particularly when the aerosol extinction levels are enhanced by an eruption. This process is primarily a way to find the altitude (and the associated extinction coefficients) of the volcanic layer in each profile where it can vary from profile to profile within a temporal bin and over the months following the eruption. Other strategies were tried but none particularly changed the results. We have clarified the text.

L. 161, p. 5 and Figure 4: What do the authors mean by a “9-point window”? Is each point in Figure 4 such a “maximum” in a 9-point window?

Figure 4 is for 20.5 km. We have clarified the text that the window is effectively a 4 km window in each vertical profile (9 points in the 0.5 km vertical resolution profiles).

L. 162, p.6: “main aerosol layer” vs “entire layer”: do the authors refer to the volcanic cloud, as opposed, say, to the combination of the Junge layer and the volcanic cloud?

In this case, it works out to basically the same thing but we are referring (and have clarified) that we focused on the volcanic cloud.

L. 165, p. 6: What are the authors speaking about? “To produce a mean value” of what? “we required a minimum of 6 points”: which kind of points and over which selection?

We moved this discussion forward in this paragraph. Particularly in the tropics, there are occasions where the number profiles in a temporal bin is very low and average statistics and sampling is pretty poor. We found that requiring at least 6 profiles in a temporal event caught all of these poorly sampled periods. We have clarified this in the text.

Figure 7: Same question as for Figure 3. Furthermore, the authors should add the Ruiz case to ease the comparison.

Done.

L. 172-178, p.6: This classification is poorly convincing. Concerning the events with “a rapid increase in aerosol extinction coefficient and ratio following the eruption”, this can- not characterize Ambae for which the increase in extinction ratio mainly spans about the 120 days preceding the eruption (as indicated by the anthors), saturating at the mo- ment of the start of the eruption while the extinction coefficient just starts to increase. The authors find the cases of Ruang and Kelut as “following their own way” while these cases have many similarities with Raikoke that they see in another category. Clearly, the categorization is subject to interpretations, what makes the classification over an 8-eruption sample poorly convincing.

The Ambae plot shows the effects of two eruptions occurring in April and July 2018. For the second event, the extinction ratio increases somewhat (on the scales shown in the plot) earlier than does 1020-nm aerosol extinction coefficient. We have simplified the categories to two types: one in which the aerosol extinction ratio increases (or remains large) relative to the baseline and ones where the initial change decreases the aerosol extinction ratio followed by some recovery to larger values.

Figure 8a: The authors should show in some way, for each eruption, where is the “before” and where is the “after”.

Since extinction coefficient always increases following an eruption, we have noted that the before points are always the left most data value is the before value and the right hand value is the after data.

L. 179-181, p. 6: How do the authors define “the first data point”? Is it the extinction ratio value corresponding to the earliest time plotted in Figure 7? In Figure 7, the time interval considered between this first data point and the eruption start time is different in each case and the choice of this time interval obviously weights on the initial value of the extinction coefficient ratio, and hence of the “extinction coefficient perturbation ratio” shown in Figure 8b. Furthermore, in at least 2 of the 8 considered cases (Cerro Hudson and Manam), there is no clear indication at all that the start of the curve corresponds to any “baseline”, and in several other ones (Kelut, Pinatubo, Ruang, Ulawun, Raikoke), the presence of such “baseline” is very uncertain. Finally, the scales used for both the extinction coefficient and the extinction coefficient ratio are different and differently related for all cases, making the establishment of parallel behaviour uncertain. Hence, drawing any conclusion on these values, taking into account the high uncertainty on the value of the maximum extinction [ratio] (See comment on L. 160, p.5) looks particularly hazardous.

The first data is the first data shown in Figure 7. As discussed in section 2, the temporal sampling from both SAGE II and III is sparse particularly in the tropics and this is one of several challenges in using this data in this analysis. Within these data sets, we observe that aerosol extinction coefficient levels at a given altitude and latitude slowly vary with time even apart from recent volcanic activity due to the recovery from past volcanic activity and seasonal processes. For the events discussed here, due to the timing of the events, these changes are very small compared to the volcanic events that follow and, in terms of the calculation of perturbation values, the exact background level has only a secondary effect on the *calculated values. As a result, the timing of the 'before' samples does not materially affect these results. We have clarified this in the text. The scales used in the plots are selected to highlight the behavior of each event and is necessary considering the several order-of-magnitude differences between events. The Ruang and Manam events occur during the 50% duty cycle portion of the SAGE II record and thus we have less data during this period. The base values are the closest in time available and occur during a volcanically quiet period and are adequate for this analysis. We have changed the reference from 'baseline' to 'before'*

L. 184-186, p. 6: See previous remark.

See previous reply

L. 197, p. 6-L. 201, p. 7: Beyond the uncertainties related to the values plotted in Figure 8b (See comment on L. 179-181, p. 6), this model is so coarse, ignoring the fact that some eruption produce more sulfur gases and some other ones more ashes, ignoring all effects related to the geolocation and consequent dynamical features of the plume dispersion as well as seasonal effects, and ignoring intrumental limitations (no- tably SAGE II's "blindness" in the early phase of the Pinatubo eruption), that it seems absolutely premature to draw any conclusions from these 10 cases among which only a handful of the considered ones follow a nice flat curve. Applying such relationship to other data sets from limb sounders where retrieved extinction values depend yet on a set of assumption, is at risk to get quite far removed from the truth. Instead of suggesting using this tool for the evaluation of interactive aerosol modules for GCMs and ESMs, it would be useful, on the contrary, to use GCMs with elaborate aerosol microphysical models to assess if this relationship is sufficiently grounded to be used elsewhere.

As we discuss in section 2, with few exceptions, the ability to infer the detailed processes within a volcanic *plume and in stratospheric aerosol from SAGE-like measurements are limited. However, the instruments do not care about the processes that create the optical properties that they measure. The measurements themselves are robust and the features we show in Figure 8 are readily apparent in the most cursory examination of the data. It would be convenient to have many more events on which to examine this relationship but such data are not available. Still, the observed relationship occurs in 80% of the data available which we find compelling. We have modified the text to reflect that, in Figure 8b, perturbation ratio is reasonably well sorted in perturbation extinction and removed the reference to linear or curved lines as probably being a little too enthusiastic. We did not originally plot a line or provide a fit in this case in part to reflect the low data amount and the uncertainty in the individual points. We agree that understanding is most compelling when there is closure between observations and models. That is why we suggest that these data provide an opportunity for closure tests. We have modified the final sentence in this paragraph to clarify our intent.*

L. 206-207, p. 7: If two closely related eruptions from a same volcano, occurring within such a short period of time, have yet to be splitted in a “regular case” and an “outlier”, it becomes very hard to find any foundation in the proposed relationship. Even if other observations could bring some evidence that the plume composition, due to this quick succession, was very different in both cases, such argument could not be invoked since this kind of consideration has been excluded in this study.

In this context, this comment reflects that the second event may, at first glance, be an outlier. Further analysis shows that it is not. Generally, there is no reason to believe that eruptions from a volcano will necessarily be the same when separated months or years.

L. 210-213, p. 7: Choosing the “initial state” of the second eruption of Ambae (with a value of 4.9 for the extinction ratio) is a highly speculative exercise ignoring the fact that the microphysical evolution is highly perturbed by the addition of fresh material. In no way, the situation corresponding to this value of 4.9 for the extinction ratio can be considered as a “baseline”. Furthermore, the given values are quite arbitrary: the value of 5 is never reached by the extinction ratio (the maximum is about 4.7), and the extinction ratio decreases far below 4.1, toward a value lower than 3.5 and that cannot be estimated from the limited curve shown in Figure 7. Hence, putting both Ambae eruptions again (and in contradiction to what is observed in L. 206-207, p. 7, see comment) in the category of “regular cases” is particularly dubious.

Again microphysical processes that may or may not be occurring do not change the observations. The April eruption of Ambae is overwhelmed by the much larger second event and we can get essentially the same perturbation values whether we use the minimum just prior to the second eruption or the values prior to the initial Ambae eruption as the ‘before’ values. We have clarified that the values reflect the initial changes observed following the eruptions. In retrospect, the 4.9 value appears to occur after the July Ambae eruption and we have modified the text to reflect that the extinction ratio changes from around 4.5 just prior to the second eruption, to 4.9 with the earliest observations of the new aerosol and then to 4.1 when the aerosol extinction coefficient is a maximum. We have changed the initial extinction coefficient ratio from ‘nearly 5’ to 4.7.

L. 220-221, p. 7: This looks like a fast conclusion about a situation where microphysics, dynamics, and multiple injections of volcanic matter combine in a complex way, as illustrated in Figure 9.

The first Ambae eruption is inconsequential to this depiction. While we are aware that there are complex processes going on, this is what is observed by the instruments and it is consistent with what is depicted in Figure 8.

L. 223, p.7: “the main aerosol layer”, “all parts of the volcanic cloud”: please clarify (See also comment on L. 162, p.6).

Updated to ‘the densest part of the volcanic plume’

L. 226-228, p. 7: What do the authors mean by “the first observations”? Also, the end value is rather 2.9 than 3.

Added ‘of the plume’

L. 228-229, p.7: Where do the authors find this value of 2.7? Such value is never reached in Figure 7, nor is indicated in Figure 8. Is the “perturbation extinction ratio” another quantity than the ones illustrated in these figures?

We’ve updated the value to 2.9 and the later value from 4.3 to 3.9.

L. 230, p. 8: What is a “compact extinction coefficient [ratio]”?

Updated to indicate that the scatter of the data is mostly compact.

L. 230-235, p. 8: It is surprising to give such an importance to outliers that in other circumnstances would be fastly overlooked.

There are sufficient numbers of observations showing enhanced extinction and low extinction ratio to impact the averages (unlike those in Figure 5 for Ruiz) so their presence cannot be ignored.

L. 239, p. 8: What do the authors mean by “the spread of the Nevado del Ruiz, Cerro Hudson and Raikoke eruptions”?

Changed to ‘differences between’

L. 243-251, p. 8: This part of the text looks like a suite of speculations without attempt to analyse them seriously, making this enumeration not very useful.

We feel obligated to enumerate issues that may impact the results we show but cannot currently explain.

L. 257-259, p. 8: The sentence is unclear. Please rephrase.

Rewritten

L. 263-266, p. 9: The sentence is unclear. Please rephrase.

Rewritten

L. 286, p. 9: What do the authors mean by “identical perturbations”?

This has been clarified in the text.

L. 285-293, p. 9 and Figure 11: Basically, the extinction coefficient at 525 nm and 1020 nm and hence they ratio, can be exactly calculated by Mie theory for fixed values of the aerosol volume density and for single-radius particles. Hence, the differences found in Figure 11 for Nevado del Ruiz and Ambae only shows in which extend this simpel

model of single-radius particles departs from the reality. Furthermore, the authors mention coagulation effects, but ignore sedimentation which is nevertheless crucial after a large eruption, and high from the early post-eruption period.

We did not make the purpose of this simple model sufficiently clear and thus have expanded the discussion of its rationale. We do not intend for it to capture all aspects of microphysical processes going on following an eruption (it is indeed far too simple for that). We do use it show to *demonstrate why we believe that the observations are consistent with the idea of the nucleation of many small particles following a small to moderate eruption (that initially may be invisible to SAGE-like measurements) followed by coagulation to larger, optically significant particles. There is an extensive discussion of the limitations of this model in the third paragraph of this section which highlight it shortcomings.*

We had extensive discussions among the authors regarding what the observations mean and the nucleation of many small particles was the only process that we can see capable of producing what is measured by the instruments. We have highlighted the lack of sedimentation as a shortcoming particularly for large eruptions though our focus remains on small to moderate eruptions. We believe that it is reasonable to offer an explanation of what we believe the measurements mean.

L. 289, p. 9: There is no single timescale used in Figure 11, nor in Figure 2, and the authors should thus not use this concept alone.

There is no timescale for Figure 11 or 2. Using particle size as a pseudo-time scale in Figure 11 is explained in the text and shortcomings related to this discussed. This is not relevant to figure 2 which is simply a plot of Mie extinction kernels as a function of radius.

L. 305-307, p. 10: From the simplicity of this model and the fact that important aspects are neglected (e.g. the absence of sedimentation which critically influences the extinction and extinction ratio), I do not think that any conclusion can be drawn about inferring primary microphysical effects from these SAGE II and III/ISS observations.

We think we have adequately demonstrated why we believe our interpretation of the results is reasonable.

However, it is clear that it is only through closure between observations and modelling that confidence in this inference can be obtained.

L. 323, p. 10: Which “measurement paradigms” are the authors talking about?

Changed to simply ‘observations.’

L. 327-328, p. 11: Using a tool to assess the data quality of a data set is only mean- ingful if this tool is not based on coarser assumptions and approximations as the ones leading to this data set. I am not sure that this is the case in the present work.

We do not understand what the reviewer intends here. In this case, we are referring to using outcomes from this study as an aid to improving data quality for instruments like OSIRIS.

L. 331-333, p.11: Among the different applications proposed by the authors, the use of an extinction ratio value inferred from a relation established from the points 6-5-8- 7-0-2 in Figure 8b to fix the size parameter used in the OSIRIS retrieval (e.g. through the Angstrom coefficient) seems to be the only one for which this simple model could have a real added value, in my opinion. It is really a pity if this model even cannot be beneficial in this framework.

The use of these results in OSIRIS retrievals is an on-going study which we hope will result in positive improvements in the OSIRIS aerosol data products in the future. WE have indicated this in the text.

Technical corrections

Caption Figure 9: It would be useful to indicate here the latitude of the Ambae volcano.

Done

L. 216-217, p. 7 and caption Figure 9: the indications of wavelength and time are not consistent between the text and the caption. Please specify the exact time duration (start and end time).

Corrected to September.

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