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Atmospheric Chemistry and Physics Discussions

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

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The manuscript ''Evidence for the predictability of changes in the stratospheric aerosol size following volcanic eruptions of diverse magnitudes using space-based instruments, Thomason et al." discusses the relationship of multiwavelength aerosol extinction 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 interesting 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 b) 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 community 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.

2) The manuscript structure has to be improved. In the present version, the main results 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 measurement 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, throughout the manuscript, to introduce the Figures sequentially in the text and not to discuss

them before presenting their content. 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

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)

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

4) L18-22: "Despite this limitation. . .particle size": these two points (the use in evaluating 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
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

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

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)

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

9) L31: "…either the measurements…": please develop a bit to clarify how measurements have been used in global models to derive climate impact of volcanic eruptions

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

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.

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

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

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

15) L61: "…subtly modulate climate…": probably it can be mentioned that the aggregated impact of these "small-to-moderate" eruptions is significant (see also Ridley, 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)

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

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

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

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.

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

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)

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

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

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

25) Are footnotes allowed in ACP format?

26) L101-102: see Major Comment #2

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

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?

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

30) L138: "…November 13…": please mention the year

31) 142: "The opposite of what…" → "This is the opposite of what..."

32) L143: "The extinction ratio becomes…": here talking about Nevado del Ruiz (not Pinatubo)?

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

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

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

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)?

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

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

39) L151-152: "Generally,…events": Can this be shown more clearly?

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

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

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

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.

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

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.

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

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."

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

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

50) L248-250: "For instance, for Raikoke...and ratio.": this makes reference to the

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(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.

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

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)?

53) 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 microphysics is quite needed

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

55) Please add equation numbering

56) 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

57) 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

58) 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.

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