Atmospheric Measurement Techniques Discussions

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Comment on amt-2021-333

Anonymous Referee #1

Referee comment on "Identification of smoke and sulfuric acid aerosol in SAGE III/ISS extinction spectra" by Travis N. Knepp et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-333-RC1, 2021

Referee Report on "Identification of Smoke and Sulfuric Acid Aerosol in SAGE III/ISS Extinction Spectra Following the 2019 Raikoke Eruption", by Knepp et al.

General Comments

This paper presents a method to distinguish smoke particles and sulfuric acid aerosols based on the analysis of the extinction coefficient at 1020 nm and of the slope of the extinction spectral dependence between 450 and 1550 nm, estimated by linear regression. The method is applied to two recent volcanic eruptions (Ambae and Ulawun) and two wildfires (2017 Canadian and 2020 Australian wildfires), as well as to the more complex case of Raikoke which was coincident with large wildfires in Russia and Canada.

The proposed method seems to be effective and provides interesting perspectives in the challenging problem of aerosol type identification. Its limits of applicability and limitations are clearly mentioned. The introduction is particularly well-written and well-documented. On the other side, the spectral behaviour of sulfuric acid, back (BC) and brown carbon (BrC) brings quite a lot of questions and sometimes some confusion related to the fact that sulfuric acid and BrC seem to show quite similar spectral behaviour. This aspect is not discussed in the paper, and it would be important that the authors take them into consideration while revising their manuscript.

Specific comments

Title: It is surprizing that the authors put the focus of their paper on the Raikoke eruption, since this event is only one of five cases under investigation and the most difficult case with the less successful results due to the complexity of the situation. They could consider changing it.

L. 49, p.2: The notation “VEI-7-8” is unclear. Please clarify.

L. 118, p.4: Does the removal of all events with errors higher than 20% have a significant effect on specific time/latitude ranges by decreasing the amount of data down to a very small number of events specifically in these time/latitude intervals ? If it is the case, could this possibly induce some kind of bias in the results shown later in the paper ?

L. 133-135, p.5: The users are probably using Level 2 CALIPSO data in the version 4.2. On the contrary, the Level 3 data are monthly averaged. This important difference between the data level as well as the time duration used for the averaging should be specified.

L. 142, p.5: It would be useful to define the concept of “attenuated scattering ratio” or to refer to some paper where it is defined.

L. 149, p.5: Do the authors mean “as early as September” ? It is hard to see any trace of any secundary plume in figure 2(c) and the indication “secundary plume” is moreover shown in Figure 2(d).

L. 185, p.7: The authors should justify their statement that BrC is more likely to be present in the stratosphere than BC, or provide a reference in this sense.

L. 187-189, p.8 and Figure 3, p.9: For the sake of clarity, the same notation notation (“1.”, “2.” or “(a)”, “(b)”) should be used here and in Figure 3. Also, the text and figure captions should be clarified. In the text: Is this simulation made for the 3 situations (sufate, BC and BrC) ? In the caption: What is the reference used for the normalization ? In figure (a), what is the mode radius ? In figure (b), what is the reference wavelength for the calculation of the spectral slope ?

Figure 3, p.3: It might look surprizing, also with respect to the data in Table 1, that the sulfuric and BrC behave quite similarly (although scattering dominates for the first one, and absorption for the second one) while the BC curve have a significantly different behaviour ? Do the authors confirm that there is no confusion between some curves ?

L. 204, p.8 - L.207, p. 9: If the attribution of the considered species in the 3 curves is correct, in view of the large uncertainty on/variability of the mode radius and taking into account the fact that sulfuric acid droplets have no reason to have similar sizes to BC particles, one has to imagine a large uncertainty around each curve, and the difference between sulfuric acid and BrC is unlikely to be really detectable using this method. This might be an additional reason in the presence of "false positives" in both volcanic eruption and wildfire cases.

Caption Figure 4, p. 11: Please repeat in the caption the relevant information provided in the figures. Character size is quite small in the figure and the information mentioned in it is difficult to read.

L. 214-216, p.9: The authors' argument is not clear. In the four cases, the light blue points corresponding to the extinction ratio with the 1020-nm channel provides flat curves, in both volcanic (with dominating sulfuric acid) and wildfire (with dominating carbonaceous aerosols) cases. Their composition is thus quite different from the background in at least one case ! On the other hand, extinction ratio values change quite strongly with the 1020-extinction coefficient in the case of the 520:1550 ratio, for both volcanic and wildfire cases. What do they mean by this sentence ?

L. 223-227, p.10: The fact that a Pinatubo-like eruption cannot be assessed by the present method is not related to a difference of process (in all cases, SO2 has to be converted in sulfuric acid using the available water vapour and within some characteristic formation time), but to the size of the resulting particles: if the resulting particles are very large, the spectral dependence is flat and the extinction ratio is close to 1; if the resulting particles are not large with respect to the wavelength, a varying spectral dependence is found. Therefore, the explanation provided in L. 223-224 seems not the right one. Also, the role of ashes is not taken into account in the present discussion.

L. 240, 245-256, p.10: In view of the case illustrated in Figure 3, the linear regression is much more reliable if you don't consider the 1550-nm channel. Why do the authors conserve this 1550-nm channel ? Starting again from the case of Figure 3, the value of the slope is likely to be very similar in the sulfuric acid and BrC cases.

L. 268-272, p.12: The authors should explain or show on z figure why the slope is more negative / flatter than the background slope for sulfuric acid / smoke.

L. 286, p.12: I suggest that the authors add the corresponding value of the depolarization ratio after "do not depolarize".

Caption Table 3: The authors should specify what they mean by "Raikoke Primary" and "Raikoke Secondary", or refer to the explanation given in Section 7.2.

L. 301-302, p.14: Where are these number coming from ? In Table 3, the fraction of misclassified events reaches a maximum of 62% at 24 km height for Ambae and 100% at 15 km height for Ulawun. Please clarify.

Caption Figure 7: Please complete the caption and describe all panels to make the figure self-explanatory.

L. 352, p.17: Where is the estimate ">81%" coming from ? From Table 3, the fraction of identified smoke events is >60% if all altitudes considered, and >86% up to 24 km height.

Figure 13: I suggest that the authors use another colour for the indication "LB" and "R", which are poorly visible.

L. 404, p.21: Large particle have to grow from condensation nuclei to large particle by all successive microphysical processes (condensation, nucleation, coagulation). They are thus likely to need several weeks (up to one month) to become large particles. Per se, they are expected to be short-lived, but to appear later. The case shown in Figure 14 was measured on 30 June 2019, about one week after the eruption. Hence, isn't it likely that these particles rather concern ash ?

L. 420, p.22: Do the authors mean: "*either* a mixture of sulfuric acid and ash *or* smoke" ?

L. 422-423, p.22: The authors try to distinguish sulfuric acid and smoke, but do not discuss the distinction between BC from BrC, although their respective spectral behaviours illustrated in Figure 3 look quite different. Actually, in view of the relative similarity between the cases of sulfuric acid and BrC, wouldn't it provide a plausible explanation for many "false positive" cases in all cases where wildfires take place (Australian and Canadian pyroCb and Raikoke) ? It is noticeable that all these cases show a significant amount of "false positive" (see Figure 9, 10, 15, and 16) while both purely volcanic cases show only very few ones (see Figures 5-6).

L. 425, p.22: Citing altitudes of 19 and 20 km could be even more convincing.

L. 453, p.24: The statement is different here from above in the text (L. 199-202, p.8). The authors should replace "there is a chance for" by "the result is most likely to be", or just repeat that the method is not applicable in this case.

Technical corrections

L. 58, p.3: Duplicated “has”.

L. 130, p.5: “which“ should probably be removed.

Caption Figure 1: I suggest that the authors reproduce the time and geolocation of the four events in the caption for the safe of readability.