1 Response to Reviewer #1

We would like to thank Mike Fromm once again for providing feedback on this revised manuscript. We also appreciate Dr. Fromm's willingness to engage in fruitful conversations about this manuscript via e-mail and telephone. Our responses are provided below (red) to Dr. Fromm's comments (black).

The authors have once again made enormous changes and improvements to their manuscript. These are greatly appreciated, and allow me to acknowledge acceptance for publication after consideration of a few very minor suggestions.

We heartily appreciate Dr. Fromm's acknowledgement of our efforts to improve this manuscript.

Suggestion: reach out to Chris Boone regarding the ACE- FTS occultations presented herein, especially the two in July. Dr. Boone has dropped those from his current manuscript, under review, on Raikoke aerosols. This is largely because of uncertainties he had with the smoke spectra.

We have confirmed with Chris Boone that the data presented in these figures is accurate and the interpretation, as presented, is correct.

Section 6.4.4. The author's conundrum regarding Figure 20 and the generally assumed SO2-sulfate conversion timeframe of ≈30 days has some particularly relevant reinterpretation regarding sulfate abundance much sooner post eruption. Guo et al. (2004; doi:10.1029/2003GC000655) discussed observations of very young sulfates in the Pinatubo cloud. De Vries et al.(2014; doi:10.5194/acp-14-8149-2014) documented stratospheric sulfates in the active Nabro eruption umbrella cloud and in the days immediately after the 13 June 2011 eruption. In addition, two works have shown substantial Raikoke stratospheric aerosol optical depth well within the first month after eruption (Kloss et al. 2021; Gorkavyi et al. https://doi.org/10.5194/amt-14-7545-2021). And since CALIOP is invoked in Knepp et al., it might be worth reviewing these data to show that the Raikoke SO2 cloud was imbedded with aerosols each day after the eruption. While some of these observations might be interpreted as ash, the important take-away is that there were indeed Raikoke particles in the stratosphere from the get go. Knepp et al. give a very nice detailed example of Raikoke sulfates on 2 July, which were imbedded in a synoptic-scale SO2 plume. Here, CALIOP provides a larger context for the SAGE layer, with signals interpreted as sulfate.

There are multiple competing factors involved. One of which is a zonal coverage of large particles (>180 nm) within days of the eruption. This is not expected, so we are comfortable leaving the text as is. However, Dr. Fromm is correct that we must acknowledge the fact that sulfuric acid particles do form rapidly post-eruption as indicated by the provided references. Therefore, we updated this section (specifically point #1) to communicate this important aspect.

2 Response to Reviewer #5

We would like to thank this anonymous reviewer for providing feedback on this revised manuscript. Our responses are provided below (red) to the reviewer's comments (black).

The authors now fairly openly and critically mention the possible issues with the applicability of the method to distinguish smoke from sulfate particles and the manuscript has improved significantly in my opinion. I also want to mention that the manuscript is very well written and easy to follow.

We appreciate the reviewer's comments and the effort in reviewing this manuscript.

There is one general aspect that should be discussed explicitly in more detail (parts of it are already discussed) in my opinion: your method will not work for volcanic eruptions that lead to larger sulfate particles (this is mentioned in the paper). Your approach to deal with this is to assume that weak eruptions (like Ambae and Ulawun) with relatively small sulfur amounts injected into the stratosphere will not lead to larger sulfate particles, but larger eruptions (like Raikoke or also Pinatubo) will. While this may well be possible, it is in my opinion not well established. The realization that volcanic eruptions will not lead to an increase in particle size (or will even lead to a decrease) is a very recent one and to my knowledge it is not yet fully understood what processes or parameters determine, whether a volcanic eruption will lead to smaller or larger sulfate particles. The SO2 amount may well play a role (it probably does), but perhaps also the injection altitude (and hence temperature of the ambient air). Certainly the relative roles of nucleation of new particles and condensation onto existing particles will be important. But I would not – considering the current level of understanding – exclude the possibility of a weak eruption that also leads to larger sulfate particles. The Kelut 1990 eruption may be such an eruption (as discussed in Thomason et al. 2021), although there may be issues with volcanic ash in the weeks immediately after the eruption.

To summarize: I think it would be appropriate to add another disclaimer stating that – while it appears plausible – the SO2 amount injected into the stratosphere may not be the only parameter determining whether the sulfate particles will become larger or not.

You are correct that injection mass alone is insufficient to predict particle size. However, if particles become smaller after an eruption then the proposed classification algorithm should correctly identify these particles as sulfuric acid (the corresponding slope will be more negative than background). Regarding the 1990 Kelut eruption, you are correct that the appearance of large particles was likely due to ash. However, as Thomason et al. 2021 demonstrated the background particle load can influence the growth rate of particles significantly. This is already discussed in the manuscript, so we see little to change. However, we now explicitly state that injected SO2 mass alone is not a good predictor of resultant particle size the "Application to the Raikoke Event" section.

I have two more general, but rather minor comments:

1. When you speak of particle radius you usually use the term "mode" radius and I'm wondering, whether this is the intended term? In the standard formulation of a log-normal distribution the variable r_m or r_0 is the median radius, not the mode radius. This affects the text and also some of the Figures, e.g. Figures 1, 4, 5 and others.

We agree that this terminology can be confusing. Unfortunately the aerosol literature is highly inconsistent on naming conventions as well as assigning variable names. This is one such case. The use of "mode radius" is one such case. This term refers to the second mode of the distribution, therefore we use this terminology within this manuscript. However, we agree that this may cause unnecessary confusion and have updated the text to clarify this point.

2. The “§” sign is used frequently to refer to different sections and subsections. I think this is not standard Copernicus terminology and suggest using “section” etc. instead.

We thank the reviewer for pointing this out and we have updated the text to remove the symbol and use “Sect.”, per the Copernicus guidelines.

Specific comments:

Line 369: “The reason for the presence of elevated sulfuric acid aerosol”

We don’t know if they were present, right? Perhaps: “The reason for the potential presence ..”

Updated per the reviewer’s recommendation.

Line 50: Please provide references for the 30+ Tg of SO2 and 1 K global temperature perturbation. The latter value seems a bit large to me. Also, the injected SO2 mass is higher than most estimates I am aware of.

I think you are correct that most estimates are around 20 Tg SO2 and 0.5–1 K. The original text represented upper limits (i.e., “upwards of”), but to remove ambiguity I changed the text to represent a 20 Tg SO2 injection and a temperature change between 0.5 and 1 K. I also added references.

Line 162: “While the real component of the BrC refractive index is spectrally flat, it ranges from 1.3 to 1.9“

With “spectrally flat” you mean: no spectral dependence, right? The range from 1.3 to 1.9 does not refer to a spectral change, but to a dependence on composition? Perhaps this can be mentioned explicitly?

That is correct, no spectral dependence. We agree this is confusing and have updated the text to: “While the real component of the BrC refractive index has no spectral dependence, previous studies reported refractive indices over a relatively broad range: between 1.3 to 1.9”

Line 185: “this model is provides” -¿ “this model provides”

Updated to the reviewer’s recommendation.

Figure 1: Definition of the slope. You carry out a linear regression with wavelength on the x-axis and log(k) on the y-axis, right? Log(k) is dimensionless and therefore the units of the slope should be: 1/nm, right?

We believe you are correct. This has been updated throughout the paper.

Line 258: Your criterion 2 will automatically exclude the possibility to have larger sulfate particles after a volcanic eruption. This is of course discussed in the paper, but in my opinion this still is a weakness of the approach, because research on the change in particle size after volcanic eruptions is still ongoing and the realization that sulfate particles may become smaller after eruptions is a very recent one.

If particles become smaller then the slope will become more negative, which will force the classification to remain sulfuric acid. We interpret this comment as just a comment and see nothing to change here.

Line 282: “In theory, the proposed classification method is straight forward and is expected to be reliable for events of a single type (i.e., either volcano or wildfire, but not necessarily mixed events).“

Only if the sulfate particles do not become larger after a volcanic eruption, right? This should probably be stated here.

This is correct. The text was updated to reflect this.

Lines 332-335: The processes may be more complicated than suggested here. An important question is, whether nucleation (of new particles) of condensation (onto existing particles) is the main sink for gaseous sulfuric acid. I'm not sure there is a general answer and the answer may depend on injected SO2 mass, injection altitude (i.e. ambient temperature) etc.

We agree with the reviewer that this is poorly understood. We see no corrective action to be taken here.

Line 334: “its impact on the spectral slope was minimal due to the consistent composition and hence spectral properties“

This sentence implies that the spectral properties depend on the composition only. However, they also depend on the particle size.

This was updated to reflect the importance of the particle size distribution.

Line 341: “vide infra”

Perhaps in english? Not all readers will be familiar with this latin expression

Perhaps, but there is no harm in learning something new. We will abide by the guidance provided by Copernicus.

Line 345: “as was seen during Pinatubo.“ I don't fully understand the implication of this part of the sentence. Do you mean that the spectra were flatter after Pinatubo because of ash or because of large sulfate particles?

This is in reference to the sulfate particles. The Pinatubo reference here is superfluous so it was removed for the sake of clarity.

Table 5: This comparison of particle sizes is interesting, but should be complemented by a bit more information. It is not clear, whether this is an apples-to-apples comparison, because the assumed particle size distributions may be different. You assumed a mono-modal log-normal distribution (Fig. 1) with a width of 1.5. What was the width parameter for the Wrana retrieval? What size distribution was assumed for the ACE-FTS retrieval? Perhaps it makes sense to determine and present effective radii, too. This may allow for a better comparability of the values.

This is a fair point. Both the Wrana and ACE-FTS methods fit the SAGE and ACE-FTS data, respectively, to the best matching particle size distribution. Therefore, there is no assumption of size distribution parameters (i.e., unlike us, they do not assume the width). You are correct that width can play a role in the extinction spectrum, though generally not substantial over small changes. We now include the sigma estimates in this table for clarity.

Are you comparing your “mode” radius to the ”median” radius by Wrana? Or are you also using the “median” radius?

As stated above there is variability in how different people communicate the mode radius/median radius. It’s the same thing, but can be confusing with the statistical mode. This has been clarified in the manuscript (see above). Felix and I both use the same value, though we call it by different names.

What’s also not entirely clear: did Felix Wrana provide the retrievals or did you implement the Wrana method and carry out the retrieval yourself?

Felix Wrana provided these data. The text has been updated to make this clear.