We would like to thank the first anonymous reviewer (AR1) for providing feedback on this manuscript. Our responses are provided below (red) to AR1's comments (black).

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.

We thank the reviewer for this suggestion and we agree that a change in title may have been applicable to the original manuscript. The 4 case study events were used to test the classification scheme, which was then applied to the Raikoke event to evaluate performance during a mixed event. We believe that, with the added information present in this revised manuscript, the focus of the paper has been shifted even more toward the Raikoke event. Therefore, we believe that the current title is appropriate.

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

Changed

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 ?

This is an insightful comment and we thank the reviewer for raising it. No, this has no impact on the date/time/latitude ranges. Rather, this just removes single data points that exceed this cutoff value. This will not result in a date/time/latitude related bias.

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.

The processing levels (L1 for depolarization ratio, L2 for vertical feature mask, and L3 for ASR) as well as temporal average time for the L3 product were added to the text.

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.

A brief definition for attenuated scattering ratio is now provided as is a reference to how this is used in the CALIOP algorithm.

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).

No, the secondary plume is just starting to be seen in August. This is clarified in the text.

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.

This is incorrect. Biomass burning produces a mixture of black and brown carbon with the ratio between the two dependent on burn conditions. The statement has been corrected with additional references included.

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?

This is an important clarification so we thank the reviewer for pointing this out. The text was updated to explicitly reference each panel as well as to explicitly state that all 3 compositions (sulfuric acid, BrC, BC) were simulated. The mode radius for Fig. 3, panel (a) was stated on line 187 of the original manuscript, but is now included in the figure caption for clarity. The method of normalizing each curve is now stated in the caption. Finally, there is no reference wavelength used in the linear regression. This is simply fitting a line to the wavelengths (independent variable) and extinction coefficients (dependent), prior to normalization.

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?

Yes, we confirmed that the lines are labeled correctly and we updated this figure.

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.

There has been much confusion over the interpretation and utility of Figure 3 so we thank the reviewer for raising this issue and allowing us the opportunity to correct this. As stated in the original manuscript, there are many assumptions that go into creating this figure; therefore, this figure should not be considered representative of actual atmospheric conditions during any of the events presented herein. Rather, this figure presents a *very generalized guide* for how particles of differing composition may change our measured extinction spectrum. Using this figure we developed the hypothesis that we *might* be able to distinguish between smoke/sulfate using the slope method. This figure does not *prove* that this is possible; rather, the case study events speak to this.

The 2 smoke curves show a range of potential values that are dependent on the composition (or degree of "complete" combustion) of the smoke. The actual refractive index for smoke is highly variable as shown be Liu et al. 2015, and the refractive indices we chose provide a reasonable representation of the BrC RI boundaries in Liu et al. 2015's Fig. 4. As stated above, wildfire burns result in a mixture of BrC and BC being released into the atmosphere and the BC/BrC ratio will be highly variable depending on burn conditions. Further, the composition of BrC determines its spectral properties (i.e., refractive index), which results in a wide range of possible refractive index values (as now discussed in the revised manuscript). Of course, this is all complicated by the lack of in situ measurements of stratospheric smoke. Indeed, it would seem that there is a great measurement and modeling opportunity here that should be seized, but is outside the scope of this manuscript.

Finally, the reviewer is correct that smoke and sulfuric acid particles are not expected to have the same sizes. This is especially true when considering background sulfuric acid aerosol (radii typically ≈70-80 nm) and smoke (radii typically ≈150-200 nm). Per Figure 3 of the manuscript, if there is 10% BC in 120 nm smoke particles then there really is no way to confuse these particles with background sulfuric acid particles. We updated this section to make these points clear to the reader as well as added a new section to discuss potential misclassifications (§§5.2 and 7.4.

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.

Font size within the figure was increased and the caption was updated to indicate extinction ratios are plotted as a function of 1 um extinction.

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?

We apologize, but we do not understand the issue pointed out by AR1. The original manuscript stated that for volcanic events the extinction ratios remained either unchanged or slightly larger (including for the 520:1550 ratio) while the fire events showed extinction ratios merging to similar, smaller, values. This seems to be in agreement with the reviewer's interpretation of the figure so we see no change to be made here.

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.

We apologize, but it seems that the reviewer's comment is in agreement with what is already in the text (i.e., large eruptions like Pinatubo inject a lot of SO2 into the stratosphere, which gets converted to sulfuric acid that goes on to coagulate into large particles that yield a spectrally flat extinction spectrum).

We appreciate the reviewer pointing out the role ash may play in the spectral slope. Ash is now mentioned in this section explicitly as well as other appropriate places within the manuscript.

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.

This may be true depending on the refractive index and PSD parameters. Therefore, how "reliable" the regression is depends on these parameters as well. That said, all regressions for all case study events were conducted in the same way. Regarding the comment on Figure 3 in general, please see the updated discussion of this figure within the manuscript as well as our response to the reviewer's previous comment on the same topic.

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.

A separate figure to explain this is not needed. First, we cannot say for certain why this happens. However, using Fig. 3 as a general guide as well as applying a general understanding of particle formation it is reasonable that the decrease in slope (i.e., more negative) is because of small particle formation; indeed this is the expected behavior for particle formation. As a general guide, Fig. 3 shows that, regardless of composition, smaller particles force the spectral slope to more negative values. This comment is now included in the manuscript.

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

We appreciate the reviewer pointing out that this needs quantified. The following text was added to the manuscript: "This feature can be used to separate stratospheric smoke from the volcanic sulfate particles which are spherical and have only low particulate depolarization ratio (¡ 0.1). Prata et al. (2017) found mean particulate depolarization ratios of 0.09 and 0.05 for the sulfates from Kasatochi and Sarychev volcanoes."

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.

A reference to section 7.2 is now in the caption and an explanation is provided within the text.

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.

The original statement in the paper is correct. The reviewer is correct about the 65% and 100% misclassification for Ambae and Ulawun at 24 km and 15 km, respectively. However, the total number of spectra identified as "enhanced" were small for these altitude/events (8 and 1, respectively); therefore, these values, while large in a relative sense, do not carry significant weight into the overall statistic.

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

The caption was updated to contain a description of all panels within the figure.

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.

The reviewer only considered 1 wildfire event within this comment. The original statement in the original manuscript was in regard to the overall performance for the wildfire case study events as a whole and this is indicated in the text. The sentence remains unchanged.

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

The marker colors were changed to red and made larger. The "LB" and "R" labels were poorly visible because they sat atop other lines within the figure. The labels were moved and are now easily readable. We thank the reviewer for a suggestion that improves this figure.

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 ?

This figure shows data collected over the field of regard displayed in Fig. 13. This region was not impacted by the Raikoke eruption by this date, so this is a demonstration of the presence of smoke in the stratosphere. No change to the manuscript is required for this comment.

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

Identification of ash is outside the scope of this study. Therefore, to this point we have only considered 2 possibilities: smoke or sulfuric acid (or some combination of the 2). However, the point raised by the reviewer is good and we now mention ash here as well.

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).

This is an important detail and we thank the reviewer for raising this concern. The chance for misclassifications was briefly mentioned in the original manuscript but has been expanded in this revised version. Indeed, BrC and sulfuric acid in Fig. 3 are quite similar. However, the updated discussion of this figure should make the uncertainty in defining the difference between BrC and BC more clear. In short, we cannot differentiate between BrC and BC but differentiating between smoke that has even 10% BC in it and background sulfuric acid aerosol should be readily achievable.

L. 425, p.22: Citing altitudes of 19 and 20 km could be even more convincing.
We appreciate the reviewer's suggestion and agree. This change has been implemented.

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.
We agree with this comment. The pertinent sentence now reads: "Second, during large-scale volcanic eruptions (e.g., Pinatubo with VEI 6), the probability for misclassifying the large sulfuric acid particles, or ash, as smoke is high."

L. 58, p.3: Duplicated "has".
Corrected

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

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.
This figure has been significantly updated from the original document. The caption was updated as the reviewer requested.