Comment on amt-2021-333
Anonymous Referee #4

The authors present new means of classifying SAGE 3 iss data to separate between volcanic and smoke. The method is applied to data after the Raikoke eruption, an event that was accompanied by large wildfires in Siberia and North America. A good deal of the aerosol signal came from smoke according to the manuscript. I imagine that a layer algorithm like this could be very useful, but the authors do not present what the method is intended to be used for, and I find their method interesting, but am not yet convinced that it has enough precision. This has to do with a few issues e.g. represented in their classification. I therefore suggest a revision.

The authors did not discuss what the results of the classification method will, or could be, used for. Large wildfire smoke events may be possible to identify in periods of moderately volcanically elevated sAODs, but OMPS (UVAI, ext coeff) and Calipso (dep ratios, col ratios) already does this. Are there any advantage of using SAGE rather than other platforms?

Can you be sure that it is only wildfire smoke and not something else? Carbonaceous components have been found in volcanic aerosol. Could it not be that one of the eruptions of Raikoke contained soot and organics, or that smoke in the area of Raikoke was entrained in the volcanic cloud?

Further, I found no difference in depolarization ratio for the assumed wildfire smoke and volcanic aerosol (after Raikoke). Why does it not show up as wildfire smoke in calipso’s depolarization ratio? You show information on particle sizes, but not on other particle
properties. I think that the depolarization ratio should be shown here since it is a very strong indicator of smoke.

Section 5: In the beginning of this section, it reads that the slope was computed via linear regression. How did these regressions look and how well did they fit to the data? From the figures, e.g. Fig4, it looks like there is a large variance in the data. It is difficult for the reader to grasp this without some type of illustration of these regressions. Aren’t the widths of these distributions rather important for your classification? The standard deviations of these linear regression models could be used to distinguish between cases where the identification is more or less 100% indicative of one class, and cases where the data points are mixtures of smoke and sulfate. I think that this could be a means of telling whether the rising stratospheric aerosol after Raikoke/Ulawun/Fires are a mixture of smoke and sulfate or only smoke.

I think that the figures showing the altitude dependent slopes are really good illustrations to highlight where the different aerosol layers are located. Does this work well when separating background aerosol from low volcanic impact? In the analysis you had to divide data into altitude segments (since extinction coefficients in rising or descending air masses becomes pressure dependent). Would it be possible to normalize the data with pressure to get an altitude independent slope for each class (backgr, volc, smoke)?

Table 3: It looks to me that there are quite some misclassifications even at times and altitudes with large sample sizes. Starting with the Canadian fires 2017, 62% of the data at 14 km altitude were classified as sulfuric acid, and at the highest altitudes (23-25 km) 57-99% are misclassified as sulfuric acid. What would be the source of this sulfuric acid? I don’t know of any potential eruptions occurring in the first half of 2017. To me this indicates big issues with the assumptions used for the classification algorithm. The same issue occurs after the Australian fires 2019/2020, but only at the highest altitude shown (25 km). I would like to see how well the algorithm does above 25 km. It is evident in the SAGE 3 iss data that the smoke rose to >30 km, and some dense smoke layers in the v5.10 data lacked data below 27 km indicating too high optical depth in the line of sight to quantify the extinction. So these are not faint layers. It is difficult to interpret the classification results after Raikoke if these issues occur in the periods of known sources. After Raikoke all the highest extinction coefficients (Fig 15) were classified as smoke. I find this surprising. Does this mean that a large fraction of the AOD elevation after Raikoke was actually caused by fires?

I wonder to what degree the small difference in refractive index affects the classification. The refractive index for black carbon differs quite from that of H2SO4. However, brown carbon and sulfuric acid has rather similar values in refractive index, indication that it is difficult to separate between the two.

Why did you not include a spectral slope for ash (Fig. 3), and in what way may this impact
The numbers in Table 3 don’t add up. I did not check them all but noticed the issue at Australia @25 km altitude (0.30 + 0.60).

L325-328, regarding Fig 7&8: You claim a rapid decrease in the slopes. I see a slope changing value over several kilometers. Isn’t this an indication of mixed sources?

Regarding the figures with calypso curtains, I suggest that you add curtains of the beta-532 signal as it is difficult to understand why there is a yellow feature in Fig 7b (same in Fig 8). The volcanic layers should be visible in beta-532.

L358: You write about an aerosol layer at 19 km altitude. It is actually visible in calypso images, but it is classified/misclassified as clouds. No cirrus should be present so far (~7-8 km) above the TP (even above the ExTL).

Section 2.1: Why was the data limited to 2 km above the TP? Was it to minimize cloud interference? And why not 1 or 3 km? Are there any risk of cloud interference that may disturb the classification?

Section 2.2: The lvl3 sAOD product has strong bias in the extratropics (Kar et al. 2019). Does this have any impact on the comparison with SAGE?

Caption Figure 3: I think that you should add the word ‘normalized’ to the ylabel.

There is strong gradient in the slope in Fig. 12d at 10-11 km altitude. Could it be clouds interfering? Also, no TP was marked in Fig.7&8. Is the TP height lower than what’s shown in the graphs?

The particle size distribution evolves with time, especially in the first month or two after
eruption (or smoke injection). This should lead to increased variance in your data.