We would like to thank the second reviewer (R2, Mike Fromm) for providing feedback on this manuscript. Our responses are provided below (red) to Dr. Fromm's comments (black).

Such extraordinary conclusions require extraordinary evidence. Although K21 provide interesting SAGE III/ISS aerosol extinction patterns in the Raikoke timeframe and previously, their thesis and presentation are wholly unconvincing. It is demonstrable that a primary reliance on aerosol-extinction spectra in the visible-to-near IR realm for inferring aerosol composition is ill advised. In short, SAGE-type aerosol extinction ratio is under constrained for such purposes. This is made obvious by invoking Thomason et al. (2021), (hereafter T21) who blended SAGE II and SAGE III/ISS extinction-ratio data in an exploration of 10 stratospheric volcanic sulfate plumes. Not only did T21 treat the Raikoke-season aerosol as an exclusively sulfate composition, they showed that sulfate-plume extinction spectra occupied a range of values enveloping K21's smoke characteristics. T21's summary Figure 8 showed that volcanically perturbed sulfate .5-1.0 micron extinction ratios ranged by a factor of 3-5 among the 10 plumes they analyzed. These 10 events displayed an equally likely positive and negative transition from background to perturbed extinction ratio. Taking T21 and K21 together, it became evident to me that SAGE smoke- plume visible-NIR extinction spectra fall within the wide range of observed sulfate-plume extinction spectra. I can only conclude that I misunderstood the arguments/findings of K21 (and the earlier T21) or that the premise is fundamentally weak. If the first conclusion applies, I would recommend a thorough clarification of K21's fundamentals and reconciliation with T21. If the second conclusion applies, this work does not merit publication.

We thank Dr. Fromm for this thorough comment. However, we believe there is a misunderstanding here. First, the role of T21 was not to identify the composition of stratospheric aerosol. Rather, T21 operated under the assumption that sulfuric acid aerosol was the only aerosol source and they presented a particle nucleation and growth hypothesis that was supported by a very simple simulation. The findings of T21 have no bearing on composition, and the key results can be modified to accommodate differing aerosol compositions.

Per Dr. Fromm's comment that "These 10 events displayed an equally likely positive and negative transition from background to perturbed extinction ratio"; this is roughly true. However, this is expected behavior and is in agreement with the premise of the current manuscript (K21). The original manuscript had statements that this analysis will fail for large and ash-laden eruptions; the very type of eruptions that yielded the decreasing extinction ratios of T21. We believe that this was, perhaps, not clearly communicated in our manuscript, so we have reiterated these limitations throughout as well as now discuss the limitations with interpreting the Raikoke secondary plume.

T21 and K21 are not directly comparable because they consider different time periods and do not share a common goal or methodology (e.g., one used extinction ratios the other uses spectral slopes (the 2 are inversely related), one seeks to infer composition the other aims to understand aerosol physics immediately after an eruption). T21 focused on the time surrounding the background period and *maximum extinction coefficient* , while the K21 analysis used a wider time frame. This is important because, for example, while Kelut and Ruang showed a decreased extinction ratio when extinction was maximized (Fig. 7 of T21), the ratio increased to above-background levels shortly thereafter. Raikoke looks like it's about to follow suit, but the time series in T21 was cut off too soon. Therefore, while Kelut and Ruang showed decreased extinction ratio early on, they reversed course to look more sulfate-ish per the K21 classification criteria.

This leaves the Pinatubo event, which we cannot address. We do not see the 2 studies as being contradictory to each other. While the two are quite different in methodology and purpose, we see the two studies as complementary to each other.

Figure 1. This sets the stage by displaying 4 stratospheric aerosol layers that K21 attribute to Raikoke. If the date and latitude/longitude coordinates of the 4 profiles are accurate, 3 of the 4 layers are not Raikoke material. K21, elsewhere in the manuscript, correctly state that by the time of those profiles the Raikoke plume had not advanced to 3 of the 4 positions (eastern Atlantic to Europe). Only the profile in Figure 1d is in a location (Canada) consistent with the spreading Raikoke plume. Panels a-c show smoke layers connectable to a Canada pyroCb in mid-June 2019. This is a major concern only in that it demonstrates an internal inconsistency, introduces the reader to 3 misleading profiles, and combines sulfate and smoke profiles under a single sulfate banner. If this figure is to be retained, K21 are encouraged to re-select profiles for display and rigorously qualify them based on convincing complementary data (such as maps of Raikoke SO2).

We thank Dr. Fromm this pointing this out. This figure is now updated to include SAGE extinction coefficient profiles for a single event, a CALIOP backscatter profile, as well as TropOMI SO_2 map.

Line 95. "A unique combination of volcanic and pyroCb events occurred in 2019 when the eruption of Raikoke was preceded by pyroCb events in Canada and Russia." K21's characterization is inaccurate. There was nothing unique about 2019. PyroCbs occur every year. They were also abundant/notable in 1991, 2008, 2009, and 2011, when Pinatubo, Kasatochi, Sarychev Peak, and Nabro created massive sulfate plumes. Indeed Fromm et al. (BAMS, 2010) pointed out a significant pyroCb injection in summer 1991 that was sampled by SAGE II, and may have been a contributor to the "new mode" of aerosol particle sizes suggested by SAGE II extinction spectra that year (Thomason, 1992). K21 are correct in recognizing that the co-presence of smoke and sulfate presents a measurement-interpretation challenge—in 2019 and other years. The realization that smoke has been on multiple occasions a non-negligible neighbor of stratospheric sulfates, and T21's illustration of broad ranges in visible-NIR extinction spectra in volcanic plumes (and volcano-pyroCb blends), heightens the improbability that SAGE-like extinction spectra alone are sufficient for particle-type attribution. For these reasons, K21 are advised to invoke complementary satellite data toward a more convincing discernment between volcanic and pyroconvectively sourced plume compositions based on SAGE data.

This is a good point and we appreciate Dr. Fromm pointing this out. The wording was updated to remove the claim of "uniqueness". The four case-study events presented in this manuscript demonstrate that this methodology is capable of distinguishing between smoke and sulfuric acid aerosol when sampling a single-sourced plume. We realize this is not without limitations and we have expanded the discussion of potential misclassifications. However, Dr. Fromm's greater point here is that distinguishing between smoke and sulfuric acid aerosol in mixed events (such as was present during the Raikoke time period) is not possible. To some extent we agree with this statement. For example, if an aerosol layer contains a mixture of smoke and sulfuric acid aerosol then we agree that SAGE is not able to determine the relative fraction of each component. Indeed, the proposed classification scheme would classify this layer as either sulfuric acid or smoke. Such an "either or" classification is not without its limitations, though it still provides scientific value. While the original manuscript provided a brief discussion on this topic, the revised manuscript has added sections to discuss misclassifications as well as to aid the reader in understanding the limitations of this approach.

Section 4. In this section K21 present a theoretical approach to understanding Vis-NIR extinction spectra for absorbing and scattering media. Smoke and sulfate are distinguished solely on the basis of the spectral variation of refractive index. This is interesting and informative. However, K21 state explicitly that their theoretical construct is "...in no way intended to be representative of actual conditions." This can be seen as a major weakness in that Figure 3b shows that an effective radius disparity between brown carbon and sulfate can be as small as ≈10 nm for an equal slope of - 1.5, ≈25 nm for -1.0, and 60 nm for -0.5. One can infer from the work of T21 (their Fig. 8) that the range of effective radius for a variety of sulfate plumes is quite large, perhaps exceeding 60 nm. Given the paucity of information on stratospheric smoke-plume and sulfateplume mode radius, and the reasonable expectation of systematic differences, it seems absorption systematics might be potentially inconsequential in the case of certain "actual conditions." In short, the theoretical experiment offers only one real-life particle-population systematic when others (to wit, particle size) will likely muddle plume distinction. If this paper is to rely strongly on theoretical underpinnings, the simulations must be multi-faceted.

We disagree that this is a major weakness of the methodology because this section presents the continuity of thought we applied in this analysis (from question to hypothesis to results) and Fig. 3 could be removed entirely from the paper without changing the analysis, results, methodology, or interpretation. Indeed, even without this figure the results from the 4 case study events would stand on their own. However, we believe that despite this model being quite simple, this figure is instructive in understanding the generalized pattern of what we observe. This figure has been updated to include more realistic BrC/BC mixtures and we have updated the discussion surrounding this figure. The 2 main points of the added information is: 1. BrC has a highly variable refractive index that may be similar to sulfuric acid or it may be almost as high as black carbon; 2. when a small amount of BC (10%) is added to the BrC simulation the spectral slope changed substantially. The BrC curve in the original manuscript was intended to act as a lower-limit for smoke. Indeed, *if* a smoke layer is composed solely of BrC that has the refractive indices specified in Table 1 would be difficult to distinguish from sulfuric acid aerosol. However, if the smoke particles have a small contribution from BC and they have radii at the lower-end of the expected values for smoke, they are easily distinguishable from background sulfuric acid aerosol. This discussion was added to the manuscript.

Section 7.1. K21 state "...there were two pyroCb events in the northern hemisphere during the summer of 2019..." and cite Kloss et al. (2021), Vaughan et al. (2021) and Bachmeier (2019). Neither Kloss et al. nor Vaughan et al. provide concrete details of any pyroCb event; neither goes much farther than to claim that pyroCbs occurred in Canada and Russia. No information is provided on the massiveness of these injections, a crucial element. The Bachmeier citation refers to a blog post about a pyroCb in eastern Siberia on 30 April 2019. It is known, and can be gleaned from the Bachmeier post, that that pyroCb is highly unlikely to have been a major contributor to the stratospheric aerosol burden. Moreover, it occurred seven weeks prior to the Raikoke eruption. If this was considered a candidate for all the smoke K21 detected in 2019, it is incumbent on them to investigate that event much more deeply and quantitatively. The same goes for the Canada pyroCb event. K21 rightfully acknowledge that the pyroCb action in 2019 did not match noteworthy pyroCb events such as the ones in British Columbia, 2017, and Australia in 2019/20. These, and a few others, were quantitatively massive, long lasting, and involved stratospheric plume lofting to the altitudes of the Raikoke plume in 2019. Presumably, any pyroCb that would have made a suitable contribution to stratospheric smoke in 2019 would be easy to identify. Relying exclusively on the vague information and citations provided herein is insufficient to buttress the extraordinary claims made by K21. In truth, there were in excess of 30 boreal pyroCbs in 2019. Some occurred prior to the Raikoke eruption, and several occurred thereafter. At least three were demonstrably large enough to create traceable intercontinentally transported plumes. Evidence of one of these plumes was inadvertently demonstrated by K21 in Figure 1. Hence, as in the Pinatubo summer of 1991, it is likely that stratospheric smoke was competing with Raikoke sulfates. But as argued above, this implies an obligation to apply much greater rigor in composition determination, necessarily involving several additional complementary data sets. SAGE data alone are insufficient. K21 are encouraged to either challenge that assessment or radically bolster their data analysis.

An example of the above suggestion was demonstrated by Cameron et al. (2021), who combined profile retrievals of SO2 and aerosol extinction in an examination of several stratospheric volcanic plumes, one of which was Raikoke. By exploiting coincident volcanic-gas and aerosol profiles, they presented a firstorder confirmation of sulfate particulate matter. In the case of Raikoke, Cameron et al. demonstrated close association of SO2 and aerosol enhancements in what K21 consider the "Raikoke Primary" and "Raikoke Secondary" plumes. This of course does not rule out some minor influence of biomass-burning-generated aerosol, but it clearly shows a picture of volcanic material over the altitude/zonal/temporal range examined by K21. Presumably, if K21's assertion of smoke-dominant presence is verifiable, complementary data embodying biomass burning signatures such as carbon monoxide would aid in identifying a sulfate/smoke blend. To make their case, K21 are encouraged to leverage data sets such as ACE-FTS and Aura Microwave Limb Sounder (e.g. https://acp.copernicus.org/articles/21/16645/2021/ in addition to CALIOP (exploiting its depolarization ratio data item).

We have reconsidered the interpretation of the Raikoke "secondary plume" that was observed over the tropics. We now demonstrate that smoke was detected betwen 25°N and 52°N up to ≈20 km as observed in the ACE-FTS data. We now discuss potential reasons for misclassifying the sulfuric acid particles in the secondary Raikoke plume as smoke (at higher altitudes) and present this as a limitation of this methodology and the importance of not limiting analyses to a single dataset.

However, we did demonstrate in the original manuscript that smoke was present in the higher latitudes within days of the Raikoke eruption. Further, Ansmann et al. 2021, Ohneiser et al. 2021, Osborne et al. 2021, and Johnson et al. 2021 reported observing smoke in the UTLS from 50°N to ≈80°N around the Raikoke time period. We see these studies as supportive of the claim that smoke was present in the northern latitude's UTLS around the time of Raikoke (up to 8-10 km). The manuscript was revised to include this discussion as well as a discussion on potential misclassifications.

Finally, we believe there is possible a misinterpretation of our results. When dealing with mixed events the classification scheme operate in an "either or" manner, which is a false dichotomy. In reality, these aerosol layers have a high probability of containing both aerosol types. Therefore, when a layer is classified as smoke this does not mean that that layer consists solely of smoke. It does indicate that this layer had enough smoke in it to push the classification into the smoke regime. This was discussed above, and this discussion is included in the revised manuscript for clarity.

Table 3. Canada. The breakdown between sulfate and smoke shows that sulfates represent more than 12% of stratospheric aerosol enhancements at all altitudes, and dominate at 23-25 km. Whereas boreal pyroCbs occur every year, extratropical volcanic events of VEI=4 do not. To my knowledge, there was no 2017 boreal volcanic eruption. K21 do not cite any evidence of such an eruption. Thus, what is the rationale for assigning such an overwhelming number of SAGE observations to sulfate, especially at altitudes >22 km? Unless there is to be a claim of an unpublished, suspected volcanic eruption that year, it does not seem logical to categorize any SAGE measurements as sulfate. More logically, these are an indication of smoke that overlaps into sulfate extinction-spectra space, or simply uncertain. Please justify classifying any of the 2017 aerosol enhancements as sulfate.

We thank Dr. Fromm for pointing this out as this is a limitation of this methodology. A brief explanation of this phenomenon was in the original manuscript, but we provide a brief summary here. First, this is a simple classification scheme that depicts general patterns between smoke/volcanic events. Because the stratospheric background is primarily sulfate, this scheme is potentially biased toward sulfate identifications (i.e., it requires a enough smoke raise the extinction coefficient above the imposed statistical threshold for background conditions AND push the spectral slope over the imposed statistical threshold for background conditions). Therefore, we believe this is a conservative estimate of elevated smoke events. Second, where SAGE samples is important. It is possible that we sample through the edge of a plume wherein we observe enhanced extinction, but not enough smoke to push the slope over the statistical threshold. This would result in a false negative for identification of smoke in this example. While this was in the original manuscript, we have updated this discussion in the revised version.

Section 7.1, Lines 367-369. Indeed, it is true that there were no 2019 pyroCbs in the class of the 2017 Canada and 2019/20 Australian events. Then how to explain smoke rising to 25 km and lasting 7 months? Except for citing Kloss, Vaughan, and Bachmeier, no attempt is made to assess the magnitude of the 2019 pyroCb plumes to determine if they had the ingredients to generate such a lasting and self-lofting plume. It seems as if the authors are satisfied that the information presented in the cited works makes it self-evident. Either the 2019 pyroCbs had sufficient heft to exert the extraordinary impact reported by k21 or they didn't, in which case a novel interplay between sulfates and smoke occurred. K21 should undertake a more rigorous 2019 pyroCb survey and, depending on their finding, offer a cogent explanation for the processes that support their high-altitude, persistent smoke+sulfate anomaly.

We support the claim of identifying smoke in the mid-latitudes by including data from ACE-FTS. The objective of this manuscript is not to link these observations with specific biomass burning events, rather it is to evaluate a classification scheme for identifying smoke or sulfuric acid particles. That said, we agree that, above ≈20 km we see no support from ACE for the smoke classification. This is likely a misclassification due to large sulfuric acid particles that were present after Raikoke. We also note that we saw no support for the presence of ash within this secondary plume.

Line 436, 437. "Unfortunately comparison with CALIOP was not possible for the secondary plume." K21 have missed a strategic opportunity to fully exploit CALIOP to help inform the sparser SAGE data at low latitudes. SAGE coincidences are not necessary determine the likelihood of smoke by way of CALIOP data. CALIOP depolarization ratio data are abundant from low to high latitude and throughout the life of the Secondary plume. It would be straightforward and advisable to analyze depolarization ratio for low- and high-latitude stratospheric layers to see if there is support for smoke.

It is true that a proximate match to a SAGE profile is required to make use of CALIOP data. However, this is not just a coincidence issue. The CALIOP signal at the corresponding altitudes of the secondary plume is quite low and making a reliable depolarization measurement at these altitudes is challenging. However, as noted above, we have updated our text to both support the claim for stratospheric smoke in this time period and to indicate that we see no support for the presence of smoke, at these latitudes, above ≈20 km.

Introduction, starting at Line 46. The next two paragraphs are interesting and well composed. But how relevant is this background to the issue at hand? I encourage k21 to prune the material here to improve the focus on the volcanoes in the satellite era.

Introduction has been updated.

Line124-126. What is the benefit of interpolating to 520 nm versus just adopting either 455 or 755 nm channels for analysis? I.e. what is special about 520 nm?

There is nothing is special about the 520 channel. There is a known issue in the 520, 601, and 676 nm channels as mentioned in the manuscript. While it is reasonable to expect that this channel, in the middle of the extinction spectrum, would play a relatively minor role in the slope calculation we did not operate under that assumption. Therefore, while building the analysis code we retained the 520 nm channel and explained our method of mitigating the aforementioned problem for this channel. However, we verified that excluding this channel does not significantly change the slope or subsequent analysis. The average of the absolute values of the percent difference (with_520 - without_520 / with_520) was 0.03%, the median of the absolute values was 0.27%, and the 90th and 99th percentile of the absolute percent difference were 0.8% and 1.5%. The 520 nm channel could be dropped from the analysis without effect. This is now addressed in the manuscript.

Line 137-138. A citation is needed for the range of injection altitudes, especially "19 km."

Citation added.

Line 138; callout of Fig. 1. Please see my previous comment about Fig. 1 and modify this sentence accordingly.

Figure 1 was updated and this sentence was updated to indicate the Raikoke plume was detected within 10 days as opposed to 1 week of the eruption.

Line 288-289. "Both depolarization ratio and the VFM were used herein to corroborate the identification of sulfuric acid aerosol and smoke within the SAGE data." That was apparently not done for Fig. 1. CALIOP near coincidences are available and show the requisite depol. ratio for smoke. When reworking Fig. 1 and the attendant discussion, please include CALIOP coincidences.

Figure 1 now has a CALIOP backscatter profile and the granule is referenced. The TropOMI SO_2 product is also shown in Figure 1.

Section 4. This is where K21 introduce the idea of a calculated slope as an alternative to extinction ratio. But I could not find where they precisely defined how slope was calculated. This would be essential for readers who would like to replicate K21's method and results. Please elaborate on the slope calculation.

Section 4 is where we introduce the hypothesis that slope can be used. The first paragraph in section 5 (Detection and classification method) describes how this is calculated.

Line 434, 435. The physical process described here is confusing and unclear. Smoke was shed from what? What is the cited precedent for smoke acquiring sulfate?

This section was updated in the revised manuscript.

Response to comments within the manuscript. All line and figure references correspond to where Dr. Fromm's comment appeared in the original manuscript.

Citation added to line 140.

Line 141: no, the colon (":") is the correct mathematical symbol for a ratio

Fig. 1: The tropopause was below 10 km for these figures (now indicated in the caption) and the line breaks are where the SAGE algorithm failed to produce valid data (now specified in the text).

Line 169: updated to specify wildfires were coincident in time.

Line 201: Changed "will have" to "may have"

Table 1: You are correct. The table has been updated.

Line232: corrected to read “three”

Line 235: The bias is mitigated using the power law. We have yet to implement a permanent correction for the 520 channel within the algorithm so we explain here that the impact of any unknown residual bias in the 520 channel should be mitigated.

Table 2: Altitude column was removed and the altitude range is now specified in the captions for Table 1 and Figure 4.

Figure 4: Yes, only data collected between 14-25 km were used (now specified in the caption).

Line 253: Updated to read 2σ

Line 313: This is a common Latin term for ”see below”. Common Latin phrases are allow by Copernicus (for example: e.g., i.e., et al. ad hoc, post hoc, vide infra, vide supra, in situ, etc.)

Lines 376–379 and Figure 14: You are correct that SAGE extinction showed values comparable to background. However, the slope profile indicated the presence of smoke, which was corroborated within the CALIOP depolarization ratio and VFM curtain plots. Regardless of the interpretation of the SAGE extinction data, the CALIOP data are unambiguous in their identification of smoke. The text has been updated to emphasize this.

Line 392: Comment already addressed in 2 location above.

Line 404: Text now updated to exclude comment about lifetimes.

Line 408: We stated that that hypothesis earlier in the paper. The manuscript was updated to indicate this.

Line 423: “near constant” is something that is not changing (i.e., second derivative is near 0) whereas “flattening” is a slope that is closer to 0 (i.e., first derivative is near 0).

We apologize, but we do not understand the issue with this part of the manuscript. Above ≈1E-3 (19 km) and ≈7E-4 (at 20 km) there is a clear divergence between the smoke and sulfuric acid designations. This is the “bifurcation” we alluded to in the manuscript; this is just a statement of an observation.