We have complemented our analysis of the January 4 event and its transport into the stratosphere with daytime CALIOP curtain plots, which are now also included in the supplementary file. The analysis of the daytime images fully support the results we have presented from the nighttime curtain plots. The nighttime and daytime curtain plots together provide several vertical images of the smoke clouds per day.

See our answers to the reviews comments below.

Reviewer #1

Review of revised version of Friberg et al.

The revised version of the manuscript of Friberg et al., 2023 can almost be published as is.

It is visible that the authors put a lot of effort in the revision process of the manuscript. All concerns and questions were thoroughly addressed.

The authors convincingly argued about their two independent methods to retrieve the short decay time of 10 days and that this finding is not in contradiction with existing literature.

Thanks for improving the literature, introduction, and figures. Now all of the figures are better readable. The font size of latitude and longitude in Figs. 1 and 2 could, however, still be improved.

We have updated these with larger font size for latitude and longitude, according to the suggestion by the reviewer.

The used values of 61 sr and 49 sr for the Australian wildfire smoke appear too small for me. It is okay that you use the recommended values in Martinsson et al., 2022, but maybe you could refer to the fact that also other lidar ratios are used in literature for the same smoke plume.

The lidar ratio we use are effective lidar ratios, i.e., they are affected by multiple scattering. That makes the lidar ratios smaller. The reason is that the scattering is enhanced by the multiple scattering. This means that these two quantities are affected in opposite directions by multiple scattering, causing a cancelation in the product of them (which is used to compute the AOD). Therefore literature data on the lidar ratio should be avoided for measurements, like CALIOP, affected by multiple scattering. This is explained in Martinsson et al., 2022.

Reviewer #2

Review of revised Friberg et al., "Short and long-term stratospheric impact of smoke from the 2019/2020 Australian wildfires." (Hereafter "F23").

Reviewer: Mike Fromm

F23 are to be commended for the efforts to update and improve the material they presented. Their track-change document is very helpful to the reviewer. These changes reduce, in many ways, the areas of uncertainty and confusion that I harbored regarding the initial submission.

However, these changes were not persuasive in terms of F23's assertion that 1. the January-phase plume had an insignificant initial stratospheric component and 2. that January-phase Australia smoke was transported from the troposphere to the stratosphere. Each of these is discussed in turn, in the next section.

My overarching concern with F23 is that they posit diabatic self-lofting from the troposphere to the stratosphere as a pathway leading to a stratospheric smoke pollution event greater than the direct pyroCb pathway acknowledged as having occurred five days earlier. This is an extraordinary claim, which requires extraordinary evidence in support. Given the acknowledged circumstances (the a priori existence of a major stratospheric smoke plume), this paper must show incontrovertible evidence of a slow, diabatic intrusion to the stratosphere that is as definite as the already published material on the Australia Black Summer pyroCb event. The manuscript does not meet that challenge, in my assessment. The ANY event still calls for explorations such as this. But the complexity of the atmospheric situation is not resolved or clarified by F23's analysis to my reading. Absent that outcome, I cannot recommend this publication. Major changes are called for.

Below, we explain why atmospheric stability and cloud formation does not prevent diabatic transport of smoke from the troposphere to the stratosphere. Nevertheless, we do not claim that diabatic heating is the <u>only</u> possible explanation. It is clear in our manuscript that we argue that diabatic heating is <u>a likely explanation</u> for the addition of smoke more than one week after the PyroCb formations. This is for example evident in Section 3.8 (on Line 345 in the track changes version of the revised manuscript) where we write: "...*The increasing potential temperature over time indicates that they were subject to self-lofting by radiation heating...*".

Writing that we see indications of self-lofting shall not be seen as "...extraordinary claims...". It is rather the opposite. We present evidence to support this. For example, in Figure 8 (and Figure 11), where it is evident that additional smoke entered the stratosphere more than one week later than the PyroCb formations in January.

The reviewer points to previous estimates of smoke by Peterson et al. (2021). We argue that these data are less trustworthy than our CALIOP data since the method used in Peterson et al. leads to misclassification of tropospheric smoke as being stratospheric. Please see our discussions on this below.

We have performed extensive data analysis with a high-resolution aerosol dataset from CALIOP. CALIOP is the only satellite sensor that can retrieve the smoke layers' position relative to the TP and simultaneously quantify the amount of smoke. Please see discussions on this below.

General Concerns

Regarding point 1 above, F23's messaging is that their data and interpretation were supported by Peterson et al. (2021). But my assessment is that the data are still contradictory and the Peterson et al. results are mischaracterized. F23 downplay the January-phase stratospheric injection by citing Peterson et al.'s revelation that the December-phase injection mass was 2-8 times greater than that of the December phase. However, the January-phase injection mass was by itself on par with the 2017 Pacific Northwest pyroCb event (PNE), which was at that time unprecedented in the satellite record for stratospheric smoke perturbation. Hence it is reasonable to imagine an Australia event that consisted only of the January phase. The Peterson et al. stratospheric smoke-mass injection could logically have resulted in a lasting plume on par with PNE. Peterson et al. calculated the stratospheric smoke source term by two methods, one based solely on the UVAI. The UVAI maximum, combined with the stratospheric area of the UVAI plume, showed that the January-phase event was on par not only with PNE, but all other historical documented pyroCb events as well.

We disagree. Our results are mostly in line with those presented in Peterson et al. However, we note that our results differ in the analysis of smoke from the January 4 fires. The tropopause altitude presented in their figure of CALIOP data of smoke from the Jan 4 fires (their Figure 8) are several kilometers lower than the TP included in the CALIOP files provided by NASA. The authors refer to TP data from radiosoundings at the Wagga Wagga station. We checked the temperature radiosonde data from Wagga Wagga (see Figure 1 below) and find TP altitude of >16 km @Jan 4 and >15.5 km @Jan 5. This is 3.5-4 km higher than what Peterson et al. used when estimating the amount of smoke injected to the stratosphere by PyroCbs. Hence, upper tropospheric smoke was misclassified as stratospheric, resulting in overestimation of the smoke mass.

Furthermore, their mass estimates are largely based on UVAI. Peterson et al. used a UVAI value of 15 as marker for stratospheric smoke. This value is not a hard limit between tropospheric and stratospheric smoke, which is evident in their figures (Figure 8 in Peterson et al.) where mid-tropospheric smoke (at 3.5-6 km altitude) is apparently misclassified as stratospheric (UVAI >15).



Figure 1. Temperature soundings at the Wagga Wagga station, southeast Australia, on January 4 and 5, 2020.

The UVAI does not contain high-resolution vertical information, limiting the possibility of distinguishing its altitude relative to the TP. The UVAI values are dependent on not only altitude, but also on the particle properties, concentration, and mass. High UVAI values can therefore appear also in the mid or upper troposphere leading to misclassification of tropospheric data as stratospheric. Hence, high UVAI values cannot be viewed as hard evidence for stratospheric smoke intrusions.

In summary, the UVAI based approach used in Peterson et al. holds uncertainties and possible errors affecting the interpretation of the event. CALIOP's altitude information is reliable.

As useful and advantageous as CALIOP data are for the purpose of characterizing nascent pyroCb plumes, they are—like all other remote sensing data items--subject to the vagueries of optimal sampling. The nature of sampling and the naturally small footprint of a nascent smoke plume is that they are often missed or incompletely captured with satellite data like CALIOP. My intense experience with the study of the ANY event, no doubt the same as F23's, revealed that the January-phase smoke plume was missed by CALIOP such that ideal plume height and concentration was delayed for several days. There were missing complete orbits or orbit segments on some days, and other days where CALIOP was just unlucky. Our survey of CALIOP showed that CALIOP made some luckier samples of the nascent December plume, as shown in F23's figures. This goes to the heart of why I asked in the first review about daytime CALIOP curtains and OMPS-LP, even conceding the poorer quality of those data. F23 showed in their supplement several daytime CALIOP curtains, implying the recognition of their value toward a more complete characterization of the nascent smoke plume. The unfortunate lack of perfect CALIOP sampling should not be convolved in the interpretation of plume altitude and concentration. In the case of ANY and PNE, the UVAI-based estimates of the immediate stratospheric presence of smoke are an independent and robust marker

of the stratospheric smoke source term. In short, there is a large, built-in uncertainty to the early sampling of CALIOP, even if all the CALIOP data are utilized. Unless F23 contend that the Peterson et al. January-phase stratospheric smoke mass is largely overestimated, the Peterson et al. results stand as an indication that the 4 January pyroCb event impregnated the stratosphere on par with almost all prior pyroCb events. If the veracity of the Peterson et al. estimates are not disputed, the challenge of showing additional smoke entering the stratosphere thereafter is considerable. F23 are encouraged to consider this argument and make whatever changes are called for in their line of analysis.

We have made additional analysis including the daytime data after the Jan 4 event. This analysis fully support the results in the nighttime data. Only very minor fractions of the smoke clouds from the Jan 4 fires were directly injected into the stratosphere. Together the daytime and nighttime data provide good coverage of the smoke clouds. The movement of the smoke clouds and the CALIOP orbits ensure that different parts of the smoke clouds are surveyed on different days. It is highly unlikely that a large stratospheric injection of smoke aerosol from the Jan 4 event would be completely missed by CALIOP. Our conclusion is that only small amounts of smoke entered the stratosphere via PyroCbs. Most of the smoke from the 2nd event entered the stratosphere later. This is shown in the supplementary, where we have added daytime curtain plots, and in Figures 8 and 11 (the first ~10 days after the Jan 4 fires). The daytime curtain plots are not used to estimate the height of the individual smoke clouds since these images contain high levels of noise, in particular in the depolarizing ratio. We have also added two adjacent nighttime curtain plots not previously included in the supplementary and estimated the height for this smoke layer in these plots (Figs S33 d-g). This has resulted in an additional data point in Figure 11.

CALIOP is the only reliable satellite instrument available that can distinguish both the smoke particle's position relative to the TP and quantify the smoke AOD. Other satellite instruments with vertical resolution become saturated. Finding the top of smoke layers is not the same as quantifying the smoke impact on the stratospheric aerosol load. This is true for all sensors.

- Regarding OMPS-LP, it cannot acquire quantitative data for dense aerosol layers. It is not reliable until more than a month after the PyroCbs during the Australian fires (Dec 29, and Jan 4).
- Regarding the UVAI, it cannot acquire vertical resolution data. One can only make assumptions on UVAI values (as described above). Hence, it cannot be used as hard evidence for a smoke layer's position relative to the TP.
- Regarding the mass estimates in Peterson et al., the TP altitudes are lower in their Figure 8 than those provided by NASA and lower than the radiosounding data as described above. This must have resulted in higher mass estimates in Peterson et al. (2021). Furthermore, their Figure 8 illustrates an example of smoke from the Jan 4 fires. It shows UVAI values >15 for tropospheric smoke, i.e. misclassifying tropospheric smoke as stratospheric. Using the tropopause altitude in NASAs curtain plots leads to almost no stratospheric impact at all. This is true for all CALIOP data. We see little direct stratospheric impact after the Jan 4 fires.
- Regarding cloud top temperatures, Peterson et al. show brightness temperatures (BT), of which the lowest BTs coincide with those in the upper troposphere in the CALIOP curtain plots shown in our supplementary file. BTs in the TP region do not tell the exact cloud top positions relative to the TP due to the minima in the temperature profile in the TP region (inversion in the stratosphere).

We have performed extensive analysis on the fire events' impact on the stratosphere and studied the time duration of stratospheric impact. We have evidence in the particle depolarization ratios that large smoke layers enter the stratosphere more than one week after the PyroCb formations at

January 4 (Figure 7 and 8). We see that the stratospheric scattering ratio, extinction coefficient, and particle depolarization ratio all increase more than one week after the January 4 fires. By investigating individual smoke layers, we find evidence that stratospheric smoke layers rise into the stratosphere from the troposphere (Figure 11). It is evident in the smoke layers' tops, mid-points, and bases.

On the contrary, no one has shown that all stratospheric smoke impact from the Jan 4 fires were a result of PyroCbs. The more we investigate the data, the less probable that scenario looks.

On the second point above, F23 made it much clearer in this revision how they accounted for the separation of December and January event smoke layers. Their CALIOP survey in Figures 1, 2, and Supplementary was more complete; their connection to phase 1, phase 2, and "other" was reasonable. From those data they built the central figures 7 and 11. This made it easier to understand these two important figures. But in my assessment, it did not adequately prove a diabatic pathway from the troposphere to the stratosphere, which is their central claim. Figure 7 contains only stratospheric observations. The CALIOP data assigned either to December or January are wholly or largely tied to two isolated, contained, and circulating smoke plugs followed by Allen et al. (2020 https://doi.org/10.1175/JAS-D-20-0131.1), Kablick et al. (2020), Khaykin et al. (2020), and Schwartz et al. (2020, https://doi.org/10.1029/2020GL090831). F23 expressly tied their December-phase data to one of these "isolated" entities. They did not directly tie their January-phase CALIOP data to an isolated smoke entity, but Figure 7a maps out the path of the plume in a way that compared almost perfectly with that of a smoke-vortex element illustrated in the above-cited papers. The important point is that the morphology of both of these smoke entities is wholly achievable by quasi-Lagrangian means due to their confinement and the absence of compromising uncertainties regarding smoke decay. These two contained smoke vortices perceptibly rose diabatically, in comparison to the extravortex ANY plumes (as hinted at by F23's "other" CALIOP data points in Figure 7).

We disagree. Our central claim is that smoke was transported from the troposphere to the stratosphere more than one week later than the PyroCb formations on January 4. We write that our data <u>indicate</u> diabatic transport (see for example Section 3.8 where we write: *"...The increasing potential temperature over time indicates that they were subject to self-lofting by radiation heating..."*).

F23's expressed and incidental following of these contained smoke vortices stands in contrast to the tropospheric CALIOP data they present. No attempt was expressed to follow these CALIOP layers materially. F23 clearly outline the difficulty of characterizing tropospheric smoke morphology because of the various, dominant, ubiquitous processes such as wet deposition. Canonical application of these forces to tropospheric smoke would lead to a vertically stratified decay profile that could appear as identical to the evolution of the CALIOP tropospheric smoke observations in Figure 11. For this reason alone, the apparent slope upward in the troposphere in Figure 11 has more than one explanation, unlike the same slopes shown for the stratospheric subset.

We can think of two atmospheric characteristics: atmospheric stability and cloud formation. The first point is less restrictive in the troposphere than in the stratosphere and should thus facilitate self-lofting compared with the conditions in the stratosphere. Cloud formation is mentioned in one passage by the reviewer:

"... The fact that all of the ANY contained plume elements shown by Schwartz et al. were defined by water vapor enhancement, and that they all were based in the lowermost stratosphere, indicates a commonality, that being a direct pathway via pyroconvection. If that is a defensible statement, the

challenge for F23 is to show that slowly ascending air masses in the troposphere can deliver not only smoke through the cold-point tropopause, but water vapor plumes as well..."

The last sentence seems to suggest that the vertical wind-speed must be high for the smoke to pass the cold point tropopause. Two of the three authors of this paper are experienced cloud scientists with several articles on clouds to their names (Martinsson (in the 1990ies) and Sporre (from 2010 onwards)), and we disagree. Martinsson et al (2022) examined the water content of the PNE smoke layers. We (Martinsson et al 2022) found H2O concentrations of 7 – 14 ppmv in the smoke layers at atmospheric pressure levels lower than 110 hPa, implying a maximum H2O vapor pressure of 0.16 Pa corresponding to a few percent relative humidity (RH). If we move this dry air to the ground the H2O concentration will increase by about a factor of 10. At the same time the temperature increases. A typical ground temperature in the summer is at least 15° C. Comparing the compressed H2O vapor pressure with the saturation vapor pressure at that temperature we find a RH of the order of 0.1%, This implies that almost all the water was precipitated before the PNE pyroCbs reached the stratosphere, e.g., if the air originally held 50% RH at the ground, 99.8% of the water was precipitated on the way to the stratosphere. That did not prevent large amounts of smoke from reaching the stratosphere. The water content of the December ANYSO was within a factor of 2, implying that almost all the water was lost on the way to the stratosphere also in this case. Smoke that reaches the upper troposphere by pyroCb transport has experienced similar air mass history because the upper troposphere is almost as cold as the cold point tropopause.

Would then a slow passage of the cold point tropopause make a drastic difference compared with a fast, convective passage? Fast vertical transport means fast cooling rate and thus high production rate of condensable water. Because of a lag in the condensation rate, the maximum supersaturation of the cloud will be high compared with slowly rising air with the same aerosol content. As a result, more cloud particles can form in fast vertical transport and take part in the later formation of precipitation. The maximum supersaturation is also strongly dependent on the amount of aerosol present. Dense aerosols, like wildfire smoke, strongly limit the maximum supersaturation of a cloud because of the increased size of the water sink and hence the condensation rate becomes much higher with a low maximum supersaturation as the result. Therefore, a larger fraction of the aerosol remains as unactivated interstitial aerosol particles. The reason that wildfire smoke reaches the stratosphere in such large amounts is the extremely high aerosol concentration, whereas a "normal" Cb loses almost all its aerosol mass because of the high production rate of condensable water in the high updraught velocities is not curbed by a large water sink, making a large fraction of the aerosol available for cloud particle formation. In conclusion, the dense smoke lifted by the pyroCbs to the upper troposphere has already withstood losses of 99% of the water or so due to precipitation. The final percent or permilles of water cannot precipitate all the remaining dense smoke, only the aerosol particles with most affinity for cloud particle formation, regardless if the transport is fast or slow.

In conclusion, smoke that has reached the upper troposphere has already lost most of the water to precipitation. A large fraction of the aerosol escapes precipitation because the smoke is dense.

The difficulty of analyzing tropospheric data lies in the frequent presence of clouds. Aerosol layers below the smoke layers complicate the attenuation correction used to compute the AOD.

Regarding the slope in Figure 11, there are no indications of large immediate stratospheric impact in the entire set of CALIOP swaths. As mentioned above, CALIOP is the only satellite sensor that can adequately distinguish smoke layers' position relative to the TP.

The supplement plots by themselves cannot be used to infer upward transport. Static images 24 hours apart allow no such definitive statement. Moreover, I could not discern a systematic rise in the

tropospheric smoke from these figures. If indeed there is such a signal, it would be essential for F23 to analyze and defend that scenario.

We have now included daytime data increasing the time resolution to 12 hours. CALIOP sampled the smoke multiple times each day. None of these curtain plots show, indicate, or even suggest any <u>large direct</u> stratospheric impact of smoke from the January 4 fires. Figure 11 is a direct compilation of the CALIOP curtain plots presented in the supplementary file. The figure does in itself provide this information.

F23 should be expected to offer other potential explanations for the pattern seen in Flgure 11. Given that wide-scale tropospheric smoke pollution on par with that seen in this case is not uncommon, and other such occurrences were not followed by significant stratospheric pollution, alternate explanations should be given at least equal weight to diabatic lofting.

We disagree. This is not a random event with faint smoke layers that dissipates quickly. The horizontal extensions of these layers are 100s to 1000s of kilometers, and PyroCbs injected them to high tropospheric altitudes. It is not surprising that these dense large smoke layers are transported differently than smaller wild-fire injections into the troposphere.

It is noteworthy that the smoke vortex associated with F23's January-phase CALIOP data was tracked by Schwartz et al. by virtue of MLS stratospheric water vapor enhancements. If this smoke entity had its origin in the troposphere, it would have been subject to the cold trap at the tropopause, a limiting factor on its water vapor content. The fact that all of the ANY contained plume elements shown by Schwartz et al. were defined by water vapor enhancement, and that they all were based in the lowermost stratosphere, indicates a commonality, that being a direct pathway via pyroconvection. If that is a defensible statement, the challenge for F23 is to show that slowly ascending air masses in the troposphere can deliver not only smoke through the cold-point tropopause, but water vapor plumes as well.

Please see our answers above.

Figure 8 is composed of zonal averages. The range from 20-80S would embody tropopause heights generally above 14 km in the northern realm to lower in the southern realm. Since this analysis is done with an absolute altitude scale, it is to be expected that the data below 14 km is a blend of tropospheric and stratospheric aerosol. Since these data almost certainly represent some unknown blend of stratospheric air, it is uncertain as to how to assess these lowermost data points in relation to those above 14 km.

<u>Tropospheric data are not included in the figure</u>. The figure <u>shows stratospheric air only</u> (as indicated in the figure caption). Data below 14 km was not used for the analysis in Figure 9.

This figure shows that additional smoke entered the stratosphere more than one week after the Jan 4 fires. There is a large <u>significant</u> difference in particle depolarization ratio between smoke from the December and January fires. This shall not be neglected. Figure 8 is hard evidence. There is no sign of large immediate stratospheric impact. Instead, we observe addition of smoke in the stratosphere more than one week after the Jan 4 fires.

Both phases produced UTLS smoke with equally small depolarization ratio (depol for short). E.g., see the intense stratospheric layer in Figure S3b,c. The fact that the December phase produced such low depols at such high altitude prompts the question as to the true difference between the December and January nascent UTLS plumes' particle-shape populations. Could F23 comment on that? This CALIOP curtain is the only indication of such low depolarization ratio from smoke from the 1st fire event. We do not know why it is lower than in the remaining CALIOP curtain plots.

There is a clear separation in smoke depolarization ratios between the two fires. The particle depolarization ratios for smoke from the 1st fire increased before smoke from the 2nd event entered the stratosphere. This is shown in Figure 7 and 8.

F23 have convincingly shown that the January-phase stratospheric depol is somewhat less than the December phase. But what does that necessarily say about its origin altitude? Wouldn't we expect the photolytic process to drive both depol populations to the same eventual value? If so, what would account for the difference in the December and January aged plumes at stratospheric altitudes?

It is well known that depolarization ratios are lower for tropospheric than stratospheric smoke. A likely explanation for this is that the soot agglomerates collapse when smoke particles are exposed to water. Smoke particles from the December fires are therefore expected to be more irregularly shaped than smoke particles from the January fires. The fact that the particle depolarization for smoke from the January fires is so much lower than for smoke from the December fires is a strong indication that the smoke was processed in the troposphere before entering the stratosphere. The particle depolarization ratio may not necessarily become equal for smoke from these two fires. Smoke particles with collapsed soot agglomerates (January 4 fires) may not reach as high final particle depolarization ratios as smoke that were not processed in the troposphere before entering the stratosphere.

Peterson et al. showed that, by number, there were many more pyroCbs in the December phase that injected only to the troposphere than on 4 January. That may or may not have been evident in the CALIOP curtains shown in F23, but it is plainly evident in the full set of CALIOP curtains that there was abundant tropospheric smoke from 29 December onward. Given Peterson et al's accounting, doesn't it seem reasonable that there was much tropospheric smoke from the December phase that was in place by the time of the January phase?

We separated data using a combination of the UVAI, backscattering coefficient, and depolarization ratio. This is a reliable method to distinguish the separation of smoke layers in time and space (both vertically and horizontally). Tropospheric smoke from the December and January fires were separated in time and space enabling the classification of smoke.

Technically, F23 do not show diabatic transport from below the tropopause. They infer it. The stratospheric fraction of observations can be accepted as reflecting diabatic lofting largely on the strength of prior publications such as Khaykin et al;, Kablick et al. and Schwartz et al., as well as the several papers on the PNE pyroCb event. But the troposphere-to-stratosphere mechanism has still not been proven with observations.

It is unclear for us why the reviewer claims that transport of smoke from pyroCbs terminating in the upper troposphere to the stratosphere is unexpected, see explanations above.

We present evidence for additional cross-TP transport of smoke occurring more than one week after the PyroCb formations. The data <u>indicate</u> diabatic lofting from the troposphere. Such transport was neglected until recent studies.

No study has shown that PyroCb formation is <u>the only</u> transport path to the stratosphere for wildfire smoke. In a recent study Ohneiser et al. (2023) investigated the potential for self-lofting from the troposphere to the stratosphere. They find that dense smoke layers can rise from the troposphere

via self-lofting. Our data <u>indicate</u> that diabatic heating transported smoke from the troposphere to the stratosphere.

In terms of the flow of argumentation in F23, I found several instances where they assert their conclusion about the diabatic lofting from the troposphere prior to any detailed analysis. In this way they seemed to put the cart before the horse. It is advised that they not only bolster their analysis proving the tropospheric diabatic lofting, but also withhold any conclusions/assertions until the reader sees the proof.

We did not present self-lofting before showing the evidence for it. In the revised manuscripts (submitted in June and August), self-lofting is mentioned in the Introductions section together with citations to previous studies. Next time it is mentioned is at the end of the Discussions section (section 3.8 Smoke transport into the stratosphere). Note that all 11 figures were presented and discussed before we mentioned Self-lofting in the discussions section.

F23, on occasion, refer to the smoke-layer tops in the LMS as "minor" in relation to the bulk of the aerosol layer. This is reasonable from a descriptive perspective, but considering the strong vertical stratification of aerosol lifetime, this may be prejudicial. There is no doubt that typical pyroCb events distribute smoke throughout the troposphere up to the UTLS, leaving what may appear to be a minor portion at the topmost altitudes. But that "minor" part has a much greater potential to last than the eye-catching tropospheric parts. Moreover, sampling by CALIOP may give the strong appearance of the topmost smoke as being small in proportion to lower plumes. A telling example of this is shown in Figures 6 and 7 here: https://doi.org/10.1029/2021JD034928. It shows an early view of the PNE smoke plume that captured a small footprint of the pyroCb smoke plume. This "minor" feature represented the most consequential early indication of the smoke that polluted the stratosphere. Hence F23 are asked to reflect on the "minor" indications of the January-phase LMS smoke as early as 6 January illustrated in Figure 11.

It is clear from the CALIOP curtains that large amounts of smoke enter the stratosphere more than one week after the PyroCb event Jan 4. This was not the case for the 2017 North American fires, where dense smoke layers were observed up to two kilometers into the stratosphere on the second (Aug 14) and third day (Aug 15) after the fire.

CALIOP obits the Earth 14.5 times per day resulting in 29 times of sampling per 24 hours (14.5 night + 14.5 day data). It evidently passed over the smoke layers on multiple occasions each day. This is shown in the supplementary file. The curtain plot on Jan 6, which the reviewer refers to, shows a faint smoke layer with backscattering coefficients more than <u>one order of magnitude lower</u> than the smoke layers from the North American fires in the study that the reviewer refers to (see Figure 2 below). The CALIOP data and curtain plots show that the smoke had little direct impact on the stratosphere.

Dense layer above the TP, from the North American fires two days after PyroCb formation Faint layer at the TP, from the Australian Jan 4 fires two days after PyroCb formation



Figure 2. Illustration of smoke layers taken two days after PyroCb formation from the North American fires (left), and from the Australian January 4 fires (right). Arrows mark the dense smoke layers above the TP from the NA fires (left), and a faint smoke layer at the TP from the Jan 4 fires (right).

F23 responded to my initial review's question about exploiting CALIOP color ratio. They argued that the differential attenuation made that an insurmountable hurdle. However, they did exploit CALIOP color ratio in Martinsson et al. (2022) in a manner I was alluding to. Hence, I still wonder if one could discern systematics in the temporal evolution of the color ratio. Both the color ratio and the differential attenuation are associated with particle size. There might be qualitative as well as quantitative tactics to assess the temporal changes even while acknowledging the attenuation issue. Since this group met that challenge in Martinsson et al., they are encouraged to do the same here or explicitly address the reason why they avoid that in this work.

The 1064 nm signal is tricky to use because of high noise. The weak signal from air molecules at 1064 nm makes the method we use to estimate the lidar ratio of 532 nm difficult (impossible) to use for the longer wavelength. We tried a different method this time compared with Martinsson et al (2022) but failed, probably due to the noise problem. Rather than going back to all files and changing to the method we used in Martinsson et al (2022) we opted to skip the color ratios this time.

Katich et al. (2022, DOI: 10.1126/science.add3101) used in situ aircraft data to develop a "fingerprint" of pyroCb stratospheric smoke, in comparison to non-pyroCb smoke, in terms of an especially large coating thickness around the BC core. F23 may wish to review this paper and comment on any potential conflicts with their hypothesis of photolytic processes reducing OA material mass.

There is no conflict with our results and Katich et al (2022). Their paper indicates that smoke particle sizes did not change from 2 months (Oct 2017) to 9 months (May 2018) after the NA fires. In Martinsson et al. (2022) we reported a rapid increase in the particle depolarization ratio during the first month. Also in the present paper we see a rapid increase in particle depolarization ratio during the first month, after which the particle depolarization ratio keeps a constant value. Hence, organics are depleted during the first month. Remaining particulate matter, i.e., soot and residual organics remain in the stratosphere until transported to the troposphere or depleted by other processes.

Targeted Comments/Concerns

Below, line numbers, figure numbers, and F23 quotes are in **bold**. My reaction is in plain text.

L30: The vertical definition of the LMS is not given herein. The term is used sometimes qualitativley, but also used as part of a targeted calculation of AOD. The details of that calculation need to be provided.

We added this information to Section 2.1, where we describe how the AODs where computed.

L53-54, "At least 38 PyroCbs injected smoke to the stratosphere during two events...": Please revisit Peterson et al. and revise this statement. According to Peterson, only a subset of the 38 pyroCb-pulse injections reached the stratosphere.

Thank you for pointing this out. We have update the text in our introductions section, i.e. 20 PyroCbs entered the stratosphere according to Peterson et al.

L59-62: Even with F23's revisions, the statements in this paragraph conflate two previous published conclusions under the banner of transport from the troposphere to the stratosphere. Only Ohneiser et al. fits within this pathway. The Peterson and Khaykin papers start the plume ascent in the lowermost stratosphere. Please add the proper nuance.

We agree that Ohneiser et al. is the paper that describes this. This is well expressed in our manuscript in the second sentence on **L59-63**, where we write: "...*Most smoke encounters in the stratosphere have been explained through upward transport by pyrocumulonimbus clouds, but studies in recent years suggest that further transport mechanisms cause cross-tropopause transport of smoke. The North American wildfires in Aug 2017 showed that self-lofting by radiative heating of the dense smoke layers caused smoke to rise from the tropopause into the LMS (e.g. Khaykin et al., 2018; Peterson et al., 2018). Ohneiser et al. (2021) suggested self-lofting of smoke from the mid-troposphere as cause of extensive aerosol layers in the Arctic stratosphere in the end of 2019 and beginning of 2020...."*.

L73, "...aerosol stayed in the stratosphere for a year...(Ohneiser et al. (2022).": As far as I can tell from Ohneiser (2022), they only show PNE smoke for ~8 months. Did I miss it? If not, please provide another citation.

We agree. Thank you for pointing this out. We have changed this in the manuscript.

L125-126, "The depolarization ratios for smoke from the 2nd fire were clearly lower than those for smoke from the 1st fire...": While some systematic difference is visually apparent, there is a considerable overlap in depol between the two phases. The December phase generated some very low-depol layers above the tropopause, as did the January phase. There are several additional CALIOP curtains attributable to the December phase, not shown here, that reinforce the realization of overlap in the depol ratio between phases. In general, it appears the free tropospheric smoke depol has single-digit depol, stratospheric has decidedly double-digit depol., and tropopause-level smoke has a wide range, as manifested in both phases. Would the authors care to comment on that?

We agree that the depolarization ratio is lower for tropospheric smoke, but there is a clear <u>significant</u> difference in the particle depolarization ratio values for the December and January smoke in the stratosphere.

L125 paragraph and figures called out: It would be helpful to have marks such as arrows on the figures pointing to features the authors want to highlight to make their point.

We do not wish to make the figures more busy. The differences in depolarization ratio is clear with or without arrows.

L127: What property? F23 describe two populations of smoke depol, but not in contrast to other particle types. Please elaborate.

We have changed the sentence to "...This difference remains for more than one month, i.e., smoke layers from the 2nd fire continues to have lower depolarization ratios than smoke from the 1st fire..."

L231-232, "Peterson et al. (2021) reported much larger stratospheric impact from the 1st fire, based on studies of the fires' immediate impact.": Yes, but the stratospheric mass from Phase 2 was equivalent to PNE, according to Peterson's Figure 1. So, on its own merits, the Phase 2 plume was a major stratospheric presence.

We find that the methods used in Peterson et al. misclassifies tropospheric smoke as stratospheric. Please see answer above. Hence, the mass estimates in Peterson cannot be used as a baseline for comparison.

L197 and elsewhere: "elevation" is regularly used to characterize an increase in AOD. This term also denotes changes in altitude. It might be advisable to choose another descriptor of the AOD amplitude change.

We agree that this could be confusing and have made changes in the manuscript using the word "increase", instead of elevation.

L232-233, "10 days after the PyroCb formations we start to see more stratospheric influence (Figure. 7).": Figure 7's first January-phase data point is on 14 January, ten days post event. But Figure 2 and especially supplementary figures show January-phase CALIOP curtains dating to 5 January (1 day post event). Moreover, Figure 11 starts on 5 January. But the reader is first introduced to the January-phase smoke by the callout to Figure 7. So, it seems to be misleading to support the above statement by this figure callout. Figure 11 shows stratospheric influence from the January phase being first detected by CALIOP on or about 6 January. The weight of Figure 2, 11, and the supplementary figures indicates that a re-characterization of this sentence is called for.

With the words "...we **start to see more** stratospheric influence..." we point to increasing smoke abundance in the stratosphere.

L233: It's not clear what **"stratospheric influence"** means. Figure 7 simply follows two smoke vortices. This doesn't represent the entirety of the smoke plume. Please consider rephrasing this.

Stratospheric influence means that the smoke influences/impacts the stratosphere.

L236, "Over time, more and more smoke...": This is not obvious from the CALIOP curtains in Flg 1,2, and Supplement. This is a conclusion stated before any proof is given.

We have changed the sentence to "Over time, more and more smoke appears in the stratosphere".

L239, "…rose by at approximately the same rate as…": Again, Figure 7 simply follows two vortices, one of which was spawned by the January plume. So it is no surprise; the ascent of that vortex has already been documented by Khaylin, Kablick, and Schwartz.

We too see ascension. We do not claim that this our major finding in the manuscript.

L257-258, "Some of the smoke from the 1st event reached the UT (Figure. 1) and may have risen later along with smoke from the 2nd event contributing somewhat to the second AOD peak.": This is putting the cart before the horse. The reader has yet to see any analysis proving tropospheric lofting.

We have deleted this sentence in the updated manuscript.

L259, "Smoke from the 1st event rose markedly in the stratosphere before smoke from the 2nd event entered the stratosphere (Figure 7b).": This conclusion cannot be drawn from Fig 7. F23 started the plots on 14 January for the January plume. The various CALIOP curtains prove that there was stratospheric smoke from the Jan phase many days earlier. See also Peterson et al.

Figure 8 (and 7b) shows that additional smoke appeared in the stratosphere more than one week after the PyroCb formation, Jan 4. See also our comments with regards to the Peterson et al paper above. We added a reference to Figure 8.

L265, "...the 2nd event that ascended later into the stratosphere.": At this point the reader has not been shown proof that tropospheric smoke ascended into the stratosphere. The material that has been presented at this point does not perform that task.

We disagree. Figure 8 shows that smoke appeared more than one week after the PyroCb formation (Jan 4).

L295-297, "This explains the low depolarization ratios for smoke from the 2nd event...": See my comments about the copious observations of low-depol. tropospheric smoke in the December plume. Note also the multiple CALIOP observations of low-depol, tropopause-level smoke from both events.

There is a significant difference in the particle depolarization ratios for smoke from the 1st and 2nd event fires.

L326-327, "From the 1st event we do not see evidence of extensive crosstropopause transport beyond the initial PyroCb...": The reader does not get any information aligned with this conclusion. What analysis did F23 perform to elicit this finding?

It is evident in the supplementary file as well as in Figure 8. We added a reference to these to the sentence.

L354-355, "A continuous crosstropopause transport over the course of several weeks also affects the AOD evolution.": This pathway is taken as a given here. Cross-tropopause transport is not quantified in a manner to support the claim that it occurred over several weeks.

We changed this sentence to "...The AOD evolution (Figure 9a) suggests cross-tropopause transport over the course of several weeks..."

L356-357, "Our study indicates that smoke from the 2nd event had larger long-term impact...(Figure 9.": Nowhere, to my reading, did F23 explain how Figure 9a was constructed. Elsewhere in the paper they described tracking the two phases of smoke only out to 4 February. How did the smoke-phase distinction over several months get accomplished?

Figure 9a and b is based on stratospheric zonal means as stated in the figure legend. Tracking the smoke until February refers to analysis of individual smoke layers. Regarding how the figure was constructed, it is described in the manuscript a few lines above (Section 3.5): "...We use this minimum to separate smoke data from the two fires (dashed lines in Figure 8) to form the AOD of the two events and investigate their individual impact on the stratospheric AOD..."

We changed the figure caption to "...Smoke decay in the stratosphere. a) 8-day running mean of the background subtracted stratospheric zonal mean AOD at 20-80°S above 14 km altitude for the 1st and 2nd event, respectively. b) Daily means of background subtracted AODs for the 1st event only, and c) smoke data from individual smoke layers from the dense isolated smoke from the 1st event

(scattering ratios, SR, from CALIOP) normalized with water vapor concentrations (cH2O, from MLS). The exponential fits correspond to a smoke half-life of 10 ± 2 (b) and 10 ± 3 days (c)..."

Figure 5: Do F23 wish to opine on the secondary AOD increase in the winter months? This period is not a focal point of the paper but the feature of increased AOD is in stark contrast to the decay signal, thus potentially more consequential than the earlier AOD dip and rise on which F23 focus.

In periods with stratospheric background (no smoke or volcanic aerosol) the stratospheric AOD varies with season due to air transport within and out of the stratosphere, and due to varying TP height (the extratropical stratosphere contains more air in the winter. This is well-known and well documented in literature, and have little impact on the shape of the stratospheric AOD after the fires.

Figure 7 caption, "...smoke transport and chemical evolution...": "chemical" is not shown, only optical properties. Chemistry is inferred from simulations, which is discussed in the text. But the figure caption should describe what is shown.

We agree. We have changed this in the manuscript and write "smoke particle evolution" instead of "chemical evolution".

Figure 9 caption, "Smoke decay in the dense isolated cloud from the 1st event..": This first sentence is confusing. The figure panel a shows both events. And panel a's description is "zonal means." How are events segregated while calculating zonal means?

We agree, and changed to "Smoke decay in the stratosphere".

Figure 11. The fit lines are not described in the caption.

We have added this information to the figure caption.

Supplementary Material, "Some Caliop curtain plots were excluded from the analysis since the aerosol attenuation signals were too weak even though the UVAI indicate presence of smoke aerosol.": This is unclear. Since there is no one-to-one relation between night CALIOP and day UVAI, how did F23 determine the weakness threshold used for excluding CALIOP layers?

We realize that this text was unclear and have rewritten this section in the supplementary. It now reads: *"All CALIOP curtain plots with distinct aerosol layers were included in the analysis"*.

Technical Comments

Figure S1: Please separate the left and right columns. The continuous black background makes it difficult to see the east edge of the left column and west edge of the right column.

We agree that this is helpful and have made this change in the supplementary file.

Throughout the text, use subscript notation in "SO2."

We agree, and now use subscript notation.