We thank the reviewers Kevin Ohneiser and Michael Fromm for all constructive comments and that helped us improving our manuscript. The reviewer comments are reported in black text, after which you find responses in this blue color. Changes in the manuscript are shown by 'Trach Changes' by <u>red</u> <u>underscored text</u>.

The manuscript of Friberg et al. focuses on stratospheric impact of smoke from the 2019/2020 Australian wildfires. They define two events of smoke injection for their study and find a half-life time of the smoke of 10 days. They claim it to photochemical processing of organic aerosol. The manuscript should contain more references and a more convincing argumentation that photochemical processing played a significant role in the decrease of smoke AOD – in the current version of the manuscript it is not convincing. Below, the main concerns are listed in more detail:

Main concerns:

What lidar ratios at 532nm are used to evaluate CALIOP observations for volcanic sulfate and Australian smoke? We computed the effective lidar ratios of individual smoke layers using the methods described in Martinsson et al. (2022), where we also corrected the CALIOP data for attenuations by molecules (including ozone) and particles. The mean values used for smoke from the Dec 29 fires and Jan 4 fires are 61 sr and 49 sr, respectively. In the remaining periods, and for the background, we used 50 sr.

Please check the papers Ohneiser et al., ACP, 2020 and Ohneiser et al., ACP, 2022. These papers may serve as reference for all the satellite observations (CALIOP, OMPS, AOD).

We computed the effective lidar ratios of individual smoke layers as in Martinsson et al. (2022). In that paper we compared our CALIOP data to OMPS-LP. It showed good agreement in periods when the OMPS-LP sensor could provide quantitative data (see for example Figure 5 in Martinsson et al. (2022)). We are somewhat confused by this comment. We have all expertize required to process and analyze satellite derived data on aerosol. We have more than a decade of experience with producing in-house satellite products using CALIOP level 1 data (the least processed version provided by NASA). For example; We reported that previous studies had underestimated the climate impact of explosive volcanic eruptions by neglecting the aerosol load in the LMS (Andersson et al., 2015, Nature Communications); We studied a decade of volcanic impact on the climate by correcting CALIOP lidar data for light attenuation caused by stratospheric aerosol particles (Friberg et al., 2018, ACP).; More recently, we developed methods for handling CALIOP data of stratospheric smoke layers (Martinsson et al., 2022, ACP).

You argue that the life-halftime is 10 days because the smoke particles dissolve. How could Ohneiser et al., ACP, 2022 observe the smoke then for 1-2 years after the emission? Please comment on that.

The organic part of the aerosol (90% of the nearfield aerosol) had such short life-time. We argue that organic aerosol is susceptible to photo-oxidation (as predicted by modelling). This portion of the aerosol has a half-life of 10 days. Aerosol stripped of organics remain in the stratosphere throughout the period studied in our work, i.e. during one year. We describe this more clearly in the updated manuscript, and also cite the finding by Ohneiser et al. (2022) suggested by the reviewer.

20°-80°S mean, this is from the tropics to the polar region. What about 30°-70°S? That can be better compared with the reference lidar observations above Punta Arenas at 53°S.

We attempt to study the "total" stratospheric impact of the smoke by capturing regions impacted by smoke. This way we can study the evolution of the smoke without impact of latitudinal mixing that otherwise would have resulted in transport induced variability of the AOD. This enables the study of the AOD decay in Figure 10.

Referencing seems to be arbitrary, especially in the Introduction. A good overview: What is already available (regarding this record-breaking event) together with appropriate references would be helpful! Which gaps are left and filled by this paper?

We aim at explaining the background needed to understand the importance of our study. We moved one of the paragraphs from section 3.8 to the Introduction section, and added additional references on smoke transport to the stratosphere. We were not aware of the simulations of radiation heating and self-lofting performed by Ohneiser et al., 2023 at the time of writing. That study fits well with the topic of our paper regarding cross-TP transport of smoke, and has been added to the Introduction section.

We tried to contribute to filling (at least) two knowledge gaps, i.e. a) how the smoke was transported to the stratosphere, and b) how the smoke impacts the stratospheric aerosol load in the short- and long-term.

- a) We agree that cross-TP transport could be introduced to the reader already in the Introduction section (and not only in the discussions section). We therefore added information and references to troposphere-stratosphere transport in the Introduction section. Here we also added the findings from Ohneiser et al. (2023).
- b) We find rapid decay of fresh smoke in the first month after the 1st fire and compute the aerosols life-time, verifying our previous findings in Martinsson et al. (2022) on smoke from the Aug 2017 North-American fires. Necessary background is already explained on this matter in the Introduction section.

Specific concerns:

9 lofted We changed this according to the reviewers suggestion

12 It was so inhomogeneous, how could one determine half-life? Decay is also a function of horizontal meridional dispersion.

We determined the half-life by two methods: 1.) directly from the AOD, and 2.) by studying aerosol - water vapor composition of selected smoke layers, both leading to a half-life of 10 days. Studying the smoke half-life directly from the AOD (Method #1), we tried to encircle the entire region impacted by smoke. Here we used data from all longitudes, latitudes from 20-80 degrS, and altitude from the tropopause to 35 km. With Method #2, we studied the evolution of the aerosol relative to the water vapor composition of individual smoke layers.

Both methods lead to a half-life of 10 days. This is the same half-life as in our recent study on stratospheric smoke from the North-American fires in August 2017.

21 Source We added a reference to the review on stratospheric aerosol by Kremser et al. (2016).

22 define extratropics We changed to midlatitudes and polar regions to be more specific.

34 References We have added the references.

41 eruptions without `We changed this according to the reviewers suggestion

51-56 Aging does not just remove/decrease smoke mass. How is that linked to the findings in Ohneiser et al., 2022: 20 months measurements, slow decay or even Canada, Siberia 5-8 months residence time. What about dispersion? Please add references.

In that section we refer to the smoke evolution in the first few months after the smoke injections to the stratosphere after the Aug 2017 North-American fires based on our recent study Martinsson et al. (2022). We claim that 80-90% of the smoke is lost in the first few months. The remaining aerosol stayed in the stratosphere for much longer. We have added a sentence stating that the remaining aerosol stayed in the stratosphere for a year (Martinsson et al., 2022) or even more than one year (Ohneiser et al. 2022).

73 kilometers We changed this according to the reviewers suggestion

80 brackets away around Martinson source We changed this according to the reviewers suggestion

80 define SH We have defined it now. Thanks' for pointing this out.

104 show instead of shows We changed this according to the reviewers suggestion

170-171 AOD CALIOP: What lidar ratios are used? Background and Calbuco and Australian fires. Please compare all these also with ACP, 2020 and Ohneiser et al., ACP, 2022.

We computed the effective lidar ratios of individual smoke layers using the methods described in Martinsson et al. (2022). The mean values used for smoke from the Dec 29 fires and Jan 4 fires are 61 sr and 49 sr, respectively. In the remaining periods. For the background and for volcanic aerosol we used 50 sr.

216 Figure 2 instead of Fig 2 We changed this according to the reviewer's suggestion

226 Decreasing depolarization ratio: put it into context with the findings in Baars et al. 2019 (for example) who found decreasing depolarization ratios after stratospheric smoke injection

Our results are not in contradiction with Baars et al., (2019) since we studied different periods. We studied the smoke evolution over the first 2 months, whereas they studied the evolution from the first 2 months to the first half a year after the fires. Decreasing particle depolarization ratio is expected after the initial depletion of organics, due to mixing with the stratospheric background aerosol which is dominated by spherical particle constituents, i.e. sulfuric acid and water. This effect should be more pronounced once the initial very rapid depletion of organics has occurred, i.e. after 1-2 months in the case of these fires. Hence, our findings of an initial increase in the particle depolarization ratio does not contradict the findings by Baars et al. (2019), but add information on the smoke aerosol evolution during its first 1-2 months in the stratosphere. We have added a reference to Baars et al., (2019) in section 3.4.

Baars et al. (2019) studied smoke from the Aug 2017 North-American fires, and write that the particle depolarization in the first two months were higher than during months 3-4 and 5-6 (0.15-0.25 in Aug-Sep, 0.05-0.10 in Oct-Nov, and <0.05 in Dec-Jan).

For the Australian fires, we find increasing particle depolarization ratio for both fires during the first 4 weeks for both the Dec 29 and Jan 4 fires. After 4 weeks the particle depolarization ratios reached constant values for smoke from the Dec 29 fires. Similarly, we found increasing particle depolarization ratios for smoke layers during the first 4 weeks after the Aug 2017 North-American fires (Martinsson et al., 2022). (For the Australian Jan 4 fires we could not continue to follow the smoke for longer than one month due to faint layers).

233-244 Same as earlier comments: What about sedimentation, what about dispersion, why can the smoke be observed for such a long time, when you write that half-life is 10 days?

This question is thoroughly answered in Martinsson et al., (2022), the first three paragraphs of the Discussion section. We have considered transport out of the stratosphere, sedimentation, cloud formation, and hygroscopic growth as explanations of the decline and found that loss of material from the particles by photolysis is the plausible explanation for the decline (Martinsson et al., 2022). This is also supported by laboratory experiments (Molina et al., 2004; Sareen et al., 2013) and modeling (Hodzic et al., 2015). We added this information in Section 3.6 together with the references.

The majority of the aerosol had such short life-time. We argue that organic aerosol is susceptible to photo-oxidation (as predicted by modelling). This portion of the aerosol has a half-life of 10 days. Aerosol stripped of organics remain in the stratosphere for the entire period studied.

239-244 typically, the findings tend to a decreased BC fraction with time as the coating with organics increases with time. This is in contradiction with your findings please comment on that.

In the troposphere, organic VOCs oxidizes to less volatile species and form SOA, within hours. SOA is not thermodynamically stable and simulations indicate that it becomes photo-oxidized, depleting the organic aerosol. This process is difficult to study in the troposphere, due to dominating wet-scavenging.

273 you could add Ohneiser et al. 2023 in that context (Ohneiser, K., Ansmann, A., Witthuhn, J., Deneke, H., Chudnovsky, A., Walter, G., and Senf, F.: Self-lofting of wildfire smoke in the troposphere and stratosphere: simulations and space lidar observations, Atmos. Chem. Phys., 23, 2901–2925, https://doi.org/10.5194/acp-23-2901-2023, 2023.)

We added the following sentence to the manuscript: "...Most recently, Ohneiser (2023) computed heating and lofting rates for light-absorbing smoke layers throughout the troposphere and the lower stratosphere. Their studies indicate that smoke layers can rise from the UT to the stratosphere via radiation heating..."

282-283 Rieger et al., GRL, 2021 show the opposite. Most of the smoke went south to 70-80°S. Why should the efficient transport pathway be to the north? In your Fig. 4, also most of the smoke is located south of 60°S.

The UVAI Figure (old Figure 11) shows the first weeks after the fire events, and is used here to discuss the increases in UVAI in the days after the PyroCb events. The instrument used in Rieger et al. (OMPS-LP) does not provide quantitative data during this period of dense smoke layers. We agree that our discussion on this topic was somewhat confusing. We have changed the text to better explain how we interpret the UVAI: "...the temporal evolution in the UVAI (Fig. 10a) indicates that most of the smoke remained north of 40°S in the days following each fire event when most of the UVAI was generated..."

312 Aging works also the other way around: Condensation of gases onto the particles, therefore they can have a long lifetime.

As stated above, SOA formation occurs on short time-scales. It is an intermediate, not thermodynamically stable. Simulations indicate that it becomes photo-oxidized, depleting the organic aerosol. This process is difficult to study in the troposphere, due to dominating wet-scavenging.

365-373 These three literature entries are not included in alphabetical order Thank you for pointing this out. We have put them in alphabetical order.

Fig. 1: Too busy, no text in the figure readable, too many subfigures, no legend, no continent boundaries visible, no latitude and longitude range visible

We agree that the Figure was difficult to interpret. We have added a more extensive illustration of the separation of smoke from the two fires as a supplementary (Figure S1).

Fig. 2: X axis text missing, too many figures, too small text, use (a), (b), (c)... in the figure to be able to refer to different parts of the figure. The figure organization is very confusing. Always show the same right below each other and not all the subfigures in different sizes. Please use less subfigures if not every figure is important for the paper.

We have updated this figure based on the reviewer's suggestion. It is now less busy, with fewer CALIOP scenes and with indexing of the subfigures (a ,b ,c ...). To give a more complete picture of the smoke layers from the two fire events, and to further illustrate the differences in the smoke's depolarization ratio from the fires, we also added more than one month (December 31 to February 4) of **CALIOP curtains** of attenuated backscattering, and depolarization ratio for smoke layers (Figure S2-S43).

Fig. 3: Same as Fig. 2, too small, too confusing, too much.

We have updated this figure. It is now less busy. To give a more complete picture of the smoke layers from the two fire events, and to further illustrate the differences in the smoke's depolarization ratio from the fires, we also added CALIOP curtains of smoke layers for December 31 to February 4.

Fig. 5: What is the orange point? What about self-lofting impact?

The orange dot indicates the location and time of the 1st event. We have added this information to the figure caption. We illustrate the stratosphere with three layers that captures the "ordinary" stratospheric transport. The impact of self-lofting is shown in subfigure a), where it takes time for the smoke to enter above 470K (the deep BD-branch).

Fig. 6: What was the used Calbuco lidar ratio and Australian fire lidar ratio? 30-70°S would be better.

As mentioned above, we computed the effective lidar ratios on individual smoke layers using the methods described in Martinsson et al. (2022). The mean values for smoke from the 1st and 2nd event are 61 sr and 49 sr, respectively.

Smoke was present in the region 20-30°S. We used 20-80°S in an attempt to encircle the entire region impacted by stratospheric smoke from the Australian fires.

Fig. 7: How are these results in agreement with the only reference dataset in Ohneiser et al., ACP, 2020 and Ohneiser et al., ACP, 2022? There could be saturation effects.

We do not understand the comment on saturation effects. We corrected the CALIOP data for attenuation by molecules (including ozone) and particles using the methods described in Martinsson et al. (2022).

Fig. 10: What about using latitude subregions: 30-40°S, 40-50°S, 50-60°S, 60-70°S?

Using a narrow latitude range would conflict with the purposes of the present manuscript. We tried to capture the region impacted by the smoke. In latitude-subregion-graphs most of the variability comes from transport in and out of the latitude regions. We therefore tried to encircle the regions impacted by smoke and study the "total" stratospheric impact of the smoke.

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

Reviewer: Mike Fromm

Overview

F23 present a new satellite-observation-based analysis of the Australia New Year (ANY) 2019/20 fire/pyroCb event. Their aim is to quantify the stratospheric smoke burden and evolution, resolving the two separate contributions from the December and January phases of ANY. There are three distinctive and new contributions herein. 1. determination of smoke AOD decay via photolysis, 2. non-pyroCb-pathway stratospheric pollution by the January-phase plume, yet 3. dominant influence of the January-phase smoke on the overall ANY stratospheric perturbation. In this pursuit, F23 employ aerosol data from nadir-viewing OMPS absorbing aerosol index, limb-view OMPS-LP aerosol extinction, CALIOP backscatter, and H2O data from MLS.

This is a pursuit worthy of study and natural for ACP. The ANY case, individually and by comparison to other stratospheric smoke and volcanic events, is still imperfectly understood and the subject of varying accounts regarding transport pathway, physical evolution, composition, and radiative impact. This study intends to reduce the uncertainties in at least two important ways.

However, F23 have taken on a very complicated scenario, involving continuous smoke generation in southeast Australia in late 2019 and early 2020, punctuated by episodes of pyrocumulus (pyroCu) and pyrocumulonimbus (pyroCb) activity, in a dynamical meteorological setting. I was not convinced by F23's core new finding—that the January-phase smoke was injected only into the troposphere and didn't rise into the stratosphere until ten days post event. F23 build their case with a subset of their data items that under-samples the downstream smoke to the detriment of accurate plume-height and transport-pathway characterization. F23's analysis of aerosol-layer stratification in their illustrations is vague. In addition, their considerable reliance on OMPS UVAI maps (Figure 1) to aid in plume distinction as the smoke blends together is problematic.

We used UVAI smoke observations to indicate the geographical position of smoke in the stratosphere and upper parts of the troposphere. CALIOP observations were used to track the altitude distributions of the smoke, since the UVAI cannot tell the smoke's position in relation to the tropopause. Here, we used mostly nighttime data due to its much higher signal to noise ratio. The UVAI is daytime data, resulting in a temporal displacement. We cover this using the wind patterns at the altitudes of the smoke (as indicated by CALIOP). Furthermore, the depolarization ratios were much higher for smoke from the 1st fire compared with that of the 2nd, which is clearly shown in the supplementary and in the old Fig 2, 3, 8, and 9. Hence, misclassifying smoke would have resulted in overlapping particle depolarization ratios in Fig. 8, which we do not see. We are therefore confident in the separation of smoke from the two fire events.

F23 comes on the heels of two other papers attributing ANY's unprecedented mass of stratospheric smoke to a non-pyroCb pathway. Hirsch & Koren (2021) found no pyroCbs, only random oceanic thunderstorms to be the pathway. Magaritz-Ronen, & Raveh-Rubin (2021) concluded that pyroCbs did not suffice, and that a single Pacific-Ocean synoptic-scale cyclone on ~2 January performed the task. Now F23 attribute the preponderance of ANY stratospheric smoke to tropospheric self-lofting as the pathway while briefly dismissing the above two explanations. Given that Peterson et al. (2021) demonstrated clusters of pyroCbs in both December and January that penetrated the tropopause, and unprecedented UVAI plumes immediately following each impulse, is it F23's position that neither the pyroCb nor the cyclone nor thunderstorm pathway can explain the long-lasting stratospheric smoke observations? If so, their analysis of satellite-data, transport, and lofting needs to be much more exacting than what is presented herein.

These previous studies were based on the initial phase (first days) after these intense fires. Peterson et al, 2021 studied the PyroCbs role as transport paths to the stratosphere. Our results are mostly in agreement with their work. We also see PyroCb impact on the stratosphere after the Dec 29 event, and also after the Jan 4 event. We have made this more clear in the revised manuscript.

CALIOP shows only a small immediate impact from the Jan 4 fires. Most of the smoke clearly entered the UT, but not the stratosphere as pointed out also by Peterson et al., (2021). The UT smoke remained in the UT for a week or more. This is clearly shown in CALIOP curtain plots (see Supplement). What is also evident in these plots is a gradual transport of smoke from the UT into the stratosphere, which is illustrated in the new figures added to the revised manuscript. We cannot find evidence that large amounts of smoke were injected to the stratosphere directly by PyroCbs during the 2nd event. This is simply not in line with our observations. Instead, the data points to later transport to the stratosphere.

Smoke from the 2nd fire had lower particle depolarization ratios than did smoke from the 1st fires. This is clearly shown in the curtain plots in Figure 2 and 3, and in Figure 8 and 9 (now Figure 1, 2, 7, 8). This difference is evident already at the first smoke observations and the difference continues as the

particle depolarizations increase over time. Figure 9 (now Figure 8) and the new Figure 11 show the addition of smoke to the stratosphere more than a week later than the PyroCb formations after the 2nd event (Jan 4). This is visible in all three subfigures in Figure 9. It is more than an indication of a later smoke addition to the stratosphere.

The figures provided by the Reviewer indicate that only little smoke entered the stratosphere directly via PyroCbs from the 2nd fire, although it is difficult to interpret limb-oriented measurements in dense layers. The CALIOP curtain plot from Jan 8 show a small contribution. The OMPS-LP curtain plots are difficult to interpret due to the instrument's limited vertical resolution (~2 km), but they indicate only small contribution of smoke. Furthermore, the OMPS-LP cannot discriminate smoke from ice clouds. Hence, the smoke layers in the UT cannot be resolved with OMPS-LP. The figures provided by the reviewer do not show the UT smoke due to instrument limitations.

My assessment is that if F23 can mount a convincing argument of a diabatic troposphere-tostratosphere pathway for the January-phase plume, the balance of material could become defensible and thus merit publication. But as will be elucidated in my report, F23's analysis of the December and January ANY smoke impulses is unclearly developed and plausibly incorrect. Hence, I recommend major revisions of this manuscript after considering all the concerns I list below.

Peterson et al. (2021) showed that pyroCbs brought smoke into the stratosphere mostly Dec. 29 - 30, and to a lesser extent, Jan. 4, because the elevated tropopause in the latter case resulted in termination of most of the pyroCbs rise in the UT. Their study of the instant events, the pyroCb formations, set the foundations for the developments of the Australian fires. Our evaluation shows that the transport of smoke into the stratosphere continued long after the events studied by Peterson et al. (2021). Just as these authors we find from CALIOP that large amounts of smoke were injected into the UT from the Jan. 4 fires. We see a gradual transport of smoke into the stratosphere from strong pyroCbs that reached the UT, primarily during Jan. 4. The basic mechanism is PyroCb formation, where a delay of transport into the stratosphere is caused by the high tropopause during the Jan. 4 PyroCbs.

The new Figure 11 show that more and more smoke entered the stratosphere over the first weeks after the 2nd event. They also show that the potential temperature for these layers increased over time. The conclusion we draw from this is that radiative heating played a role in transporting smoke from the troposphere to the stratosphere. Quantifying each mechanisms impact on this additional cross-TP transport is outside the scope of this article, but we are happy to collaborate with others on the matter in future studies.

General Concerns

A major concern of mine is that F23 discounted a crucial element of the pyroCb "smokestack" phenomenon. The ANY event, like all pyroCb cases, involves the direct injection of smoke, ice, and biomass-burning gases to the top of the pyroconvective column. Like all convective exhaust, these materials disburse at column-top altitudes. These injection heights are quantified in the case of ANY and several other pyroCb events (such as Black Saturday, PNE, Chisholm, etc.) by weather-radar reflectivity and/or infrared brightness temperature image data. In the case of each, it is known that the column-top estimates are generally conservative, i.e. low biased. This has been documented (Fromm et al., 2021, https://doi.org/10.1029/2021JD034928 and references therein). Hence the source term for any such pyroCb event is reliably known to the extent that the pyroCb clouds are thus characterized. In the case of ANY, Peterson et al. (2021) provided an exhaustive accounting of all of the December- and January-phase source terms. Both the December and January phases embodied plumes that topped out in the LMS. Detection of those plumes post-pyroCb are then subject to spotty sampling by any and all satellite instruments. It is regularly the case that pyroCb-plume case studies involve these imperfect satellite data items convolved with various time lags between injection and ideal sampling. Because these fire events also involve somewhat continuous emissions throughout a wide range of vertical transport that the downstream plume picture is embodied by thick and thin smoke plumes from the surface to the top most injection heights. Incomplete/imperfect sampling of these plumes can be suggestive of a multitude of pathways and processes. In the case of ANY, we can state with certainty that the December and January pyroCbs created smoke plumes that topped out above the tropopause ambient on the active dates. . In the case of the 4 January pyroCbs, echotops exceeded 16 km (and Θ exceeding 380 K). Consistent plume heights were observable and traceable to these pyroCbs, 2-3 days post pyroCb. But even if such downstream sampling was non-existent or woefully incomplete, it wouldn't change the underlying veracity of the "smokestack" pathway and endpoint. Therefore, the challenge for identifying any other contributing pathway to the stratosphere in the case where direct injection is clearly established is thereby heightened. F23's Figure 2 and 3 appear to show confirmation that both December and January ANY smoke matches the smokestack expectation. Additional support for this end-to-end connection is provided in the supplement at the bottom of this review. F23 are encouraged to weigh this argument and evidence, and consider how it informs their approach.

PyroCbs reached the stratosphere on Jan 4-5 (we do not disagree with Peterson et al., (2021) on this). The cloud-top altitude indeed shows the maximum altitude of the cloud, but do not tell the vertical aerosol distribution. CALIOP shows that large amounts of smoke was injected to the upper troposphere. The presence of smoke in the stratosphere increases over the following week. This is

clearly shown in our figures, e.g. the new Figure 11 added to the manuscript as well as in Figure 9 (now Figure 8). The new Figure 11 show that more and more smoke entered the stratosphere over the first weeks after the 2nd fire. It also shows that the potential temperature for these layers increased over time. Our study adds information on the fate of the smoke that has been little studied in previous work.

As mentioned above, our evaluation shows that the transport of smoke into the stratosphere continued long after the events studied by Peterson et al. (2021). Just as these authors we find from CALIOP that large amounts of smoke were injected into the UT from the Jan. 4 fires. We see a gradual transport of smoke into the stratosphere from strong PyroCbs that reached the UT, primarily during Jan. 4. The basic mechanism is PyroCb formation.

Figures 1 (daily UVAI maps for ~5 weeks), 2 and 3 (ensembles of CALIOP curtains) are used to make the case that the December-phase of smoke injections immediately reached the lowermost stratosphere (LMS) but the January-phase smoke did not. As one would expect, the smoke emissions over the weeklong pyroCb event generated plumes that were straightforward to separate for a brief period, then became indistinguishable based on the UVAI alone. F23 blended the UVAI data with selected nighttime CALIOP curtains to attribute a portion to December and the balance to January. They discuss how they evaluated plume transport to connect these elements with the two phases, but that transport was only inferred via the day-to-day change in the UVAI plume positions. I could not find any additional tool for source-receptor connection such as Lagrangian trajectories. By its nature, the UVAI depends largely on AOD and altitude such that a low, dense layer's UVAI could be indistinguishable from a high, optically thin layer. It is obvious from F23's Figure 2, 3 that multiple layers, from the free troposphere to the LS, were the rule downstream of ANY. The flat color gradation used in the UVAI maps limits any meaningful discernment of low/high, dense/thin features. And as mentioned above, after the first week of January, the previously distinct UVAI plumes become inseparable. F23 recognize this and qualify their analysis with their selection of CALIOP curtains matched to the UVAI features. However, every CALIOP curtain used by F23 is from a nighttime orbit segment. These are systematically ½ day offset from the UVAI-measurement time. This might be OK if there is only one layer at stake and that layer is governed by very light winds. But given the true complexity of the smoke layering and natural wind-shear involvement, the association of the CALIOP features with the UVAI is wholly uncertain.

We used mostly night data due to its higher signal to noise ratio for this separation, and connected CALIOP to the UVAI maps using data on wind patterns. As stated above, possible misclassification

would have showed up in the particle depolarization. Instead, the depolarization ratios differ markedly. Figure 2, 3, 8, and 9 (now Figure 1, 2, 7, 8) show that the smoke was separated both in altitude and in depolarization.

F23 might have invoked other imagery-based retrievals available at night, such as IR-based CO, but they did not.

We preferred to use satellite products that we are familiar with. The 60 m vertical resolution of CALIOP (at the tropopause) is unmatched by any other instrument.

F23 might have invoked daytime CALIOP curtains, but they did not.

We used nighttime data since it has far higher signal to noise ratio and (as pointed out above) misclassification would have shown up in the particle depolarization ratios.

Hence the reader is inadequately informed about the true association of the CALIOP and UVAI features. On its merits, this complicated ANY smoke event requires a more rigorous and precise accounting between the CALIOP/UVAI and the two ANY injection phases.

We disagree. Previous satellite based studies did not perform thorough analysis on particle optical properties in the first month after these fires. We are to our knowledge the first group that performed such analysis. **The particle properties verifies that we have classified the smoke successfully.**

For defensible reasons, F23 do not employ OMPS-LP aerosol extinction data as they do CALIOP for the assessment of the nascent plume altitude. However, this may have been a missed opportunity, given the limb-view data's coincidence with the OMPS-NM UVAI. While acknowledging F23's cited concerns about OMPS-LP utility in the presence of optically thick plumes, its natural combination with the UVAI can and does allow for a confident characterization of LMS plume-top. Moreover, it will be shown that such a combination reveals a finding of the LMS position of the young January-phase plume days before F23's conclusion.

OMPS-LP shows that some smoke from the 2nd event fires entered the stratosphere on Jan 4-5, and so does CALIOP. We do not disagree on this.

However, telling where the plume-top is does not answer the question on how large impact the plume has on the stratospheric aerosol load/AOD. OMPS-LP does not show how much smoke it was since the aerosol extinction coefficients cannot be quantified, and its relatively limited vertical resolution (2 km) add to the difficulties of using its data in cases where the smoke is positioned close to the TP. F23's main analysis leading them to conclude that the January-phase pyroCb smoke was almost totally relegated to the troposphere (until it diabatically lofted across the tropopause ~13 January) exploits the CALIOP curtains shown in Figure 3 and selected point observations in Figure 8.

The zonal means in Figure 9 (now Figure 8) show evidence of additional smoke transport to the stratosphere more than a week after the Jan 4 PyroCbs. Furthermore, the new Figure 11 show that more and more smoke entered the stratosphere over the first weeks after the 2nd fire. It also shows that the potential temperature for these layers increased over time. The conclusion we draw from this is that radiative heating played a role in transporting smoke from the troposphere to the stratosphere.

Figure 3 has 12 panels, curtains from 5-10 Jan. Most panels show dense smoke straddling or above the tropopause.

The figures show weak signals from stratospheric smoke layers and optically dense tropospheric smoke layers. Yes, there were a direct impact on the stratosphere, but much more smoke lingered below the TP in the week(s) following the Jan 4 fires than what initially entered the stratosphere.

Presumably F23 do not attribute these to 4 January. But it is unclear how they interpret Figure 3. The text discussion calls out Figure 3 but does not offer any in-depth explanation of the various aerosol features. On its face then, Figure 3 seems to contradict F23's premise. A sufficiently detailed discussion, commensurate with the many Figure 3 panels and the multitude of aerosol layers therein, is essential. By the way, this recommendation also applies to Figure 2 and attendant analysis.

We understand that the discussions on Figure 2 and 3 were to brief. We have revised these figures, and added a supplementary file which curtains plots of the smoke layers, extending to Feb 4. We have also added an additional illustration (new Figure 11), showing the 2nd fire's smoke layer's position relative to the tropopause. That graph shows that more and more smoke entered the stratosphere over the first weeks after the 2nd fire.

F23 exploit CALIOP depolarization ratio in an interpretation of particle morphology. Two separate threads are presented. 1. They evaluate a temporal increase of depolarization in the stratospheric smoke, in combination with AOD decay, as evidence of photolysis-imposed loss of organic mass fraction. 2. Small depolarization of the January-phase tropospheric smoke as a sign of aging in a humid environment with a transformation to more spherical particles.

These questions are answered in the following paragraphs.

Regarding point 1, two questions arise. First, if the particles are losing mass, one might expect to see a change in CALIOP color ratio commensurate with particle-size reduction. F23 introduce CALIOP attenuated color ratio in Figures 2 and 3, so it is natural to ask if they analyzed the temporal variation of that quantity in relation to depolarization ratio. If so, those results would be to F23's advantage to report. If not, it is reasonable to suggest that F23 perform this analysis to more fully support their line of argument.

It is unfortunately not possible to use the color ratios of the entire smoke clouds. CALIOP data must be corrected for the strong attenuation of the lidar beam in the dense smoke clouds. It is not possible to use the attenuated (uncorrected) data for color ratios, since the two wavelengths become attenuated to different degree. This attenuation correction is only possible for the shorter wavelength (532 nm).

Also, Baars et al. (2019; https://doi.org/10.5194/acp-19-15183-2019) showed that the 2017 Pacific Northwest stratospheric pyroCb plume decreased in depolarization over the course of ~3 months. Might F23 cite that work and comment on the implications of such transformation? Would that be consistent with a resupply of organics or other shell material on the BC core? Is there evidence of CALIOP depolarization ratio decline after the flattening shown in Figure 8?

Of course that can be mentioned in the paper, but on the other hand this always happens. The depolarization ratio decreases as smoke mixes with background air that normally contains spherical particles, regardless of external or internal mixtures of the aerosol material. We study young smoke clouds, whereas Baars et al (2019) compared young clouds with aged, well-mixed clouds, where in the latter case the background aerosol affects the results.

The individual smoke layers became to faint to follow them for months. We do not know if a decline in particle depolarization ratios occurs after the flattening. Particle depolarization ratios are increasing over time for smoke from both the 1st and 2nd fire events, as well as after the Aug 2017 North American fires (Martinsson et al., 2022). Tropospheric SOA (secondary organic aerosol) formation occurs over time-scales of hours in the boundary layer to days in the free troposphere, suggesting that it should not impact the aerosol properties over time scales of weeks or months. The stratospheric conditions may be different though. Furthermore, our results are not in contradiction with Baars et al., (2019) since we studied different periods. We studied the smoke evolution over the first 2 months, whereas they studied the evolution from the first 2 months to the first half a year after the fires. Decreasing particle depolarization ratio is expected after the initial depletion of organics, due to mixing with the stratospheric background aerosol which is dominated by spherical particle constituents, i.e. sulfuric acid and water. This effect should be more pronounced once the initial very rapid depletion of organics has occurred, i.e. after 1-2 months in the case of these fires. Hence, our findings of an initial increase

in the particle depolarization ratio does not contradict the findings by Baars et al. (2019), but add information on the smoke aerosol evolution during its first 1-2 months in the stratosphere. We have added the Baars et al., (2019) finding of decreasing depolarization to the revised manuscript.

Regarding point 2, is it reasonable to speculate on how particles that have a collapsed, nearly spherical BC core would become less spherical during the aging in the stratosphere? What is the evolution of the depolarization ratio of the tropospheric smoke? Is there a perceptible decline in time, consistent with the proposed aging?

We have not made a special study on this subject, probably the relative humidity is an important parameter. Observations with CALIOP from this fire show constantly a lower depolarization ratio for UT smoke. We recall Tandem-DMA measurements where smoke aggregates almost instantly collapse (during the brief transport time between the two DMAs).

A more concrete concern regarding this point is that F23 generalize the ambient condition of the tropospheric January-phase smoke as "humid." While it is indisputable that the stratosphere is dryer than the troposphere, the speculation here about the ambient conditions implies (to me) that the smoke particles are far below the relatively dry UT. Is it F23's expectation that the January plume was largely lower than the UT (as several of the lower, dense layers in Figure 3 are) and thus lofted by several km before entering the stratosphere? See my earlier comment about the need for a much more precise treatment of the features in Figure 3.

The term "humid" refers to the conditions in the upper troposphere, which are more humid than the stratosphere. We have changed the sentence to address this comment. It now says *"more humid tropospheric conditions"*.

We point on the large presence of smoke in the troposphere as the source delayed transport into the stratosphere. Our investigation does not deal with the fate of each and every smoke layer in the troposphere. It is obvious from the numerous CALIOP curtains in Figs 2 and 3 (now Figure 1 and 2), and in the newly added supplementary, that the smoke in the UT has lower depolarization ratio than smoke in the stratosphere.

I was confused by the general treatment of the December and January plume discussion. It appears that F23 are dealing separately with the vortical, rising plume elements and the overall ANY stratospheric smoke. Given that the two compact plume sub-elements represent a minor contribution to the overall AOD of the ANY smoke, the discussion of the global AOD evolution of the two phases seems to be unrelated to the compact-feature evolution. Perhaps I did not catch on to the interplay between the two themes. Please point out to me what I am missing, or otherwise add some discussion material to help the reader appreciate the two seemingly independent themes.

The confinement within the vortex enabled us to follow those individual smoke layers for longer. Smoke layers outside the vortex became too faint to follow for as long (Figure 8, now Figure 7). In Figure 9 (now Figure 8) we show the zonal mean based on all CALIOP nighttime swaths. Both figures reveal a temporal evolution of the particle depolarization ratios. When computing the decay of smoke aerosol we performed one estimate using only data from the isolated cloud (Figure 10a, now Figure 9a), and one estimate based on all data (Figure 10b, zonal mean, now Figure 9b)

F23 claim that the January-phase smoke plume ascended gradually from the troposphere to the stratosphere over the period of 4-14 January. Figure 3, which chronicles the perceived 4 January smoke via CALIOP imagery terminates on 10 January. There are no additional granular CALIOP curtains filling the gap to 14 January. Presumably F23's suggested rise of the smoke to the tropopause would be discernable beyond 10 January (Figure 3). The reader should have the access to the full interpretation of the smoke evolution to be convinced of F23's evidence. The 4-day gap in this timeline stands as a barrier to this understanding. It is essential for F23 to present this entire timeline or explain how the evidence they show in Figure 3 is sufficient to secure their claim.

We have added curtain plots until Feb 4 in the supplementary file. They show the gradual increase in stratospheric smoke from the 2nd fire caused by transport of dense smoke layers across the tropopause. This is also shown in the new figure (Fig 11), where the individual smoke layer's position relative to the tropopause are illustrated.

Section 3.8, "Smoke transport to the stratosphere" is perhaps the most consequential section of this paper. Yet it is populated by weak, confusing, and misleading points. Next, these are shown in bold followed by my reaction in plain text. **"The NA wildfires in 2017 showed that self-lofting by radiative heating of the dense smoke layers caused smoke to rise from the UT into the LMS (e.g. Khaykin et al., 2018; Peterson et al., 2018)."** This is an apparent mischaracterization of these two cited papers. Neither made that argument, or even hinted at it. The diabatic lofting observations they showed all started at stratospheric altitudes. If I missed these points in the two cited papers, please point them out to the reader. We changed "UT" to "Tropopause".

"Ohneiser et al. (2021) suggested self-lofting of smoke from the midtroposphere as cause of extensive smoke layers in the Arctic stratosphere during in the end of 2019 and beginning of 2020." F23 consider this statement might balancing by citing Boone et al. 2022; https://doi.org/10.1029/2022JD036600), who categorically refuted Ohneiser's characterization of extensive stratospheric smoke at that time.

We have changed this sentence and added the findings of Boone et al. (2022). It now reads: "…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. Whether those aerosol layers consisted of sulfate or sulfate-covered smoke particles is under debate (Boone et al., 2022; Knepp et al., 2022)…"

"Hirsch and Koren (2021) argued that smoke injections to the stratosphere may have occurred in the first week of January via cross-tropopause transport by convective clouds south of the fire region (38°S), where the tropopause height is lower. However, the temporal evolution in the UVAI (Fig. 11a) indicates that most of the smoke remained north of 40°S, and only a minor portion was located south of 45°S." But Figures 1 and 2 show that there was abundant smoke south of 45S in late December and the first week of January. This comment is in no way an endorsement of Hirsch & Koren's claim; it is meant to point out that F23 show data that are in apparent direct conflict with this statement.

We do not fully understand this comment, since we have interpreted Figure 11a (now Figure 10a) similar to the reviewer. We do not see evidence of Hirsch & Koren's claim. We have added the following sentence to Section 3.8 to clarify this: "…From the 1st event we do not see evidence of extensive cross-tropopause transport beyond the initial PyroCb caused smoke injections in the CALIOP data…".

The 2nd event fires (Jan 4) positioned dense smoke layers in the mid and upper troposphere (Fig. 3). Figure 3 shows tropopause-level and/or LMS smoke in 9 of the 12 panels. As I commented above, F23 do not examine Figure 3 (or 2) in granular detail or perform an explicit feature-by-feature source attribution. The reader is given no framework on which to match the above statement with the illustrations.

The newly added supplementary file show that most of the smoke was injected below the stratosphere. These regions are the UT and the mid-troposphere, e.g. ranging from the tropopause to many kilometers below the TP. In the newly added figure (Figure 11) we illustrate the 2nd fire's smoke layers position relative to the tropopause (based on the curtain plots provided as supplement).

We see evidence of vertical transport during the following week. What is this evidence? If this is referring to Figure 3, the evidence is not at all clear to me. For the reader to see F23's evidence, a much more detailed analysis of Figure 3 is required.

This is shown by the new figure (Figure 11). Although Figure 9 (now Figure 8) already shows this. It is evident that additional smoke entered that stratosphere more than one week after the PyroCb formation (Jan 4). This impacted all parameters illustrated in Fig 9, i.e. the extinction coefficients, the scattering ratios, and also the particle depolarization ratios.

The generally low depolarization ratios (<0.10) are not indicative of cloud formation. What is the relevance of this statement? Since this is referring to Figure 3, it is apparently meant to compliment F23's argument that this is smoke from 4 January Australia. Moreover, there are several large non-cloud features in Figure 3 that have depolarization ratio >>10%. Hence, this statement requires expansion toward a full characterization of the various scenes provided.

We understand that this may be confusing. We have changed the text (section 3.8) to better describe what we mean, i.e. that those dense features in the CALIOP curtains are smoke and not clouds.

"...we suggest self-lofting by radiation heating and isentropic cross-tropopause transport as the cause of transport to the stratosphere for smoke from the 2nd event, thus following the rising trend in the stratosphere, as demonstrated in Fig. 8b, also in the upper troposphere." "We suggest" implies a considerable amount of uncertainty (as opposed to, e.g. "we find" or "we demonstrated"). Prior to this statement F23 claim to see "evidence" of self-lofting in the troposphere. So, the reader needs to know if their evidence is inconclusive, which would leave F23 in the position of just "suggesting" it. If their evidence is shown to be conclusive, then their presumed position would be that tropospheric self-lofting was demonstrated. Please explain the claimed evidence and discuss their confidence level in the interpretation.

The new figure (Figure 11) and Figure 9 (now Figure 8) show that smoke is added to the stratosphere more than one week after the PyroCb formation from the 2nd event (Jan 4). CALIOP shows no evidence of <u>large</u> immediate stratospheric impact from the 2nd fire, instead it reveals large amounts of smoke in the UT, and that more and more smoke enters the stratosphere over the first weeks after the Jan 4 fires.

This statement also mentions isentropic cross-tropopause transport as a pathway to the stratosphere. They made no prior claim of finding evidence of this for the January-phase smoke. Their only treatment of this pathway was in reference to and partial dismissal of Magaritz-Rohnen & Raveh-Rubin (2021). Hence, it is unclear what justification there is for the suggestiveness of isentropic cross-tropopause transport. Please elaborate, or consider dispensing with this suggestion.

We do not dismiss isentropic cross-TP transport as transport path for smoke to the stratosphere from the 2nd fire. CALIOP curtain plots show that more and more smoke enters the stratosphere over time, but large amounts of smoke from the 2nd fire does not enter the stratosphere as early as Magaritz-Rohnen & Raveh-Rubin (2021) suggested from their trajectory studies. We cannot completely rule out that their suggestion of isentropic cross-TP transport in the first few days after the 2nd fire event (Jan 4) caused smoke to enter the stratosphere, but our new figure (Figure 11) and the zonal means of CALIOP data (old Fig. 9: extinction coefficients, scattering ratios, and particle depolarization) tell that the majority of the smoke entered later. Furthermore, the potential temperature of the smoke layers indicates self-lofting (the new figure).

Targeted Concerns

L198-200: "The 2nd fire event occurred on January the 4th, but smoke from this event showed only little immediate stratospheric influence (Fig. 3) in line with observations by Peterson et al. (2021)." What are the observations from Peterson et al. that support this statement?

We refer to their estimate that the 1st fire event gave a larger contribution to the stratospheric aerosol load. We have changed our sentence to clarify what we mean: "…*The 2nd fire event occurred on January the 4th, but smoke from this event showed only little immediate stratospheric influence (Figure 2, S8-S13). Also Peterson et al. (2021) reported much larger stratospheric impact from the 1st fire, based on studies of the fires' immediate impact (2021)…"*

Section 2.3: In this introduction of the MLS water vapor, it is called for F23 to cite Schwartz et al. 2020; 10.1029/2020GL090831) who did an in-depth analysis of the ANY MLS H2O data.

We did not cite Schwartz et al. (2020) since the focus of our study is the aerosol.

Figure 1: How is the AI color scaled? What are the ascending orbits? Why are they shown? All the panels have grainy resolution. It is very difficult to discern all the additional layer features.

We agree that the resolution was low. We have added this information in the supplementary to better illustrate the transport of smoke in the stratosphere. The supplementary file shows the UVAI maps

together with CALIOP curtain plots of smoke from the two fire events. (The orbits shown in the supplementary are from CALIOP). UVAI: Low threshold: 0.75-1; max 50 (red); typical max in fig 15 - 20 (clear yellow).

Figure 1. I was not able to find a thorough description of how the green-line demarcation between December and January phases is justified. Please explain in a way that applies to every figure panel. We found the approximate separation line by combining the horizontal information (UVAI) with vertical information (CALIOP curtains) and wind directions. Here, we aim at separating CALIOP data from the two groups. It is worth noting that both the smoke layers altitudes and their depolarization ratios differed markedly adding robustness to this method. These differences in altitude and depolarization ratio is seen in Figure 7 (old Figure 8) and in the newly added supplementary.

Figures 2 and 3: The 532 nm attenuated backscatter panels all have the meteorology-data overlay. These are not described in the captions or main text. Moreover, many of the panels are cropped such that the isoline labels are not displayed. If the meteorology data are necessary, they should be appropriately labeled and exploited in the main text. If they are non-essential, please consider reducing clutter by showing just the lidar data.

We have modified these figures based on the comments from both reviewers. We wish to keep the isolines to make the figures more similar to the supplementary. The information is now added to the figure captions to those figures, as well as to the supplementary file.

Define "main layer" and "minor layer".

We changed these terms since they were misleading. We now refer to the two categories of smoke layers from the 1st event as 'dense isolated' and 'other'.

Figure 8b: The earliest triangle is ~10 days after the fire event. But the earliest triangle on the map is not where the phase-2 plume was on 14 Jan (10 days post event). This point appears to be close, geographically, to the position on or about 7 Jan when Khaykin et al. started following this plume element. What is the date of this earliest triangle? If it is indeed ~7 January, then the triangles in Figure 8b are offset incorrectly. If instead the x-axis is relative to the December phase alone, this should be explained.

Those data are from 10 days after the fire event (14 Jan). Please see the CALIOP curtain plot S24d. Also, we updated the figure, since the color scale did not properly match the smoke data.

L287-288. "However, the depolarization ratios during the first week of January does not indicate any frequent cloud formation connected to the smoke layers." Meteorologically, what is meant by "frequent cloud formation?" What are the smoke layers to which F23 are paying attention? The reader has no basis on which to accept this assertion. Please bolster it with a callout to and detailed analysis of Figure 3, or providing an additional figure in support.

We understand that this may be confusing. We have changed the sentence to "...do not indicate cloud formation connected to the smoke layers..."

Conclusions, L322. "Smoke was injected to the stratosphere from two events." The first event was attributed by F23 to the December pyroCbs. What was the second stratospheric injection event? If F23 maintain the claim that the January impulse was strictly tropospheric, should this sentence be amended? If the second stratospheric injection event was from the January pyroCbs, this contradicts the arguments that are the basis of F23's findings. Please clarify.

We understand that "injected" sounds dramatic and hints of PyroCbs. We therefore changed to "added" to clarify that we point to addition of smoke to the stratosphere, and not specifically to PyroCbs penetrating the tropopause. As mentioned above, we study both the stratospheric and tropospheric smoke from the 2nd event. The 2nd event PyroCb injected smoke into the upper troposphere and in part to the stratosphere. We find that the UT smoke was transported into the stratosphere over the course of weeks. This further highlights the importance of PyroCbs as smokestacks, since they can not only impact the stratosphere directly via injection of smoke the stratosphere.

Technical Points

Figure 2. "Four days CALIOP curtains...": Insert "of" before "CALIOP."

We changed the figure caption.

Figure 2 caption. "The day of the fire...": What is meant by this? Should it be "day of the pyroCb? Considering that this was a multi-day pyroCb phase, it would be advisable to revise this terminology.

We have changed the figure caption.

Figure 2 caption. "malfunction": A description of the malfunction is called for, along with a citation so the reader knows what the anomaly is.

We have changed the figure to make it less busy, according to comment by Reviewer #1. This resulted in a change in the caption too. All those curtain plots are available in the new supplementary file.

Figure 2, and throughout the document. "29 Dec.": The first phase of the ANY pyroCb event was a multi-day affair, 29-31 December. Please consider describing this phase in that way.

We describe that the event started on Dec 29, both in the abstract and in the main text, and now also in the caption to the Figure.

Figure 3 caption. "Same as figure...": Missing Figure number.

Thank you for pointing this out. We have added the number.

Figure 9a: A scaling factor for the extinction coefficient is missing.

The lidar ratio was computed based on the methods described in Martinsson et al., (2022). It has a mean value of 61 sr (1st event) and 49 sr (2nd event). For further details, please see answer to comments from Reviewer #1.

Figure 9c: "1.0" on the color bar: Should be "0.10"

Thank you for finding this. We have changed to 0.10.

3.2 Compared to volcanism: Please state what is being compared to volcanism in this section heading.

We changed to "Wildfire smoke compared to volcanism"

Supplement



Aerosol Extinction Values Retrieved at 997 nm(km⁻¹)

















532 nm Total Attenuated Backscatter, km⁻¹ sr⁻¹ UTC: 2020-01-08 23:25:16.9 to 2020-01-08 23:38:45.6 Version: 4.10 Standard Daytime



24-hr back trajectories from CALIOP lead back to prior day's AI max.

