Dear Reviewer (Dear Mike)!

We thank You for careful reading and for the critical, constructive, fruitful comments and suggestions. After considering it all, we think that our revised manuscript now is in a good shape. We fully agree with your criticism concerning our tropospheric self-lofting part (observations vs simulations). The discussion was not convincing and straight forward. We also apologize that we obviously were not clear enough in our statements that smoke self-lofting in the troposphere remains a hypothesis. You are right! We did not observe self-lofting directly.

The topic we study in our manuscript is still a very new aspect in atmospheric science. Even if results of our measurements aimed to demonstrate that self-lofting played a major role in vertical smoke transport processes in the troposphere are not comprehensive enough, our simulations, the impact and uncertainty study (Sections 3.1-3.6), and the comparison of stratospheric CALIOP Canadian and Australian smoke observations with ECRAD simulations (Section 4) events are worth to be published and shared with scientific community. No doubt. On the other hand, we extensively improved the manuscript along your comments.

One of the main motivating points for the entire study was, that Raikoke sulfate aerosol alone cannot explain the observed AOTs of 0.1-0.2 (at 550 nm, in August 2019, as discussed in Ohneiser et al., 2021). The Raikoke-related AOT (according to the emitted SO_2 mass) was of the order of <0.025 at northern latitudes. Impossible to have a Raikoke AOT of 0.05. Furthermore, pyroCb activity was low over Siberia in the summer 2019 (that was even mentioned in an e-mail by D. Peterson to us, when we asked him for his opinion about the pyroCb situation over Siberia in the summer of 2019). So, how to explain the found of massive pollution in the stratosphere, if Raikoke aerosol cannot explain it? That was the basic and main driving question for us to perform this detailed self-lofting study.

Back to the revision, as a consequence of all the shortcomings in the discussions in the submitted version, we completely removed those contents in the article that were dealing with the comparison of tropospheric observations with respective simulations. Now we present a very different discussion, based on new data and a new analysis strategy, that we believe is more convincing that tropospheric smoke self-lofting obviously occurred. This discussion is given in Section 5. We analyzed the entire CALIOP data set (day by day, scene by scene) from the Raikoke eruption on 21-22 June 2019 until the end of October 2019 to have an overlap with the MOSAiC Polarstern lidar observations that started in the end of September 2019. The focus is on smoke layers (pyroCb-related, self-lofting related) but also on features produced by Raikoke sulfate aerosol. We concentrated on the high northern latitudes >65°N. We found a number of fingerprints and arguments for self-lofting in the upper troposphere as discussed in Section 5, however, it remains a hypothesis that self-lofting really occurred. This is emphasized in the revised version several times.

Another point: New articles were published on wildfire smoke (Xian et al. 2022) and on stratospheric aerosol typing (Knepp et al., 2022, Boone et al., 2022) that forced us to consider them in our discussions. Especially the Boone et al. article contains a number of, to our opinion, misleading, incomplete, and thus incorrect conclusions on the aerosol mixture in the Arctic stratosphere so that we were forced to provide a short reply in Section 5, but also to write a comment article to Boone et al. article that we submitted to JGR on 25 October 2022.

The first part of the article (Sections 1-3), and also the discussion of comparisons of observations vs simulations for stratospheric ascending Canadian and Australian wildfire smoke layers (in Section 4) remained widely unchanged. Section 5 is, however, completely new.

Now to the item by item response:

Our responses are in BLUE.

All changes in our revised version of the manuscript is highlighted in BOLD.

Before we start, please note that the title of the article has been shortened and we added a co-author (Fabian Senf), an expert for atmospheric dynamics and modeling.

Review of Ohneiser et al. (2022), "Self-lofting of wildfire smoke in the troposphere and stratosphere caused by radiative heating: simulations vs space lidar observations" Reviewer: Mike Fromm

Ohneiser et al. explore the phenomenon of diabatic ascent of solar-radiation-absorbing smoke particles with a combination of satellite lidar- and visible-wavelength imager-based aerosol observations and modeling. The diabatic lofting in their focus is broken down into two categories: free-troposphere-to-stratosphere (hereafter "FTTS") and exclusively stratospheric. The latter of these two has been unequivocally observed in multiple case studies. The former (FTTS) has been hypothesized but not confirmed observationally.

To be more specific, in our revised version, the main goal is the ascent of smoke plumes and layers from typical injections heights of 2-6 km to the tropopause. Further stratospheric ascent is similar to pyroCb-related lofting events. PyroCb convection is usually restricted to heights below or around the tropopause, and only in the minority of cases, the smoke directly reached the lower stratosphere. Self-lofting from the tropopause towards greater heights is already well documented in many other papers (Kablick et al, Torres et al, and others, all cited in this article) and thus needs not to be investigated and highlighted in our manuscript.

It is the FTTS aspect that is the driver of this work, as revealed in the Abstract: "One of the main goals is to demonstrate that self-lofting processes can explain observed smoke lofting in the free middle and upper troposphere up to the tropopause and into the lower stratosphere without the need for pyrocumulonimbus convection." This is the logical extension of this author-group's prior publications that argued for non-pyroCb stratospheric smoke being the dominant particulate constituent in the Arctic in 2019-2020 (as opposed to Raikoke volcano sulfates, put forward in a host of additional recent publications). Hence, this manuscript attempts to test their hypothesis with observations and modelling support thereof.

Yes, after presenting all the pure simulation results (in Sections 2-3), the goal is to find convincing arguments, signatures, and fingerprints that indicate that self-lofting may have contributed to the smoke transport from the injection heights (2-6 km) up to the tropopause. This is the main goal of the article (and therefore main topic of the discussions in Section 5).

The authors' observational test includes not only the Raikoke summer of 2019 but an apparently similar scenario in August 2021 when Siberian fires also produced dense, synoptic- scale, multi-day smoke plumes. They focus on CALIOP backscatter curtains for a few selected days each year over the smoky domain. From those curtains the authors mark vertical locations representing a smoke-layer centroid that presumably represents a traceable ascent of the smoke they attribute to self-lofting. In addition to the CALIOP observations, the authors show HYSPLIT forward trajectories in association with the CALIOP plume heights over the multi-day plume evolution. Back trajectories, briefly described and unshown are also invoked to argue for diabatic ascent of the smoke layers.

This part was removed from our revised version. .

I found the entirety of the Siberia CALIOP and trajectory analysis (Section 4.4) to be confusing, arbitrary, speculative, error-prone, hence unconvincing. Below I will point out the various issues I found in greater detail. But in summary, my assessment is that the authors did not demonstrate observational evidence of diabatic self-lofting of Siberian tropospheric smoke in either 2019 or 2021. Absent that evidence, the reader has no basis on which to accept the modeling results presented herein and thus be convinced of the novel hypothesis presented in foundational work of Ohneiser et al. (2021; "O21").

We agree. And this comment motivated us to make a new effort to look at the CALIOP data again in this Raikoke summer 2019, but without trying to compare observed scenes directly with simulations. In Section 5 of our revised version, no simulations are shown.

Moreover, the authors clearly did not show incontrovertible observational proof of the FTTS diabatic self-lofting pathway required for the presumed stratospheric source term for the Arctic MOSAiC lidar-based conclusions of O21, on which Engelmann et al. (2021), Ansmann et al. (2021a), and Ohneiser et al. (2022) depended. From O21's Figure 13, one can project that the nascent, <u>tropopause-level</u> Siberian smoke resulting from FTTS transport would have an AOT of ~0.2 to 0.3 in August 2019. Layers embodying such an AOT would present as an unmistakable signal in CALIOP data. By their own analysis in Section 4.4 (Fig. 17), the Siberian smoke in 2019 had ascended only to 5 km (~5 km below the tropopause). Presumably, had there been detectable, efficient self-lofting to the tropopause, the authors certainly would have shown it. In the apparent absence of smoke FTTS proof, the authors were left to state: "Fractions of this smoke plume must have reached the tropopause and later on the lowest stratosphere." Thus, even if their observational interpretations are robust, the authors did not achieve one of their "main goals." The reader is left to rely on hypothetical modeling such as already presented in O21 and updated herein. From the Summary and Outlook section: "the large number of open parameters generates large uncertainty / sensitivity to the modeled lofting rates." Hence, we are no closer to a fire-emission/stratospheric-plume-pathway connection in boreal 2019 as before.

We agree. To our opinion, it is generally impossible to show 'incontrovertible observational proofs' in the case of slow tropospheric self-lofting processes, i.e., slow compared to these cases of explosion-like pyroCb events. As a consequence, we spent quite a long time to carefully check all CALIOP data, back and forth (mainly over the Arctic, >65°N), to find more solid, clearer, and more reasonable indications for self-lofting processes. These indications are described in Sections 5. One of the most interesting features (for us at least!) was the occurrence of a near-tropopause aerosol layer. This layer is in line with our simulations. There may be a number of other reasons that may also be used to explain such a layer, but the near-tropopause layer is predicted by the model in cases of long-lasting fire events, and that counts here for us. Nevertheless we clearly state that our argumentation is still a hypothesis, as all other explanations would also be hypotheses.

The authors' main goal was to support the contentions of O21, Engelmann et al. (2021), Ansmann et al. (2021a), and Ohneiser et al. (2022) that a boreal stratospheric smoke plume was present in 2019/2020 with AOT and persistence on par with certain volcanic injections and major pyroCb outbreaks. Their approach was to establish proof by observation in support of O21's hypothesis. My assessment is that the observational proof was not given in spite of expectations. If my assessment is accurate, the authors have strategically falsified the O21 hypothesis by looking for and not finding a suitable source term for the stratospheric smoke that O21 and associated papers proffered. This is a valid and valuable scientific conclusion. The current manuscript is thus positioned to challenge those previous conclusions. The authors are asked to consider using this current work to acknowledge the lack of observational evidence for Siberian smoke FTTS transport and the implications for O21, Engelmann et al. (2021), Ansmann et al. (2021a), Ohneiser et al. (2022), etc.

We agree. We believe that our new approach presented in Section 5 of our revised version of the manuscript is now more convincing. As mentioned already, the most important finding is the development of a diffuse tropopause aerosol layer that is fully in consistency with the self-lofting simulation study (Figure 7b), i.e., the decreasing ascent rate with height in the upper troposphere (and the minimum in the ascent rate profile at the tropopause). This leads to accumulation of smoke aerosol at the tropopause.

If my assessment is incorrect, it is incumbent on the authors to establish observational proof of the 2019 Siberian-smoke pathway to the stratosphere. The modeling work had essentially been done by O21; the modelling in this paper on its own does not serve as proof.

Here we partly disagree. The modeling work was NOT essentially done in Ohneiser et al. 2021. In that paper (as well as in Ansmann et al., 2021), we presented (some kind of) preliminary simulation results. We never presented the simulation tool itself. We were always planning to present the simulation scheme together with an extended uncertainty analysis in an independent article, i.e., in this article here. And even without a clear conclusion regarding the impact of smoke self-lofting in the troposphere, our manuscript presents so many new aspects and results from the simulation study. Even if we cannot present clear proofs, then the next generation of scientists can test our hypothesis, and work on the open questions. Otherwise, there is nothing to motivate future work in this field. Sure, others can use our work (published as ACPD version), but is that a correct way to push science forward? In this regard, we partially present the evidence and highlight what are gaps that still need to be studied in deep. All this can be found in the sections 4 and 5 and the summary section 6.

Regarding the observations and modeling the authors present on the second diabatic pathway, exclusively stratospheric, the simulations make a valuable contribution to the literature. As acknowledged by the authors, the stratospheric scenario is much cleaner and simpler to follow. Moreover, prior publications have already provided a foundation for tracking the smoke-plume features studied herein. Considering my concerns with the FTTS aspect of this manuscript and the implications for publication merit, I will focus my comments on that rather than the stratospheric part.

Thank YOU for this statement!

Major Concerns

L49, “Recently, smoke self-lofting was observed in the middle and upper troposphere (Ohneiser et al., 2021; Engelmann et al., 2021).”: It is more accurate to say that self-lofting was inferred. Only a single CALIOP curtain was displayed by O21, giving a static view of vertical aerosol placement. Inferences were made based on Lagrangian trajectories and modeling.

We agree. We re-arranged the text to avoid such an impression.

L55-57, “…Siberia in the absence of pyroCb convection as spaceborne lidar observations indicated (CALIPSO, 2022).”: How can CALIOP data be used to show there was no pyroCb action? The cited reference gives no help. In this Introduction section, it is presumed that any cited material will support the specific statement made. The reference here is a generic identifier of the CALIPSO data repository. In point of fact, there were 5 pyroCbs in the Sakha region of Siberia in July and August, 2021. PyroCb alerts and follow-up discussion in 2021 were shared in near real time on the Worldwide pyroCb Information Exchange (https://groups.io/g/pyrocb), for the specific purpose of engaging researchers across the globe and documenting individual pyroCb eruptions. The authors are asked to review the communication on this open platform and revise their description of the 2021 boreal and Siberia pyroCb occurrence.

Thank You for these hints! We checked the communication and the different data sources. It remains to say, that such big pyroCb events as observed on 12 August 2017 (Canada, British Columbia) or in the beginning of 2020 (Australian fires) did not occur over Siberia in July and August 2019. And that was also mentioned in the paper of Knepp et al. (2022). Furthermore, the observed very low depolarization ratios support the absence of pyroCbs. In all cases with pyroCb-related smoke, the depolarization ratio was clearly enhanced (Canadian smoke, Australian smoke). Even in June and the beginning of July 2019, several pyroCb-related smoke layers were detected by CALIOP (probably formed over the North American continent), enhanced depolarization ratios were observed, indicating smoke lofted by pyroCbs.

L57, "Part of the smoke again entered the stratosphere in August 2021.": No doubt. One or more of the reported pyroCbs (see above) naturally injected smoke to or across the tropopause. Moreover, many other boreal pyroCbs were catalogued in 2021 (see the above link to the reporting medium). It is inappropriate for the Introduction to this paper to combine the claim of no pyroCbs and the supposed self-lofting from Siberia to the stratosphere.

We agree, and kept this in mind, when re-arranging the text. In 2021 (but also for 2019), there are contributions from both pyroCb-related lofting and smoke self-lofting.

L59-65, Discussion of smoke heating, buoyancy, self-lofting: It is important to treat the arguments of Boers et al., de Laat et al., Torres et al., and Yu et al. separately. Torres et al. and Yu et al. dealt explicitly with a puroCb-generated source term at the tropopause and primarily stratospheric transport whereas Boers et al. and de Laat et al. considered lofting of non- pyroCb-generated smoke in the lower to middle troposphere (analogous to this manuscript's Siberia hypothesis). The distinction I call for is because stratospheric self lofting has been well characterized observationally (to which the Kablick, Khaykin, and other citations in this paragraph attest) whereas the FTTS pathway has not. It also must be noted that the Boers/de Laat pathway on Australia's Black Saturday has been reinterpreted as a classic pyroCb event, with rapid injection to the tropopause (Fromm et al., 2021, https://doi.org/10.1029/2021JD034928). Please revise the discussion here to reflect the disparate levels of certainty regarding pyroCb vs. non-pyroCb smoke pathways.

We agree (and thank you for the details!). We re-arranged the text accordingly, and separated the approach of Boers et al. and deLaat et al. from the one presented in the articles of Torres et al. and Yu et al. (see Section 1).

L199, "Chemical aging is assumed to lead to a spherical shape…": On what basis is this assumed? Please defend this statement. There is abundant literature showing that aged tropospheric smoke particles embody depol. ratios that are significantly greater than the limiting value for uniform spheres, e.g. Dahlkötter et al (2014; doi:10.5194/acp-14-6111-2014), Burton et al. (2015; doi:10.5194/acp-15-13453-2015), Hu et al. (2022; https://doi.org/10.5194/acp-22-5399-2022), Liu et al. (2022, https://doi.org/10.1016/j.jqsrt.2022.108080). Hence, Table 2's "2%" "spherical" characterization is apparently an inaccurate simplification of non-pyroCb, tropospheric smoke.

First of all, we removed Table 2.

As requested we extend the explanations on this (in Section 3), i.e., regarding particle aging, aging periods, and the consequences for particle shape and observed depolarization ratios. However, as experimentally working lidar scientists, we must clearly say: If we measure depolarization ratios of 2-3% then the particles MUST be spherical. Even small deviations from the spherical form will already introduce a significant jump in the depolarization ratio. Furthermore, real world lidar observations can never deliver 0% depolarization ratio which should theoretically be measured in the case of ideal spheres and in total absence of any multiple scattering effect. There is always signal noise and the remaining uncertainties in all the channel calibration efforts make it impossible to have at the end 0% for the particle depolarization ratio. Rayleigh scattering already introduces 1-2% depolarization (that must be corrected).

Please revise to reflect the literature. Since the weight of the above-mentioned papers rests on the idea that aged smoke is better represented by an aspherical model than monolithic spheres, the implications of changing Table 2 are momentous. The authors are asked to reconsider their assumptions for aged tropospheric smoke and the implications for the optical properties they observed in the stratosphere during MOSAiC.

We disagree. If aspherical particles are present, then they cannot be aged, they are fresh particles. These fresh particles, producing enhanced depolarization ratios, had not enough time to age and to develop a compact spherical shape. We discussed all this already in many papers (Haarig et al. 2018, Ohneiser et al., 2020, Ansmann et al. 2021, review on smoke optical and microphysical properties in ACP). With time, all smoke particles will end up as spheres when the aging process is completed (after days in the humid troposphere, or weeks to months in the dry upper troposphere and especially in the stratosphere, as Baars et al. (2019) showed). We think the improved discussion in Section 3 will help to better understand these differences of aged and fresh smoke particles and the consequences for measured depolarization ratios.

Regarding the concern above regarding the authors' assumption of spherical, aged smoke in Siberia (and eventually in the stratosphere), it is important to note that the only other published work describing non-pyroCb, FTTS transport is de Laat et al., (2012). Even if it is maintained that their "solar escalator" was an accurate depiction, the resultant stratospheric smoke from Black Saturday had depolarization ratios weeks after onset so great that the CALIOP observations thereof triggered a feature classification of "ice" (https://acp.copernicus.org/preprints/acp-2021-117/acp-2021-117-RR1.pdf). Hence these aged smokes were wholly inconsistent with the assumptions shown in Table 2. The authors are asked to comment on this inconsistency and make the appropriate revisions to their current assumptions on smoke aging.

We avoided to discuss the deLaat results in this lofting paper, to keep the discussions as short as possible. And we removed Table 2. Regarding the Black Saturday smoke: The depolarization ratio for the aerosol above 20 km is clearly enhanced. That means the particles were non-spherical. If self-lofting in the free troposphere was involved then the upper troposphere must have been super dry and condensable gases were not available so that particles could not age quickly and could not develop a compact and spherical. To our opinion, the detected Black Saturday smoke above 20 km, showing enhanced depolarization, was lofted by pyroCbs.

Section 4.4. This section is problematic. My concerns are many and deep. This part of the paper is seriously flawed. Below I list the issues individually.

We totally agree with you, it was our fault to present it. We apologize for that. We removed the content of Section 4.4. Our revised version now contains a new section 5 with a discussion on potentially observed fingerprints for tropospheric self-lofting of smoke over central eastern Siberia and the adjacent Arctic in the summer of 2019 based on CALIOP data (June to October 2019).

L482, "The smoke at different heights was transported in the same air column.": Meteorologically, I cannot make sense of this. What does "same air column" mean? The trajectories take different paths and have very different endpoints. This implies considerable speed and directional shear, which by itself seems to suggest a variable air column. Please explain.

As mentioned, all this is removed now.

Discussion of Figures 17 and 18: How “old” are the smoke layers on 12 August 2019 and 4 August 2021? Without knowing that, the reader has no idea how all those smokes got to that start point. Almost assuredly they didn’t all arrive in a single “air column,” but no information is given. I.e. these seem like arbitrary start points and hence it is unclear how to assess transport after the start points in relation to transport to them.

As mentioned, all this is removed now.

L482, 483, “The trajectories analysis in Fig. 18a indicated similar wind speed and directions at different heights in the middle troposphere.”: I cannot reconcile Fig. 18a with this description. There are significant differences in both aspects. If the authors want to convince the reader that these trajectories are similar, they need to quantify that and show what dissimilar trajectories would look like.

It was excluded from our revised version.

L483, 484, “The smoke slowly ascended from around 3-4 km height to 5-6 km height.”: The trajectories themselves account for almost a 1-km altitude gain. I.e. resolvable meteorology is responsible, not diabatic lofting. The authors need to acknowledge this.

As mentioned, we removed this section.

L484, 485, “The backward trajectories (not shown) indicate no direct link of the air mass at 5-6 km height with lower heights (and thus a direct fire smoke uptake from the sources).” For this to make sense, the trajectories must be shown. Even so, it is difficult to understand what the authors mean here. What is “direct…uptake”? What are “the sources”? Either remove this sentence or bolster it with displayed trajectories and precise terminology.

As mentioned, it was removed.

L489-494, Nearly identical trajectory discussion regarding the 2021 smoke: I have the same concerns as stated above. Please take the same corrective actions.

As mentioned, it was removed.

However, we present new analyses in Section 5.1, based on a CALIOP observation in August 2021. We combined subsequent CALIOP observations and backward and forward trajectories to investigate self-lofting effects.

L494, “Initial smoke plumes were at around 3 km height on 4 August 2021.”: What is meant by “initial” when the previous sentence refers to smoke 10 days earlier. Please expand and clarify with more precise information.

As mentioned, all this is removed now.

L495, 496, “Fractions of this smoke plume must have reached the tropopause and later on the lowest stratosphere.”: What does this mean? Why “must” the smoke have behaved like that? Was there a stratospheric, Arctic aerosol plume in Fall 2021 on par with the Raikoke/MOSAiC episode of 2019? Given the suggestiveness of the CALIOP observations in 2021 as compared to 2019 (the plume altitude and AOD are greater), one might hypothesize that a similarly strong and long-lasting plume was in evidence in 2021/22. Can the authors confirm such a plume? If so, that would be a powerful set of circumstances. If not, it would tend to shed doubt on the smoke composition of the 2019 Arctic stratospheric plume. The authors are asked to quantify the Fall/winter 2021 stratospheric aerosol load in comparison to 2019 and draw appropriate conclusions.

As mentioned, we exclude this discussion. Concerning the CALIOP observations in 2021. The troposphere was filled with smoke up to the tropopause over the Arctic, especially in the second half of August. The layering structures were different compared to 2019. Such an (isolated) diffuse near-tropopause layer was not found in 2021. Instead, often the entire troposphere between 5km and the tropopause was filled with smoke over a large latitudinal range (65-80°N) so that an extra layer around the tropopause could not be resolved. Sometimes the depolarization ratio was enhanced and may indicate pyroCb lofting, sometimes the depolarization ratio was close to zero.

It is not only a question of the aerosol conditions. Self-lofting needs probably also favorable stagnant conditions… and the meteorological conditions always vary and are never exactly the same from year to year during the burning season.

Figure 17. Unless the authors explain the rationale or algorithm for marking the “plume center height” in each panel, it is reasonable to conclude that the choices are arbitrary. If they have just been eyeballed, one could make an independent set of marks justifying no altitude gain; there are aerosols from the boundary layer to nearly 10 km in most panels. Moreover, the authors show one selected CALIOP curtain each day and thus don’t account for the full synoptic view of the smoke on each date. A fuller synoptic view of the plumes on these dates, involving multiple CALIOP curtains per day and horizontal map views of the overall plume (using something like visible imagery, UV aerosol index, carbon monoxide) are essential for the reader to have confidence in the arguments brought here. And, as mentioned above, some traceability from 12 Aug 2019/4 Aug 2021 back to a definable source is also called for.

Section 4.4, including Figure 17, is removed.

Figure 17a: The longitude “170” seems to be incorrect for this CALIPSO orbit. It should be ~150E.

Section 4.4, including Figure 17, is removed.

L486, “The observation were taken over the East Siberian Sea north of Siberia (78°N, 160°E).”: What observation? Do the authors mean “observations”? If so, what observations? 78N, 160E is not near the CALIPSO track, as far as I can tell. The orbit shown in Fig 17a is much farther west. On that track, the lon at 78N is ~123E. The center of the thick smoke layer in 17a is north of 80N, but still far west of the trajectory start point. Please clear up the confusion.

Section 4.4, including Figure 17, is removed.

Figure 18: Why are the trajectories initialized at 07 UTC? This is neither close to the night or day orbits of CALIPSO as far as I can tell. Why not initialize them at the time of 12 Aug 2019, 4 Aug 2021 CALIPSO observations? If they were initialized at the CALIPSO observation time, that should be clarified in the caption. Assuming I have found the correct CALIPSO orbits to go with the two start points, the forward trajectories launched from them are significantly different than those shown. Please clarify or explain.

Section 4.4, including Figure 18. is removed.

Figure 17a caption: Why are these heights (3, 5, 7 km) chosen? The initial layer is said to be at 3 km. Secondly, the displayed map legend says 3, 4, 5 km. The trajectories conform to these values. Are the heights in the caption the ones intended for display, or is the caption incorrect?

Section 4.4, including Figure 17, is removed.

L489-497, Discussion of the 2021 smoke: There were 5 pyroCbs in the Sakha region of Siberia between 26 July and 7 August. No accounting of pyroCbs in 2021 is given by the authors. In addition to the Siberia pyroCbs, there were numerous pyroCbs in the USA and Canada in July and August. There was even a possible pyroCb in Greece in early August. Hence, there were multiple direct inputs of smoke into the UTLS that could have been transported to the Siberia zone in the time frame under study. It is therefore possible that some of the elevated smoke in Figure 17b is from pyroCb or pyroCu injections (when a pyroCb event occurs, naturally and frequently they are accompanied by numbers of pyroCu, which efficiently pollute the free troposphere. See Figure 7 of Fromm et al. (2021; https://doi.org/10.1029/2021JD034928) and attendant discussion. The authors are asked to account for the added complexity and uncertainty regarding Figure 17 and associated analysis.

Section 4.4, including Figure 17, is removed.

Nevertheless, in the new Section 5.1, we show one CALIOP smoke case and then discuss briefly the CALIOP smoke profile observations in August 2021. It is mentioned that pyroCb-related lofting was involved in the smoke transport.

L487, "The black dots indicate the position of the smoke layer…": The smoke in each panel is seemingly much wider than the dots represent. A clearer explanation of the dot location choice is needed.

Section 4.4, including Figure 17, is removed.

Figure 17, star symbol: What is the significance of the star symbol versus the dot? The star symbol is not defined in the text or figure caption.

Section 4.4, including Figure 17, is removed.

L511-513 (End of Section 4.4), "We can conclude that even tropospheric smoke can ascend significantly from lower tropospheric injection heights up to the tropopause level within a few days and even enter the lower stratosphere as demonstrated in Ohneiser et al. (2021).": Once again, O21 did not "demonstrate" with observations that smoke self-lofted across the tropopause in amounts to justify their conclusions. They were only able to hypothesize this. In the current work, the observational aspect did not demonstrate the FTTS diabatic pathway, even if what they showed in Figure 17 was totally robust. No stepwise continuation of the smoke lofting to and through the tropopause was demonstrated. This should have been achievable given the enormous AODs and multi-season aerosol persistence O21 reported. Moreover, in all comparative respects, 2021 Siberian smoke equaled or exceeded 2019 in terms of AOD and height.

Yes, we agree. We did not observe self-lofting. All the CALIOP data analysis (even in the new Section 5) does not allow us to state: We found a proof for smoke self-lofting. NO! In our revised version of the manuscript, we carefully and critically present our results.

Considering these two primary factors as inputs to the self-lofting model, Fall and winter 2021's boreal stratosphere should embody a smoke plume at least on par with 2019. If that condition did not occur, there was effectively no observational demonstration of the FTTS diabatic lofting in either season. The authors are encouraged to assess that logic, dispute it, or consider a reinterpretation of their hypothetical model.

As mentioned, one cannot expect to see the same evolution of smoke layers in the upper troposphere and lower stratosphere in 2019 and 2021…. Meteorological conditions are never totally equal. We see smoke in the CALIOP data up to the tropopause or better around the tropopause in both years. CALIOP observation could not be used to see diffuse thin aerosol layers higher up in both years. Only in cases with pyroCb-lofted smoke one may have a good chance to see compact, isolated ascending plumes producing considerably high backscatter together with enhanced depolarization, as it was the case for the record-breaking Canadian and Australian smoke events.

The goal of our manuscript is not to illuminate in detail the wildfire and smoke situation in 2021. The goal is to discuss the 2019 smoke over the Arctic.

Minor Issues

Figure 1 (and lines 38-41): The images are offset by one day from those stated in the caption. The actual dates are 3-5 January, not 2-4 January. The thickest smoke on 3 and 4 Jan over the Tasman Sea is lower to mid-tropospheric. The UTLS smoke from the December phase of ANYSO pyroCb event had already blown east of New Zealand. The only substantial UTLS smoke in Fig. 1 is on 5 Jan. The arrows point to the plume from the 4 Jan pyroCb phase.

We improved Figure 1, and also rephrased the text in the introduction. But the message is that self-lofting can immediately produce very complex layering structures. The yellow arrows shall guide the reader to see the smoke. Many scientists may have no idea how to distinguish smoke and clouds.

L42-46, discussion of smoke over cloud: The radiative implications of smoke over cloud are very complex. Referring to them in the context of Fig. 1 snapshots seems to draw relations that are likely to be irrelevant. Smoke plumes blown by fast UTLS winds will be almost totally decoupled from most cloud systems. There won't be much time for a cloud, small or big, to have an impact on a plume blowing faster than and independently of that cloud. Making the case for cloud effect on self-lofting calls for a much more sophisticated discussion.

We re-arranged the text to better explain the cloud impact. The impact is via changed albedo conditions. As soon as the smoke is above a white cloud layer, there is almost a factor of 2 more solar radiation available for absorption.

L43, "compact layers": Please define "compact."

We changed the text (left out: compact). Words need to be self-explaining. Otherwise, should be better not used.

L67-68, discussion of dust self-lofting: The Daerden et al. paper is on Martian dust. Applicability to the Earth is unclear and not mentioned. Please elaborate or remove this citation. It was not evident to me that Gasteiger et al. discussed anything but vertical mixing, which is not synonymous with self-lofting. I could not find observational evidence therein of dust diabatic lofting. Please point it out if I missed it.

We changed the text and removed this point.

L187, "Siberian fire smoke reached the stratosphere via the self-lofting process.": As in other places in this manuscript, this is stated as fact. The authors hypothesized this in O21, and make statements elsewhere herein to the effect of “it must have happened,” but definitive observational proof does not predate this paper. The authors are asked to modify the wording here and throughout to reflect the uncertainty of the FTTS self-lofting pathway.

We agree! We carefully went through the text to emphasize: All our studies may indicate self-lofting effects, but it remains a hypothesis that self-lofting contributed to the smoke vertical transport.

L187-192: Citations are needed for the Kuwait- and Canadian-plume BC fraction. Same goes for the fuel types in Canada, Siberia, and Australia.

We improved Kuwait-fire-related information and references in the Introduction, in Section 3, and 3.5. Concerning fuel types that is, to our opinion, google-like or text-book-like knowledge, however, we cite Ohneiser et al. 2022 here. Regarding BC fractions we also give references now. All this is given in Section 3.

L193-195: “more homogeneous”: Please define “homogeneous” since it is used as a comparator to pyroCb plumes.

Done, it was rephrased! Avoid to use ‘more homogeneous’.

L196, 197, discussion of pyroCb-plume depolarization ratio: A citation is needed for the “20%” value. Also, these double-digit depolarization ratios last far longer than the “fresh” stage. Please reword accordingly.

Reference is now given!

L271, “coherent structures”: Please define “coherent.”

We do not mention ‘coherent structures’ anymore.

L312-319, discussion of Kuwaiti oil-rig fire plumes: One of the prime science topics centered on these plumes was a validation of the Nuclear Winter hypothesis. Because of the size and darkness of these smoke plumes, it was thought that this might have been a natural (or at least inadvertent) experiment in self-lofting. Yet there are no publications to my knowledge that revealed significant self lofting. Rather, the consensus seemed to be that in spite of the absorption and optical depth of the plumes, little or no diabatic lofting was observed (Hobbs and Radke, 1992; Larry Radke, personal communication, 2008). Hence the strategic value of this case study is brought into question. The authors are asked to reflect on the published findings of observed Kuwaiti smoke heights as a function of time and make suitable revisions to the manuscript.

We checked the literature comprehensively. Limaye et al. (1991) observed lofting. They found plumes at 6-7 km height in a distance 2000 km away from the sources. Close to the sources the plumes were below 3 km. We mention that in the introduction and in Section 3.5.

L446-454, Discussion of Figure 16 and MODIS AOT: This is a very interesting result. But the method needs to be explained better. Just how are the MODIS AOT data evaluated? What is done when there are no MODIS AOT retrievals due to clouds or smoke flagged as cloud? Given that MODIS data are daytime values, were CALIOP daytime curtains used for MODIS associations? And what are the implications of the results? Are the CALIOP-based AOT's low biased? Are the MODIS data a better approximation for the stratospheric plume AOT?

We describe how we used the CALIOP and MODIS data to obtain our parameterization (Section 4.1). When there are no MODIS pixels left after cloud screening then there are no MODIS AOTs. It is not necessary to have rather exact AOT values and day by day AOTs to obtain the AOT parameterizations used.

L471-476: The importance of this paragraph is not clear. The authors seem to be saying that if several sources of uncertainty all align in one direction, the overall error in final plume height can be very large. If that is a correct interpretation, isn't that self-evident? Please clarify.

We removed this paragraph to avoid confusion and to keep the discussion short here.

By the way we changed the figures (now we show more compact Figures 12 and 13 in Section 4). We reduced in this way the number of figures in Section 4.

L481, “Almost coherent smoke plume structures…”: Assuming “coherent” was already defined, what is meant by “almost coherent?”

We removed such statements.

To sum up:

All in all, we think that the manuscript is in a good shape now. We also believe that we could convincingly present a number of arguments that the 2019-2020 aerosol conditions as observed with CALIOP and the MOSAIC Polarstern lidar in the UTLS height range point to smoke self-lofting as a significant process to transport smoke upward into the tropopause region over high northern latitudes in the summer of 2019.

The key findings concerning tropospheric self-lofting over Siberia in 2019 were summarized in Section 5 as follows:

We found to our opinion clear evidence supporting our hypothesis that self-lofting processes played a key role in the formation of a smoke-dominated UTLS smoke layer. First of all, the observed AOTs were a factor of at least 5 higher AOT than expected from the Raikoke SO$_2$ emission, so there were additional aerosol particles in the UTLS height range besides the Raikoke sulfate particles. Typical optical fingerprints for aged smoke particles were found from the polarization Raman lidar observations. The inverse spectral slope of the lidar ratio and the high 532~nm lidar ratios are a clear and unique sign for the dominance of smoke in the UTLS aerosol layer. No other aerosol type was ever observed with lidars showing such a spectral behavior of the lidar ratio. The observed low particle depolarization ratios clearly show that lofting by pyroCb convection played no or only a minor role. The occurrence of pyroCb-related lofted smoke was always found in published lidar observations to be associated with enhanced depolarization ratios caused by irregularly-shaped, fastly ascending smoke particles. Then, only self-lofting is left to explain the built-up of the optically relatively dense, long-lasting smoke layer observed over the Arctic for almost a year until May 2020. The low depolarization values pointing in addition to relatively low ascent rates in the troposphere so that the smoke particles had sufficient time to age and to develop a compact and spherical core-shell structure. This feature is expected according to our simulations when self-lofting comes into play. Finally, the formation of a near tropopause smoke layer is also in line with the simulations suggesting or predicting an accumulation of upward moving particles around the tropopause because of the height-dependent ascent rate (with minimum at the tropopause). However, it remains to be emphasized that all these arguments can not be used as a proof that self-lofting processes really occurred and triggered significant upward motion of smoke-filled tropospheric air parcels. It remains a hypothesis.

Our final remark in this reply letter:

There is no reason anymore to reject such a paper that will trigger future work concerning the role of self-lofting of light-absorbing aerosols in the atmosphere. There is so much smoke around the world (central and southern Africa, in South America, western North America, western to eastern Mediterranean, Middle East and central Asia, Southeast Asia, Australia, Alaska, Canada, Siberia), that we believe, self-lofting plays a role on a global scale. However, it remains rather difficult to provide clear observation of slowly ascending of likewise thin aerosol layers. Self-lofting leads to a prolongation of the lifetime of all these light-absorbing particles in the atmosphere and this aspect is not considered in any of the numerous climate models used to predict climate change.

Finally, we should add that we presently have to review a JGR manuscript on global aspects of ascending BC-containing aerosol layers and these authors find similar results concerning self lofting in the troposphere and stratosphere as we discuss in our manuscript. One highlight is that the used Earth System Model indicates a strong increase in UTLS smoke mass concentrations (by 50%, annual mean increase) in the 8-22 km height range of the northern part of the Northern Hemisphere when BC absorption of solar radiation is considered in the simulations. As these authors pointed out in the introduction, their JGR article was motivated by our self-lofting studies presented in Ohneiser et al. (2021) on MOSAiC observations and by this ACPD manuscript (Ohneiser et al., 2022), and by other recent papers dealing with self lofting processes.

Second Reply:

Dear Reviewer!

We thank You for reading the manuscript and for your comments and remarks! We agree with your criticism concerning our tropospheric self-lofting part (observations vs simulations). The discussion was not convincing and straight forward.

As a consequence, we re-wrote the entire section on tropospheric self-lofting of smoke (now section 5, section 4.4 in the submitted version). We skipped the simulation part in Sect. 5. CALIOP observations (and one Leipzig lidar observation) are presented only in this revised section. We re-analyzed the entire CALIOP data set over central eastern Siberia and the Arctic from 22 June 2019 (at that day the Raikoke volcano erupted) until October 2019 (to have an overlap with the MOSAiC Polarstern lidar observations that started in the end of September 2019). Together with new literature on stratospheric smoke (Knepp et al., 2022, Xian et al. 2022), corroborating our hypothetical approach, we believe that we have now several indications that self-lofting contributed to the upward transport of smoke within the troposphere. However, we agree, it remains a hypothesis! And that is emphasized several times in the revised manuscript.

However, the topic is a very new aspect in atmospheric science that clearly deserves publication. Even if we are in trouble to demonstrate that self-lofting played a major role in vertical smoke transport processes in the troposphere, our simulations, the impact study and uncertainty analysis, and the applications to Canadian and Australian fire events are worth enough to be published. No doubt!

The first part of the article (Sections 1-3), and also the discussion of comparisons of observations vs simulations for stratospheric ascending Canadian and Australian wildfire smoke layers (in Section 4) remained widely unchanged with respect to the submitted version.

Now to the item by item response:

Our answers are in BLUE!

Significantly changed text or added text is given in BOLD in the revised version of the manuscript.

Before we start, please note that the title of the article has been shortened and we added a co-author (Fabian Senf), he is an expert for atmospheric dynamics and modeling.

The manuscript by Kevin Ohneiser and coauthors addresses the solar-driven lofting of wildfire smoke plumes in the troposphere and stratosphere using ECMWF radiation transfer scheme with different parameterizations and satellite observations using CALIOP and MODIS instruments. The ascent rates of smoke plumes produced by Canadian, Australian and Siberian wildfires derived from CALIOP observations are compared with the calculated ascent rates from radiative transfer simulations. The main goal of the study, as stated in the abstract, is to demonstrate that the radiative heating of intense smoke plumes is capable of lofting them from the free troposphere up to the tropopause and into the stratosphere without the need of PyroCb injections.

We changed a bit the main goal. The main focus is now on tropospheric self-lofting only, i.e., from the injection heights of 2-6 km to the tropopause. Lofting of smoke from the tropopause to greater heights within the stratosphere is already well documented and well described in the literature (as described in the introduction).

After a detailed description of the modeling setup, sensitivity tests and uncertainty discussion, the authors demonstrate in Fig. 11 that a 2.5 km- thick smoke plume with a realistic BC fraction of 2.5% and a very large AOT above 2 should rise from 3 km altitude into the lower stratosphere in two weeks. However, the analysis of CALIOP observations of tropospheric smoke from Siberian wildfires in the following section does not provide any support for the cross-tropopause transport of aerosol plumes rendering the main goal of the study unachieved and casting doubt on the usefulness of the simulation results.

Yes, this is true! Therefore, we changed the strategy of our study. As mentioned above, the study is now based on the CALIOP data set (day by day, scene by scene) from the Raikoke eruption on 21-22 June 2019 until the end of October 2019 with focus on smoke layers (pyroCb-related, self-lofting related) but also on features produced by Raikoke sulfate aerosol. We concentrated on the high northern latitudes >65°N. We found a number of fingerprints and arguments for self-lofting in the upper troposphere in line with our simulations. But we do not show any simulation here. Our 1D simulation model cannot be used to simulate 3D-air motions in the troposphere. We mention that in Section 3.4. However, even after listening a number of convincing arguments in Section 5, it remains a hypothesis that self-lofting really occurred. This is emphasized frequently!

More specifically, there are several major issues as follows.

The description of the satellite instruments, data versioning, measurement uncertainties and the approach to data treatment is totally missing in the manuscript.

We agree! However, we exactly provide similar information on CALIOP and MODIS and other instruments, data use, etc. as in our foregoing papers on Arctic smoke (Ohneiser et al., ACP, 2021) and Australian smoke (Ohneiser et al, 2022). And that was acceptable. CALIOP and MODIS are so well known that such a detailed introduction of these instruments (including, data handling, quality levels, uncertainty analysis, etc) is no longer needed to our opinion. And all necessary information, needed in this self-lofting paper, is given in Section 4.1, i.e., how we got the layer center heights (in the case of the presented Canadian and Australian smoke layers) and how we got the AOT values from CALIOP observations and (now also in more detail, from MODIS data).

As far as I understood, the authors used CALIOP quicklooks to derive the layer thickness and mean attenuated backscatter, from which the AOT is calculated using an arbitrarily chosen factor of 1.5, which should account for the light attenuation. While the derivation of layer thickness from the quicklooks may be deemed sufficiently accurate (although for compact stratospheric plumes only), it is unclear how the authors derived the layer mean backscatter from the images. Was it done by reading the colors of each individual pixels and using the color bar to retrieve the values? If so, the uncertainty of such estimates might be unacceptably high and I wonder how such estimates would compare with those by Kablick et al. provided in Fig. 16.

The factor of 1.5 accounts for the probably too low lidar ratio of 65 sr (the smoke lidar ratio in the CALIOP data base), because we observed lidar ratios of 90-100 sr for the Australian smoke (Ohneiser et al., 2022). This is now correctly mentioned in the revised version. This was not well described in the submitted version.
The use of quicklooks of attenuated backscatter (color plots) or the use of downloaded particle backscatter values doesn't matter much. At the end, the obtained AOT values may differ by about 5-10%. That is our experience. Kablick also used backscatter coefficients, multiplied by the smoke lidar ratio, and then integrated the obtained extinction values from layer base to top.

Constrained by the CALIOP AOT from Kablick et al., the simulated ascent is nowhere near the observed one and the authors opt to constrain the simulation with MODIS total AOT data (ignoring the tropospheric aerosols), which is substantially higher than both the CALIOP-derived AOT and, what is particularly puzzling, much higher than the estimates by Ohneiser et al. (2020), their Fig. 5b, reporting the lidar-derived AOT@532 of 0.1 – 0.3 for the Australian smoke plume in late January 2020 (which would be consistent with Kablick et al. data in Fig. 16). The authors thus seem to deliberately ignore their own observations for the sake of reproducing the observed lofting in the simulation.

The observations in Ohneiser et al. (2020) were done at the edge of the smoke-filled vortex and the AOT values were therefore smaller. The MODIS data overestimate the UTLS AOTs before 17 January (this is shown now in the new, more compact Figure 13) and after 17 January the values became reasonably small (after subtracting a typical marine AOT value). Then, the CALIOP AOTs of Kablick et al. and the MODIS AOTs compared reasonably well.
In the revised version, we mention that a tropospheric marine background AOT of 0.05 was subtracted from the observed total smoke MODIS AOT. The tropospheric AOT contribution was cleatly underestimated until 16 January when using a tropospheric AOT of 0.05.
As a consequence, the simulations based on MODIS AOTs start on 17 January 2020.

Section 4.4 and Fig. 17. The Siberian tropospheric smoke plumes show rather complex vertical structures, whereas the determination of the aerosol layer vertical boundaries (critically influencing the AOT estimate) appear to be somewhat too arbitrary. Personally, I do not see any significant lofting for the both cases shown in Fig. 17. It rather appears that the smoke was found in the UT from the very beginning, which would point to the PyroCb-driven vertical transport.

Section 4.4 including Figure 17 is removed now, and substituted by the new section 5.

Discussing the Siberian smoke plumes, the authors state that “fractions of this plume must have reached the tropopause and later on the lower stratosphere” without providing any supporting observations, and the only reason I can possibly think of is the absence of such observational evidence. Moreover, the simulations based on an assumption of the persistent Gaussian vertical shape of the layer (which is obviously not the case here) show even weaker lofting than what is inferred from CALIOP quicklooks. I also wonder why the simulation was not extended further in time (using, e.g. 15% daily AOT decrease) to provide at least the modeling support for the potential lofting up to the tropopause level.

Again, Section 4.4 is removed, and substituted by the new section 5. It makes no sense to compare simulations with observations in the convective and turbulent troposphere. We explain why our simple ECRAD model cannot be used to simulate 3D-air motions in the troposphere in Section 3.4.
The assumption of the real shape structures or the assumption of a Gaussian shape profile almost lead to the same results.

All in all, we think that the manuscript is in a good shape now. We also believe that we could convincingly show that the 2019-2020 aerosol conditions as observed with CALIOP and the MOSAIC Polarstern lidar in the UTLS height range can only be reasonably explained by considering smoke self lofting as a significant process to transport smoke into the tropopause region. To summarize again the main 3 arguments that point to a strong role of smoke self-lofting are:

(a) The observed UTLS AOT (caused by smoke and sulfate) was much higher (0.1-0.2 at 532 nm) than the expected sulfate AOT (<0.025) originating from the Raikoke eruption. However, the observed low depolarization ratios pointed to a minor impact of pyroCb-related smoke lofting. Consequently, another process is needed to transport smoke upward, towards the tropopause.

(b) By carefully expecting all CALIOP observations at high northern latitudes from June to October 2019, we detected a near-tropopause aerosol layer, clearly visible from mid July to mid of August (in the period of strongest fires over Siberia). The occurrence of this layer is in line with one of the main simulation results. The lofting rate decreases with height in the upper troposphere, and shows a minimum at the tropopause. In case of a steady upward transport of smoke this leads to smoke accumulation at the tropopause.

(c) The found lidar ratio characteristics (355 nm lidar ratio is much smaller than the 532 nm lidar ratio, lidar ratio at 532 is typically larger than 70 sr indicating strongly absorbing aerosol particles) unambiguously points to smoke as the dominating aerosol type in the UTLS aerosol layer. Such a unique spectral slope of the lidar ratio has never been observed by the lidar community for any other aerosol type.

Finally, we should add that we presently have to review a JGR manuscript on global aspects of ascending BC-containing aerosol layers and these authors find similar results concerning self lofting in the troposphere and stratosphere as we discuss in our manuscript. One highlight is that the used Earth System Model indicates a strong increase in UTLS smoke mass concentrations (by 50%, annual mean increase) in the 8-22 km height range of the northern part of the Northern Hemisphere when BC absorption of solar radiation is considered in the simulations. As these authors pointed out in the introduction, their JGR article was motivated by our self-lofting studies presented in Ohneiser et al. (2021) on MOSAiC observations and by this ACPD manuscript (Ohneiser et al., 2022), and by other recent papers dealing with self lofting processes.