

Dear Reviewer!

First of all thank you for careful reading and all good and constructive suggestions which improved the paper significantly (we hope). It is usually often of great and complementary help when a reviewer is not an expert in the same field as the authors. You realized certainly that we are not working every day in the field of stratospheric ozone depletion and are also not just experts for polar meteorology.

Before we answer all comments and questions, step by step and item by item, let us provide a brief overview of the essential changes:

Motivated and mostly forced by the other two reviews, we went deeply into the literature with special focus on volcanic sulfate aerosol (originating from the recent Raikoke and from the Sarychev volcanic eruptions), into our own Leipzig lidar data analysis, into CALIPSO observations and HYSPLIT trajectory analysis and conducted further simulation studies to bring together much more solid information and substantial argumentation to provide a plausible link between the Siberian fires (in July-August 2019) and the polar smoke layer we observed later on in the North Pole region (since late September 2019).

Some significant changes before we start the step by step reply:

- We show a new figure (Fig. 2, time series, MODIS AOD from 2000-2020), highlighting the extraordinarily strong 2019 fire season in central and eastern Siberia.
- We introduce a new table (Table 2), highlighting the unique signature (fingerprint) of aged wildfire smoke, namely the inverse spectral slope of the lidar ratio ($LR_{355nm} < LR_{532nm}$) together with high LR_{532} . We compare these smoke features in the new table with the ones for mineral dust, volcanic sulfate and ash, Arctic, European, American haze, etc.
- We show a new HYSPLIT plot (Fig. 5) to better explain the link between the CALIPSO overflight over Siberia and the strong fires... downwind (west!) of the flight track.
- We show a new figure with the Polarstern track (Fig. 6, same as in Engelmann et al., 2021).
- We created a new figure (Fig. 7) to better show the influence of the polar vortex on all the observations.
- We introduced a new section (Section 3) on Siberian fires, self-lifting hypothesis, consequences for aging (leading to spherical smoke particles), and also on the potential misclassification of smoke layers as sulfate layers when using the CALIPSO aerosol typing scheme (Sect. 3.1).
- We shortened the ozone-depletion/smoke/PSC section as requested, and combined the two figures of this section to one figure (Sect. 5, Fig. 17).
- We skipped several figures (layer-depth histogram, monthly mean extinction profiles) to keep the paper as short as possible.

We think, more cannot be done. Many parts of the paper are substantially improved, caused by the recommendations of the reviewers. But this kind of research must also tolerate hypothetical explanations and argumentation. We feel this approach, as we present it now, is justified.

Step-by-step reply: our answers in BLUE

Ohneiser et al. present a summary of a persistent wildfire smoke aerosol layer observed during the MOSAiC field campaign in the high Arctic. The authors primarily focus on observations obtained from the "Polly" Raman lidar operated onboard the Polarstern, but also compare with other lidar platforms (e.g., CALIPSO and KARL). With the unique observations made possible by Polly and the

MOSAIC campaign, the authors are able to provide quantitative descriptions of the optical and microphysical properties of observed aerosols for a region where such measurements are sparse. These measurements are made even more important by the unique atmospheric and aerosol conditions related to the 2019 Siberian wildfires and the unique polar vortex and ozone conditions of late-2019 and early 2020.

I am not a lidar measurement expert; in fact the editor has asked me to provide my views for portions of the manuscript related to polar vortex conditions and ozone depletion. However, I of course read through the entirety of the paper (multiple times), and so I feel comfortable saying that the paper is generally well-written. While I can not provide an expert assessment of the observational aspects and descriptions, as a "knowledgeable layperson", I'd say The authors mostly gave thorough and convincing descriptions of the observational data and their characterization. The paper is clearly cutting-edge and would be a perfect fit for ACP. However, I do have a few concerns, some more major than others; most of these are related to the aspects surrounding the polar vortex and ozone depletion in Section 4. Since Section 4 is a relatively small portion of the overall paper, I would say the revisions required to address my comments would be relatively minor to implement, but I do feel strongly that they are necessary changes.

General comments

- (1) In reading through the paper, reaching Section 4 felt like a definite shift. Up to that point, everything felt (as a reader) mostly authoritative and backed up by quantitative results. In contrast, Section 4 felt very vague and overall speculative. One paragraph particularly stuck in my mind -- p14, L10-15 -- as it included many instances of language such as "were probably ..." and "were likely". Three "precise" research questions are posed early on in the section, and yet the remaining results and discussion do nothing to answer them and instead involve results that only hint at possibilities. Section 4 essentially says "the smoke aerosol layer could have been important for ozone depletion" simply because the PSC, smoke, and ozone depleted layers overlapped in the vertical (Figures 15 and 16).

We got the point and worked on Section 4 to meet the concerns. Section 4 is now Section 5 in the revised version. We re-arranged the text. See further explanations below.

Let us briefly start with this: There are many hypothetical arguments and aspects in the paper. This must be allowed as long we state that clearly. And we do so. For example, there is no clear answer to the question: What was the most likely source for the smoke we observed over the High Arctic? We try to answer this question in the new Section 3. There is also no clear answer to the question: Is it possible that the smoke contributed to the strong ozone depletion?

We think that nobody has an idea at the moment how efficient glassy organic particles are in heterogeneous chemical processes in the stratosphere and that nobody knows how these glassy particles can influence PSC formation! It will take years to investigate this properly! You need sophisticated laboratory studies for that. Modelling is insufficient.

Our job is simply to raise our hand to tell the ozone science community that there was a persistent smoke layer over the Arctic in the spring of 2020 when a record-breaking ozone depletion occurred. Please keep that into consideration in your future studies

By the way, we operate another lidar at Punta Arenas, Chile, and detected the Australian smoke from the tropopause up to 26-30 km height, from January 2020 to April 2021. We were sure, already in March-June 2020, that there will be a strong ozone depletion in the southern

hemispheric spring season (September to November) because of the smoke.... sure, without any (precise) idea about the pathways...

There are many details that would have to be worked out in much more detail to answer the questions posed in Section 4. For instance, if the MOSAiC measurements were to be of use in answering these questions, then there would need to be details about the time-varying geometry of the vortex. While it's a fairly safe assumption that the vast majority of measurements (lidar, ozonesondes, etc) sampled air within the polar vortex, on weekly timescales the vortex and its edge are quite mobile (and similarly, so is the region of air cold enough to form PSCs). This means that measurements could sometimes sample air in different parts of the vortex, meaning some measurements would be less relevant than others in establishing where things were "in the right place at the right time".

First of all, this triggered us to present the new Figure 7 with the scaled potential vorticity and temperatures for different height levels along the Polarstern route from October 2020 to May 2020.

As mentioned, we re-phrased and rearranged the text to give the impression in Section 5 that we just want to point to a new aspect: smoke and ozone depletion. not more...

If we do not show that, others will do it. Furthermore, we see the same in the southern hemisphere: a strong ozone depletion height range, but this time right in the center of the Australian smoke layer.

As written, I do not feel Section 4 should be kept. I would suggest that it should be shortened and perhaps folded into the current Section 5, with content from Section 4 forming the basis of some discussion (i.e., providing motivating context for future work). At the very least, Section 4 should be shortened and rearranged so that the authors present the "possibility" before posing the big picture questions. The authors should also be clear (if they keep any of Figures 15 or 16) that conclusions cannot presently be drawn from their analysis.

This is done! We agree! We shortened Section 4 and rearranged the remaining text parts.

In my specific comments below I provide more detailed questions/comments related to the content of Section 4.

(2) Again, this is not my area of expertise so perhaps these are naive questions: The title of the paper and the introduction generally outline the assumption that the smoke aerosol layer measurements were tied to the Siberian wildfires, but are there not other potential confounding sources?

We changed the title! Removed 'Siberian'!

We agree, we should be more careful and therefore rephrased the introduction. But as you will see, there was the volcanic sulfate layer (with Raikoke as the source) and the smoke layer (from the huge Siberian fires as we hypothesize) and nothing else. There were many fires within the Arctic circle in 2019. But very strong and long-lasting fires were needed and probably also stagnant weather conditions to initiate self-lifting in order to end up with smoke in the stratosphere. Only if the smoke reaches stratospheric heights it will be distributed of the entire northern part of the Northern Hemisphere and will survive for months.

Can the authors provide more evidence to say that the Siberian wildfires were likely the dominant/overwhelming contribution to the observations discussed throughout the paper?

This motivated the new Section 3 on the Siberian fires, on self-lifting aspect, on smoke aging aspects, and even on the failure of the CALIPSO aerosol typing scheme to identify smoke, but instead to miss-classify that as sulfate aerosol.

The paper lacks some descriptive context as well: how severe were the 2019 Siberian wildfires in comparison to prior years? It is perhaps fortuitous that the MOSAiC campaign was able to sample aerosol conditions largely influenced by Siberian wildfires, but is there a quantitative measure of how unusual 2019 was that would better emphasize the importance of the MOSAiC measurements? These kinds of details may be present in the articles that the authors cite, but I think it's worthwhile for the authors to make them explicit where possible.

We include the new Figure 2 with MODIS AOD observations from 2000 to 2020. The maximum AOD was observed in August 2019 during the last 20 years.

Yes, it was fortuitous! And we (as aerosol lidar enthusiasts) were happy to observe this exotic aerosol layer throughout the winter half year of 2019-2020.

(3) There were a couple of times in the paper where a Figure is introduced and a portion of it is discussed, but then the discussion moves onto one or more different figures before later coming back to the initial one. An example of this is Figure 9: On page 9, line 17 Figure 9a is introduced, but then within the same paragraph the authors move on to Figure 10. The authors go back to Figure 9a, and eventually introduce Figure 11 before eventually mentioning Figure 9b. I recognize that this may just be personal preference, but I think that cases like these generally suggest that text or figures should be re-organized to better maintain the serial nature of the text.

I understand that in the final manuscript the authors will generally not have much of a say where figures will end up in relation to the text, but it is still a bit awkward for a reader to have to flip between multiple figures for a given piece of text (whether on paper or digitally). I wouldn't classify this as a major issue in this paper, but I still urge the authors to consider whether their figures or text could be restructured to make things flow more naturally.

We agree, kept these suggestions in mind, and did our best to improve the structure. We partly rearranged figures to avoid such a confusing back and forth in the discussion.

Specific Comments

P6, L17 and Figure 5: The information about the arrival height of 10km should be included in the figure. Also, what about the vertical information? How did these trajectories evolve in terms of altitude/pressure over this time period? Is this not important information for discussing the airmass origins?

We added the plot with the height information (lower panel of Figure 9, this was Figure 5 in the old version), and mention the arrival height of 10 km in the caption.

P10, L22: I do not believe the Lawrence et al ref here shows that the polar vortex collapsed on 20 April. In fact, their Figure 10 shows the vortex was unusually long-lived, lasting into May. Maps of the vortex on April 20 show that the vortex was undergoing a split, so it's probably fair to say the vortex began decaying around this time. However, if you click through further dates ..., I think it's clear that a distinct polar vortex is present well into May. This comment is also relevant to other spots in the text where the authors mention the vortex collapsed in late April (e.g., P2, L11).

We improved the text accordingly.

P13, L14-15: This requires more nuance; it wasn't just weak planetary wave forcing, but also the very strong dynamical influence of downward wave reflection events during the 2019/2020 winter season (this is discussed in detail in the Lawrence et al ref).

We agree! But at the end we removed the long paragraph with all these meteorological statements to shorten the section. We did not see a need anymore to explain why the vortex was so strong.

P14, L3-9: While questions 2-3 are interesting and are worth further exploration, I'm not sure question 1 has much scientific merit in relation to the polar vortex. There are too many issues with timing and location and other confounding factors. Early in the season, the polar vortex was generally not extremely strong or weak. Furthermore, the very strong vortex conditions in Jan-Mar coincide closely with the dynamical downward wave reflection events mentioned above.

We removed question 1!

P14, L18-22: While true, I'm not convinced this is an appropriate apples-to-apples comparison. For instance, the 2019 Australian wildfires were severe enough to be comparable to a moderate volcanic eruption in terms of the impacts to solar radiation (as noted in the Khaykin 2020 paper already cited). I do not think that the Siberian wildfires have been established as being anywhere near as severe. There are other differences, but the point is, this kind of comparison runs the risk of equating inherently different events, which could be much more coincidental than this statement suggests.

We removed this part in Section 5. Only in the conclusion section 6, we left the remark concerning the ozone hole over Antarctica and the occurrence of the smoke at the same time.

P14, L23: How much of this introductory material is actually necessary for introducing the two figures of the section, which don't actually answer the "big questions" posed beforehand? The beginning of this section sets up big expectations that aren't met.

We re-arranged the text as already mentioned.... The 'big questions' are just briefly mentioned in the discussion on the potential pathways of the smoke impact on ozone depletion...

Figures 15 & 16: These figures appear to me to be mostly redundant. Figure 16 is mostly a coarse-grained version of Figure 15 with additional information about the ozone anomalies. It seems like this information could easily be combined into a single figure.

Yes, we prepared one figure but did not find a solution to 'efficiently' combine the information in both figures. But having one figure now, it is easier to see the effects. We indicate better the different months J: January, F: February, M: March, in both figures, (a) and (b) to facilitate the study of both figures.

P16, L16-22: This paragraph and Section 4 (which has major issues) seems like a missed opportunity to provide expert guidance on how MOSAiC measurements could assist future studies that may attempt to answer such questions.

We think, Section 5 (the former Section 4) is sufficient to initiate, trigger, or stimulate further research. We are not experts for atmospheric chemistry, we are not members of the stratospheric ozone community, we are even not members of a well-organized stratospheric aerosol community. So we prefer not to stand up as a teacher to tell the pupil what to do next.