Dear Mike!

First of all thank you for your careful and detailed study of the manuscript and for the many, very 'direct' and constructive statements. We had your comments always in mind when we revised the manuscript. Before we answer your comments and questions step by step, we would like to briefly summarize what we did:

We went deeply into the literature with special focus on volcanic sulfate aerosol (originating from the recent Raikoke and from the Sarychev volcanic eruptions), and studied again many papers including the papers you suggested, we also re-checked our own Leipzig lidar data analysis, we again checked many CALIPSO observations and performed an extended HYSLPIT trajectory analysis and conducted further simulation studies (on self-lifting aspects) 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). The focus is on the potential self-lifting process, on smoke aging and on the consequences of slow self-lifting and thus efficient aging of the smoke particles for the morphological and optical properties of smoke. They become spherical before they reach the stratosphere and cause low depolarization ratios.

We did all this very carefully and even if we present a large number of reasonable and plausible arguments for our chain of hypotheses we emphasize that our entire set of arguments still remains at a hypothetical level.

Nevertheless, there is no doubt! What we observed during the MOSAiC expedition is clearly and unambiguously wildfire smoke. There is no way around. We observed this inverse spectral dependence of the lidar ratio (LR355 < LR532) together with the high LR532 around 85 sr, and that is a unique fingerprint of wildfire smoke! Note, that we observe such smoke fingerprints again and again, since 1998 (Wandinger et al., JGR, 2002). …. in Canadian, Siberian, Australian smoke, …

And one of the convincing and motivating arguments to search for a much more massive aerosol source than the Raikoke volcano was: The Raikoke-related AOD at high northern latitudes was about 0.01 at 500 nm in autumn 2019 (according to simulations of the 2019 Raikoke as well as of the 2009 Sarychev volcanic aerosol impact and corroborated also by Raikoke aerosol observations at lower latitudes, discussed in Kloss et al., 2021). But, we observed AODs around 0.1 in autumn 2019 in the High Arctic, and thus an order of magnitude higher values. We were forced to find a massive aerosol source.

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 (LR355nm < LR532nm) together with high LR532. 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 miss-classification 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. (2021), hereafter abbreviated as "O21," is a provocative study of upper troposphere, lower stratosphere (UTLS) aerosols at high northern latitude between fall 2019 and spring 2020. It is provocative in that O21 observe UTLS aerosol nearly daily in that temporal span and conclude that the composition is wildfire smoke, the source is fires in a sector of Siberia in July 2019, and the transport pathway to the UTLS is diabatic heating/lofting.

The manuscript is also provocative in that Raikoke volcano (Kuril Islands) erupted in late June 2019 and polluted the UTLS with a mass of SO_2 on par with or exceeding other eruptions that generated stratospheric clouds persisting for greater than a half year (E.g. Kasatochi, Sarychev Peak, Nabro, Kelut, Calbuco. See Solomon et al. (2016) for a tabulation.).

We checked the potential strength of the Raikoke sulfate aerosol in terms of AOD at 500 nm again, and we used especially the paper of Haywood et al (2010) for the Sarychev volcanic eruption (causing a very similar perturbation 10 years ago, in the summer of 2009), and came to the conclusion that the Raikoke-related AOD in October 2019 in the High Arctic was 0.01 to 0.015 at 500nm, but we observed 0.1, and thus an order of magnitude more, when we started the MOSAiC measurements! There is no doubt! We need a 'better' source for the aerosol we observed during MOSAiC. This is now clearly mentioned in the introduction. We must say, all the papers on Raikoke aerosol are a bit confusing because they provide observations of SO_2 (yes this is related to Raikoke) but then they show extinction coefficients (and this is related to EVERYTHING, smoke, sulfate, background aerosol etc…). And we must say, in many papers the authors obviously just did not know what they measured. And the authors were probably miss-guided by CALIPSO lidar observations and CALIPSO aerosol typing. In the CALIPSO data base, you will find sulfate layers only (if we neglect the few pyroCb smoke cases). We (at TROPOS, Leipzig) compared our own Raman lidar observations (14 August 2019) with an almost direct over flight by CALIPSO (14 Augsut2019, same time): We saw the same layer features as CALIPSO, we measured the same depolarization ratios, they classified the layer as sulfate aerosol layer (based on depolarization observations), and our lidar ratio observations ($LR355<LR532$, and high LR532) clearly told us: WILDFIRE smoke. CALIPSO is a 'simple' backscatter lidar and we use an advanced, state-of-the-art Raman lidar, and at the end, we have to defend our unambiguous observations (facts) and nobody asks: What may go wrong with the CALIPSO aerosol classification (assumptions) . Motivated by this mismatch, we introduce an extra subsection (Sect. 3.1) on the the miss-classification of smoke layers when using this quite simple CALIPSO aerosol typing scheme. Furthermore, we will write a paper on this topic (submission in September 2021).

If O21's conclusions are borne out, it will be a new insight into the polar UTLS and smoke transport to the UTLS. However, there is overwhelming observational evidence that Arctic UTLS in the second half of 2019 and early months of 2020 was blanketed by Raikoke sulfates.

The impression ….. 'overwhelming observational evidence' …. sounds strange in our ears. You mean you have rather clear arguments that all the aerosol we detected and measured in the stratosphere in 2019 … was Raikoke aerosol? We have clearly to state: We disagree! Our observations are in full disagreement with the 'overwhelming observation evidence'.

And concerning the 'evidence': In the Kloss et al. (2021) paper, for example, they show color-scaled OMPS-LP-derived AODs with high resolution up to 0.025 (ONLY) and all higher values are just given in the same color as for 0.025, simply in red. Why (?), because of saturation effects? This is a strange way of presentation, when keeping in mind that the AOD was obviously close to 0.1 in August to October over high northern latitudes (according to the Leipzig, Spitsbergen, and Polarstern lidar observations at 532 nm)! Furthermore, a rather inhomogenuous AOT distribution is shown by Kloss et al. with increasing AOT from low to high latitudes. This is in contradiction with model results which indicate a homogeneous Raikoke sulfate particle distribution from 45 to 90°N. The same was observed and modelled after the Sarychev eruption. Why was the volcanic sulfate aerosol this time (in 2019) inhomogeneously distributed from 45 to 90N according to the observations of Kloss et al., and after the Sarychev eruption it was homogeneously distributed, and thus in full agreement with the models? However, this inhomogeneous stratospheric aerosol distribution in 2019 is very reasonable when keeping the strong Siberian fires into consideration. But Kloss et al. (2021) totally ignored a potential contribution by the Siberian fire smoke.

Secondly, there is abundant evidence that the UTLS aerosol picture O21 describe over the July Siberia source sector is also dominated by Raikoke SO_2 and sulfates.

If the Raikoke sulfate aerosol was dominating (with an absolute maximum AOD of around 0.02 at 500 nm in mid August 2019 and later on with an AOD of the order of 0.01 in October 2019), how can we then measure AODs even exceeding 0.1? We observed stratospheric AOTs of 0.1 with lidar at Leipzig, at Spitsbergen, and at Polarstern in August to October 2019!!!! So, we clearly have a conflict, if we want to continue with our message: Raikoke aerosol dominated the stratospheric aerosol layer in 2019. The simple answer could be: All prediction of all the models are wrong, i.e., the Raikoke impact was totally underestimated. But then please tell us why the Raikoke effect on stratospheric AOD was about a factor of FIVE higher than the Sarychev volcanic AOD although the emitted SO2 amount was just 20 % higher in comparison with the Sarychev SO2 amount.

Thirdly, the lidar data
presented by O21 are more closely aligned with spherical sulfate droplets than smoke particles. These three points are elaborated on below.

Your phrasing suggests, stratospheric smoke particles are 'by law' nonspherical and thus produce significantly enhanced depolarization ratios. Yes, this is probably true for smoke particles reaching the stratosphere by the fast pyroCB-lifting processes, then the depolarization ratio is significantly higher than zero. The emitted fires retained their nonspherical shape when reaching the stratosphere. After 3-6 months even these smoke particles would have developed a perfect spherical form by smoke aging processes and the depolarization ratio would be close to zero (as can be found in Baars et al., ACP, 2019). In contrast: if the smoke particles are lifted slowly into the stratosphere, by self-lifting

processes, so that aging processes could be completed within the humid tropospheric environment (all this takes 2-4 days only) then the smoke particles have the chance to reach the stratosphere as spherical particles. And this seems to be the case here. The Raman lidar observations of the lidar ratios at 355 and 532 nm clearly and unambiguously indicated SMOKE (LR355<LR532, LR532 very high), and at the same time the depolarization ratio was close to zero (clearly indicating spherical particles). But now, we have a problem with the CALIPSO aerosol typing scheme, because such a case of smoke self-lifting is not considered in the CALIPSO aerosol typing scheme. As a consequence, if particles are spherical they are assigned automatically as sulfate aerosol particles.

All this is now explained in detail in the new Section 3.

<u>Point 1</u>

Kloss et al. (2021) show that the Raikoke volcanic cloud dominated the high-latitude northern hemisphere from eruption through the early months of 2020. I consulted Chris Boone, ACE-FTS Project Scientist and co-Principal Investigator, to help qualify the 2019/20 UTLS plume further. ACE not only delivers aerosol extinction profiles but profiles of SO_2 as well. These were combined by Cameron et al. (2021) in an examination of several UTLS volcanic events including Raikoke; the results further qualify those of Kloss et al. and show the strong presence of Raikoke SO_2 and sulfates at high northern latitude in summer and fall 2019. In addition, ACE IR spectra have been used to identify smoke aerosol in connection with ACE Imager extinction profiles (Boone et al. (2020)). The same technique was applied to high-latitude northern hemisphere 2019/20 ACE data while invoking published sulfate IR spectra for comparison. The findings are summarized as follows. In July 2019, the lower stratosphere in the latitude region near 60 degrees north is stuffed with sulfate aerosols from the Raikoke eruption. …

We can confirm this. Our lidar observation at Leipzig (52N) on 23 July 2019 of 355 nm lidar ratios of 45 sr (typical value for freshly formed sulfate particles) and depolarization ratios at 355 and 532 nm close to zero suggest non-absorbing, spherical sulfate aerosol. Unfortunately, the volcanic AOD was too low, of the order of 0.005…, so that we could not determine 532 nm particle extinction and lidar ratio values to that time (before August 2019, when the stratospheric aerosol increased strongly).

….. Identification of the composition is accomplished by looking at the infrared spectrum of the aerosols and noting the coincident enhancement of SO_2 and ACE Imager aerosol extinction layers. In September/October 2019, there is a blanket of aerosols in the lower stratosphere in the latitude region near 80 degrees north. ….

Coincidence (SO2 and enhanced extinction) alone is not a convincing argument to us. Again, we confirm that there was a lower stratospheric aerosol layer around 80N in September and October 2019 (by our Spitsbergen and Polarstern lidars). And we can add, the stratospheric AOD was close to 0.1 at 500nm, and thus a factor of 5-10 higher than the expected Raikoke AOD.

The blanket appears to be composed of Raikoke sulfate aerosols. …

…according to CALIPSO aerosol typing…, but not in agreement with our dual-wavelength Raman measurements of LR355<LR532 together with high LR532.

In February/March 2020, the aerosol blanket in the lower stratosphere near 80^{o}N is still present. …

Yes, again, we can confirm this and the layer was still 10 km in depth.

…. SO_2 has decayed. However, the spectra associated with the Imager aerosol layers are consistent with sulfate. At no point did we find any evidence of biomass burning smoke playing a role in these stratospheric aerosols (Boone et al., 2020). Detailed support for these findings is available upon request.

We are not sure whether these spectra contain accurate information on the aerosol type. 'Evidence' and 'consistency' sound convincing, but is that sufficient to make solid statements on the aerosol type? Is our solid fingerprint (the measurement of an inverse spectral behavior of the lidar ratio as an unambiguous fingerprint for smoke) not much more 'convincing'?

Point 2

O21 hypothesize that their MOSAiC Arctic 2019/20 lidar signals are dominated by an impressive build-up of UTLS smoke that began between ~24-28 July 2019 in a zone within Siberia centered roughly at 60^{o}N, 110^{o}E (Figure 2). They present nadir satellite imagery combining fire hot-spot data, true-color imagery, and aerosol optical depth retrievals to show an intensification of burning and smoke concentration. They also display a single CALIOP curtain on 26 July to characterize the vertical structure and ascent of smoke layers (Figure 3). There are several concerns regarding Figures 2, 3 and their interpretation, listed below.

It is simply not possible to know from a single CALIOP curtain if ascent is taking place; there is insufficient information from such a quasi-instantaneous vertical snapshot. Additional information must be brought to bear.

Yes, sure!

There are technical issues with Figure 3 and its caption. The date of the CALIOP image is given as "2016." This is obviously an inconsequential typographical error.

We improved this.

But more substantially, the latitude, longitude coordinates labeled along the bottom are wrong.

We improved this as well. We apologize for all these mistakes!

The curtain displayed is actually situated west of the Figure 2 boxed focal point. It is west of the labeled coordinates by approximately 25^{o} longitude. Compare Figure 3 with the actual orbital track and lidar data …

We now show in the new Figure 5 HYSPLIT backward trajectories, and indicate the CALIPSO flight track in this HYSPLIT map as a straight line and also show box 2 (with strong fires) defined in former Figure 2 (now Figure 3).

This is scientifically relevant for two reasons. One is that the vertical aerosol profile over the Siberia focus zone is unknown to the reader. Secondly, the aerosol profile on display is nominally upwind of the Siberia box, meaning that the history of those aerosols may be disconnected with processes occurring in the Siberia box.

The HYSPLIT backward trajectories in the new Figure 5 show that CALIPSO measured the smoke produced in box 2, because the track was downwind of box 2, and not upwind. Therefore, we are

convinced that the CALIPSO backscatter curtain plot provides an impression of the vertical distribution of smoke originating from the fires in box 2.

But is that essential? The main reason to include the CALIPSO figure is to show a convincing example: Yes, there was a lot of smoke and the smoke was everywhere up to the tropopause and even above the tropopause.

Indeed, it appears from the CALIOP curtains linked above that the "Ascending smoke plumes (mostly in green) are visible up to the tropopause at 10-11 km height as well as in the lower stratosphere…" [Quoted from Figure 3 caption] are mostly assigned as sulfate by the CALIOP version 4 aerosol subtype algorithm.

CALIPSO had no choice or chance…, as we already discussed: The depolarization ratio was close to zero, and then there is only one solution: this is sulfate aerosol!

One can see from CALIOP curtains just upwind of the Figure 2 Siberia box, on days leading up to the O21 smoke AOD buildup, an assignment of sulfate subtype to "mostly in green" backscatter in the UTLS. Here is an example from 20 July …. Similar scenes are found each day thereafter leading up to the 24-28 July O21 period of focus . This multi-day, broad, high-latitude swath of aerosols primarily defined as "sulfate/other" conforms to the findings of Cameron et al. and our deeper investigation into ACE July 2019 profiles near 60^{o}N.

As stated already, we confirm this finding: Our July 2019 lidar profiles at Leipzig provide clear signatures for volcanic aerosols. This is not surprising because the extreme fires in Siberia accumulated between 19 July and 14 August (according to the very nice paper of Johnson et al, 2021), and caused a huge jump in the AOT (in the troposphere as well as in the stratosphere) NOT before the beginning of August 2019. By the way, even Johnson et al. saw a lot of structures in the stratosphere over their field site in Alberta in August (and later on), but unfortunately they restricted their discussion on the backscatter and ozone profile observations in the troposphere (this information is obtained by personal communication).

We were a bit disappointed when evaluating the Cameron et al. paper because the most interesting month (August 2019) is missing, probably because of missing observations because of saturation effects as a result of too high particle extinction coefficients.

It stands to reason then, that if Raikoke sulfates are blanketing high northern latitudes at that time (Kloss et al., Cameron et al., and our investigation), they would also be evident over the Figure 2 Siberia box before, during, and after the hypothesized smoke uplift. The CALIOP example below shows aerosol subtype findings consistent with the prior examples on 20 July over the Figure 2 box….

Again, CALIPSO aerosol typing in July is ok, the rest is not ok to our opinion.

Hence, if the July 2019 Siberia-zone smoke is the initial condition for the O21 lidar observations in fall and winter, the presentation surrounding Figure 2 and 3 is insufficient to make that case.

Yes, such a presentation is insufficient and remains insufficient. We agree that the text in the submitted version of the paper was misleading. We gave the impression that would be able to offer a solid explanation. To keep the answer short: The goal of the new Section 3 (with the discussion of Figures 2 and 3 in the old version) is now to offer a plausible way and a lot of reasonable arguments that the Siberian fires were most probably responsible for the smoke we observed during the MOSAiC expedition. However, we clearly state that all this is based on several hypotheses. As given now more

precisely in the introduction, the motivation for the extended Section 3 on Siberian fires is simply: We observed a strong perturbation of the stratospheric aerosol layer by smoke and we realized that this perturbation is an order of magnitude stronger (in terms of AOD) than the Raikoke sulfate impact and we asked ourselves: What can cause such a strong impact on the stratospheric aerosol load? What was the source for this smoke? And the jump in stratospheric AOD toqrds 0.1 guided us to check the fire maps for July and August 2019.

Sure, there is no way at all to present a clear solution, that the smoke produced over Siberia was definitely (no doubt at all) the source of the smoke over the North Pole 3-6 months later on. Even, modelling and air mass transport analysis would not help. But we have the Spitsbergen observations from the beginning of August 2019 to January 2020, and thus a coherent link from the fire months (July and August) to October, and even an overlap (October to January) with the Polarstern lidar observations. All this is shown in Figure 12 in the revised manuscript.

<u>Point 3</u>

O21 acknowledge that the stratospheric aerosols detected by MOSAiC lidars were spherical according to aerosol depolarization measurements (Page 7, line 21). In principle, there seems to be less uncertainty as to the shape, composition, and depolarization of volcanic liquid sulfates in contrast to biomass burning smoke. Lidar measurements of tropospheric and stratospheric smoke converge on the idea that smoke is depolarizing (e.g. Burton et al., 2015; Fromm et al., 2008), suggesting some amount of asphericity. The first reported lidar observations of stratospheric smoke emphasized the unmistakable signal of depolarization (Siebert et al., 2000; Fromm et al., 2000). More recent stratospheric smoke papers, cited within O21, consistently report depolarization by smoke particles. Hence O21 would be establishing a new finding--stratospheric smoke with essentially no aerosol depolarization. Arguing for this peculiarity, O21 mention an aging process and a collapse of black carbon core. Neither process is described, and no publications are cited. Given the weight of evidence for the overarching presence of Raikoke UTLS liquid sulfates during the O21 reporting period and the MOSAiC depolarization results provided therein, the arguments for particle aging and black carbon core- shell collapse must be made more substantively.

This was another motivation to introduce the new Section 3. The first version of the text was six pages long to bring together all necessary information on particle aging, the influence of smoke aging on morphological properties and finally on the optical properties of smoke particles as given in Ansmann et al. (2021). But this would be too long in such a MOSAiC paper. So, we decided to provide a compact Section 3 only. All the published knowledge about stratospheric smoke (or better UTLS smoke) and resulting depolarization ratios measureable with lidar is linked to pyroCb-related smoke lifting. And this fast lifting of smoke by pyroCb convection prohibits particle aging and the development of a spherical shape before the smoke particles reach the stratosphere. They keep there nonspherical shape and thus produce significant depolarization of laser light. This is the present status of knowledge, and this is considered in the CALIPSO aerosol typing scheme, and this is the reason that any stratospheric aerosol layer with low depolarization ratio is classified as sulfate aerosol. In the Burton et al (2015) paper the smoke reached the dry upper troposphere, where favorable conditions (dry, no condensable gases) were given to slow down the particle aging process so that the particles remained nonspherical.

But now (in the case of the extreme Siberian fires) we had to introduce the self-lifting process. Otherwise there would be no smoke in the stratosphere, because as you told us (and we checked that also), there was no pyroCb development. And self-lifting takes some days, and this time is sufficient to finalize the aging process. And at the end of any aging of smoke particles is the spherical shape, the evolution of a perfect spherical shell indicated by the low depolarization ratios. And as we show in the

paper of Baars et al. (2019), even the stratospheric smoke is aging and reaches the spherical form, but this takes months… before the depolarization ratio goes down to zero again. By the way, we observed the same for the Australian smoke over Punta Arenas. In January and February 2020, the smoke depolarization ratio was significantly enhanced and then dropped immediately to zero in March-April 2020, at least at the lower heights of the stratosphere.

In conclusion, this is the first time that we have smoke in the stratosphere (and there is no doubt that is was smoke because of the invsere lidar-ratio spectral dependence) and that this smoke was not depolarizing. This is quite a very new aspect. And it is therefore not surprising that such a unique event is not considered in the CALIPSO aerosol typing scheme. But the other way around, it is impossible that the UTLS aerosol layer we observed consisted of sulfate particles. All the observed facts (lidar ratios, Angstroem exponent) would be in severe contradiction with this conclusion. And even the simple Mie models would be in trouble to produce lidar ratios of 55 sr at 355 nm and 85 sr at 532 nm for typical size distributions and sulfuric-acid water droplets.