Authors response to community comments by Dr. Farahnaz Khosrawi

We sincerely thank Dr. Farahnaz Khosrawi for the valuable comments. Our point-by-point response to the comments are given below. The comments are marked in bold blue font and our responses are marked in normal black font below each comment.

I have read your manuscript with great interest. While reading your manuscript I found several issues that were not clear as well as some technical issues which I provide you here in my comment. I think these will help to improve your manuscript.

Specific comments:

Title: After reading the manuscript I had the feeling that the title does not really fit to the content of the paper.

Thank you for your comment. The main objective of the study is to study the effects of aerosols from the 2019/2020 Australian black summer event on polar stratospheric clouds (PSCs). Our findings reveal that ice PSCs exhibited a significant and anomalous increase in areal coverage. We attribute this to increased stratospheric aerosol loading as revealed by Ozone Monitoring Profiler Suite measurement and by the condensation of HNO_3 on bushfire aerosols forming liquid-NAT mixture, which then rapidly transitioned into ice. Since this result, "enhanced formation of ice PSCs" is central to the paper, we believe the title reflects the most critical finding while remaining concise. We kindly request to retain the current title to highlight this key aspect.

L24: What is LNAT? I have never heard of it. NAT particles are solid particles, so why should these then be liquid? Do you rather mean supercooled ternary solutions?

Thank you for pointing it out. We changed the 'LNAT' to 'liquid-NAT mixture' following the terminology used in Pitts et al., (2018). By LNAT, we meant liquid-NAT mixtures. This is essentially a mixture of liquid supercooled ternary solution and solid NAT.

Pitts, M. C., Poole, L. R., & Gonzalez, R. (2018). Polar stratospheric cloud climatology based on CALIPSO spaceborne lidar measurements from 2006 to 2017. Atmospheric Chemistry and Physics, 18(15), 10881–10913. https://doi.org/10.5194/ACP-18-10881-2018

L59: Here you should add "e.g." and also add the references of Khosrawi et al. (2016) and Thölix et al. (2016).

Complied with.

L65: Title of subsection does not fit to the content. In this section also reanalysis data is described. Thus, this subsections should be renamed to "Satellite and reanalysis data"

Thank you for pointing out that. The subsection title is updated now into "Satellite and reanalysis data" at page no. 5, L134.

L119: "…a new methodology" Where is this methodology described? You should provide here a short description of the methodology.

Thank you for the comment. In In this revised manuscript, the methodology is Sect 3.3 from L183 in page no. 6 to L240 in page no. 7. In this methodology, to gain information about formation pathways of PSCs, we have examined the temperature history of the air parcels containing ice/liquid-NAT mixture through Lagrangian backward trajectory analysis along with corresponding changes in MLS HNO_3, and H_2O mixing ratio. Furthermore, as per the reviewer's suggestion, we fed these trajectories to the CLaMS microphysical box model, to simulate the PSC evolution and subsequent uptake of HNO_3, and H_2O and validated it against the MLS observed uptakes of these gases.

In addition, as suggested, instead of showing the results as average for the whole year and statistical analysis, in this revised manuscript, we presented results in the form of case studies along with results from CLaMS box model simulation. Totally 7 case studies have been added as described below:

- Liquid-NAT mixture formation pathways: 2 cases discussing ice-free NAT formation pathway (Case no. 1 in page no. 19 and Case no. 2 in page no. 22) and 2 cases discussing ice-assisted NAT formation pathway (Case no. 3 in page no. 24 and Case no. 4 in page no. 26).
- Ice formation pathways: 2 cases discussing NAT-assisted ice formation pathway (Case no. 5 in page no. 31 and Case no. 6 in page no. 32) and 1 case discussing NAT-free formation pathway (Case no. 7 in page no. 33).

The revised methodology is similar to Nakajima et al., (2016) and Voigt et al., (2018) who have studied the PSC formation pathways through investigating temperature history and CALPSO observed PSC type along the backward trajectories. We brief the methodology here below.

1. First, we choose the ice and liquid-NAT mixture PSCs from CALIPSO observation.

2. For these chosen PSCs, we calculated 48 h backward trajectories using CLaMS trajectories model and hourly ERA5 operational analysis meteorological data. The rationale behind choosing the '48 h' is that once the air parcel's temperature drops below TNAT and following the nucleation of NAT particles with a number density of 5×10-4 and 5×10-5 cm-3, within ~19 h (0.8 day) the NAT particles' perpendicular backscatter exceeds CALIPSO detection threshold and becomes detectable (Lambert et al., 2012). Similarly, Voigt et al. (2005) provided observational evidence from aircraft campaigns showing NAT formation within approximately 20 hours after the temperature drops below TNAT. In the case of ice formation, the 48 h period also should be sufficient (considering the average cooling rate of the stratosphere). To support for choosing 48 h, in this present study, we also provided observational

evidence of formation of liquid-NAT mixture and ice PSC within the 48 h once the temperature decreased below TNAT.

3. To determine PSC composition along each trajectory, we identify intersection points where the backward trajectory crosses the CALIPSO scan track within a ±30-minute window i.e., at the intersection points, both trajectory and CALIPSO profile time should be within ±30-minute window. At each valid intersection, the PSC composition is assigned from the CALIPSO profile with the closest potential temperature to the trajectory point. In addition, the MLS observed gas-phase HNO3, and H2O are filled along the trajectory at the time of observation of PSC from CALIPSO. This creates the comprehensive picture about temporal evolution of air parcel which leads to formation of ice/liquid-NAT mixture and help us to understand their formation pathways.

By carefully analysing backward trajectories of air parcels containing ice and liquid-NAT mixture PSCs, we retrieved their formation pathways, and relative percentage contribution of formation pathways for liquid-NAT mixture is given in Fig. 13 (c) (page no. 29 in the revised manuscript), and for ice in Fig. 19 (page no. 37 in the revised manuscript).

We also validated the MLS observed uptake in HNO_3, and H_2O against the CLaMS modelled uptake in these gases during formation of ice and liquid-NAT mixture.

These are incorporated in the revised manuscript, with the specific cases on pages pointed out above.

L160: This should rather read "occurrence" than formation.

Complied with.

L167: It is not clear how you get information on the formation pathways. You only get information on the occurrence of PSCs.

In response to the specific comment #5, we briefly described the methodology to retrieve formation pathways of PSC.

L174: Also here it should rather read "occurrence".

Complied with.

L214: It should rather read "chemistry" or "microphysics" than "dynamics".

Thank you for the comment. As there is no any evidence that bushfire aerosols changed the PSC chemistry (for e.g., chemical composition) and microphysical properties such as nucleation or growth rate of PSC. Hence, we would like to choose to use the word 'dynamics' to indicate the overall observed change in PSC, specifically areal coverage of various PSCs as a result of increased HNO_3 and aerosols due to the bushfire event.

L254: There are plenty of references for this statement, thus "e.g." should be added before the reference of Tritscher et al.

Complied with.

L273: References are missing here. There is a special issue on this winter in JGR/GRL and plenty of papers that discuss the vortex dynamics during this winter.

Complied with. We added references for the paper discussing vortex dynamics during 2020 winter at the L404 and L405

L278: Add "e.g." since these two references are not the only ones that could be cited here.

Complied with.

L289: Here ACE-FTS is used, but this data set has not been described in the method section.

Thank you for the comment. The description of ACE-FTS is given in page no. 5 and L144 to L146. The same is quoted here.

"Atmospheric Chemistry Experiment-FTS (ACE-FTS) onboard SciSat satellite provides trace gases mixing ratio by measuring limb absorption spectra and level 2, version 4.0 daily HF, H_2O, HNO_3, N_2O_5 mixing ratio (https://www.frdr-dfdr.ca/repo/dataset/c75d2c49-0def-49e5-9c69-5e74c824dc6c; Bernath et al., 2020) are used in the present study."

L289ff: I still do not understand how to read the correlations. Which part of the correlation refers to chemistry and which to dynamics? Could you mark the respective points in the plot?

Thank you for pointing out. We added a few more points between the L415 and L418 in page no. 14 and paraphrased the Fig. 4 caption in the revised manuscript. Further, we are explaining the same below.

The dynamic and chemical cause for the change in gas composition can be inferred from the scatter plot. For example, the data corresponding during 2020 (red diamond, in Fig. 4(a) in revised manuscript) are much deviated from the regression line made for the background period (blue circle, in Fig. (a)). It suggests that concentration of long-lived chemically inert species, HF does not change during this period. Therefore, the increase in HNO_3 should be due to chemical process. Similarly, during 2020, data corresponding to H_2O (red diamond, in Fig(b)) have not deviated much from the regression line. Hence, it suggests that H_2O is increased due to dynamical cause.

Figure 9 caption: No pathways of formation. To my understanding you solely look at the occurrence of specific PSC types. From CALIPSO data it is only possible to derive information on the occurrence of PSC types, not on the formation pathways. As stated above you have to better explain your methodology.

Thank you for the comment. We agree with the reviewer that CALIPSO provide the information about the PSC occurrence and type but not about the formation pathways. For this reason, we calculated the backward trajectory analysis of the air parcels containing liquid-NAT mixture and ice PSC to retrieve information about the formation pathways. We added the revised methodology to Sect. 3.3. from L183 in page no. 6 to L240 in page no. 7.

Figure 9 caption: percentage contribution to what? To all particles?

The NAT particle can form either nucleating on pre-existing ice (ice-assisted nucleation process) or on STS with solid foreign nuclei inclusion (ice-free nucleation process). The 'Percentage contribution' says, how much liquid-NAT mixture formed via ice-assisted nucleation process and how much via ice-free nucleation process.

L360ff: Here you mention Tice, but you have nowhere defined/mentioned at which temperatures the respective PSC particles form.

We thank the reviewer for pointing out. These details are added now at L86 to L91.

L381: Isn't that quite logical? Why should it be large NAT if it is obviously no PSC?

When CALIPSO classifies a grid as 'No Cloud', it essentially does not mean there is no physical presence of PSC. As Lambert et al., (2012) (in their subsection 4.2.1) mentioned, as optical properties of large NAT particles (radius > 6 µm) with very 'low number density' falls below the CALIPSO detection threshold, these types of particles are not detected by the CALIPSO but can be detected through HNO_3 depletion from MLS observation. Hence, even if CALIPSO classifies certain grid as 'No Cloud', it essentially does not mean there is no PSC but corresponds to NAT with low number density and large size.

Lambert, A., Santee, M. L., Wu, D. L., & Chae, J. H. (2012). A-train CALIOP and MLS observations of early winter Antarctic polar stratospheric clouds and nitric acid in 2008. Atmospheric Chemistry and Physics, 12(6), 2899–2931. https://doi.org/10.5194/ACP-12-2899-2012

L397: This is pure speculation. You cannot derive from your analysis any conclusions on the formation process, i.e. if it was homogeneous or heterogeneous.

As per the revised methodology, as brief in response to specific comment #5, we study the temperature history of the air parcel. Based on that, and also type of PSC which observed along the backward trajectory of the liquid-NAT mixture, we group the liquid-NAT mixture formation pathways into two: ice-assisted nucleation and ice-free nucleation as exists in literature. So, along the backward trajectory of liquid-NAT mixture, if NC/STS are observed along the trajectory and temperature not decreased 1.5 less than T_{ice} (temperature at which ice nucleates heterogeneously), we conclude that liquid-NAT mixture formed via ice-free nucleation pathways. In case, if NC/STS/ice are observed along the pathways, or the temperature of the air parcel decreased 1.5 less than T_{ice}, we conclude that liquid-NAT mixture formed via ice-assisted nucleation pathway.

L392: What about temperature fluctuations induced by waves? These are not resolved by the reanalysis data.

We acknowledge the view with thanks. We agree that ERA5 data does not resolve small scall fluctuation. For this reason, we are not discussing the formation of mountain wave induced ice, and enhanced NAT in the present study.

L411: I don't think that the solid kernel of a PSC can be detected by a lidar. They usually detect if the particle is generally liquid or solid. With their schemes they can characterize the type of PSC (NAT; ice is STS), but not if in the formation a foreign nuclei was involved.

Thank you for the comment. We understand that CALIPSO provides just PSC composition/type only but not formation process. Using the methodology which we described in subsection 2.3.1 in original manuscript, we noticed a few STS changed to liquid-NAT mixtures. This transition is possible if NAT is nucleated on the STS. Hanson and Ravishankara (1991; 1992) have shown through laboratory experiment that homogeneous nucleation of NAT on STS (i.e., STS without any foreign nuclei inclusion) is less likely for stratospheric condition. Furthermore, in general, homogeneous nucleation of NAT is kinetically suppressed as shown in Koop et al., (1995). Hence, it is highly probable that during the STS to liquid-NAT mixtures transition, NAT nucleated heterogeneously on STS (i.e., STS with solid nuclei).

Hanson, D. R. and Ravishankara, A. R.: The reaction probabilities of ClONO2 and N2O5 on 40 to 75% sulfuric acid solutions, Journal of Geophysical Research: Atmospheres, 96, 17307–17314, https://doi.org/10.1029/91JD01750, 1991.

Hanson, D. R. and Ravishankara, A. R.: Investigation of the reactive and nonreactive processes involving nitryl hypochlorite and hydrogen chloride on water and nitric acid doped ice, J Phys Chem, 96, 2682–2691, 1992.

Koop, T., Biermann, U. M., Raber, W., Luo, B. P., Crutzen, P. J., and Peter, T.: Do stratospheric aerosol droplets freeze above the ice frost point?, Geophys Res Lett, 22, 917–920, https://doi.org/10.1029/95GL00814, 1995.

L432: No, there could be temperature fluctuations by waves which are not resolved by the meteorological analysis.

We agree with the reviewer's view that small scale fluctuation by waves are not resolved in ERA5 reanalysis. Hence, we are not discussing the influence of the small scale temperature fluctuation on PSC formation pathways.

L444: As stated above. From lidar you cannot get any information in the nucleation process. Only information on the type (composition) of a PSC can be derived.

We agree with the view that CALIPSO provides just the information of PSC's presence and type, but not formation pathway. In response to the specific #5, we brief the methodology to retrieve the formation pathway. Please also kindly see the detailed the revised methodology added in the revised manuscript at Sect. 3.3. from L183 in page no. 6 to L240 in page no. 7.

L450: This is a contradiction. Why is it called liquid NAT if it is solid?

To avoid confusion, we changed the terminology ‘LNAT’ into ‘liquid-NAT mixtures’ as in Pitts et al., 2018. The liquid-NAT mixture is mixture of liquid-STS and solid NAT.

L469: chemistry should be replaced by composition since PSCs are not formed by chemistry.

Complied with.

L471: As also already stated above, the methodology has not been clearly explained and this needs to be improved to make this study more convincing.

In response to the specific #5, we brief the methodology to retrieve the formation pathway. Please also see the detailed the revised methodology added in the revised manuscript at Sect. 3.3. from L183 in page no. 6 to L240 in page no. 7.

L473: This is only speculation. I do not see any proof in your analysis for this.

Thank you for the comment. In response to the specific comment #5, we brief the revised methodology to formation pathways. Using that, in the revised manuscript, we discussed the formation pathway of ice and liquid-NAT mixture. Here, we discuss one of the cases briefly.

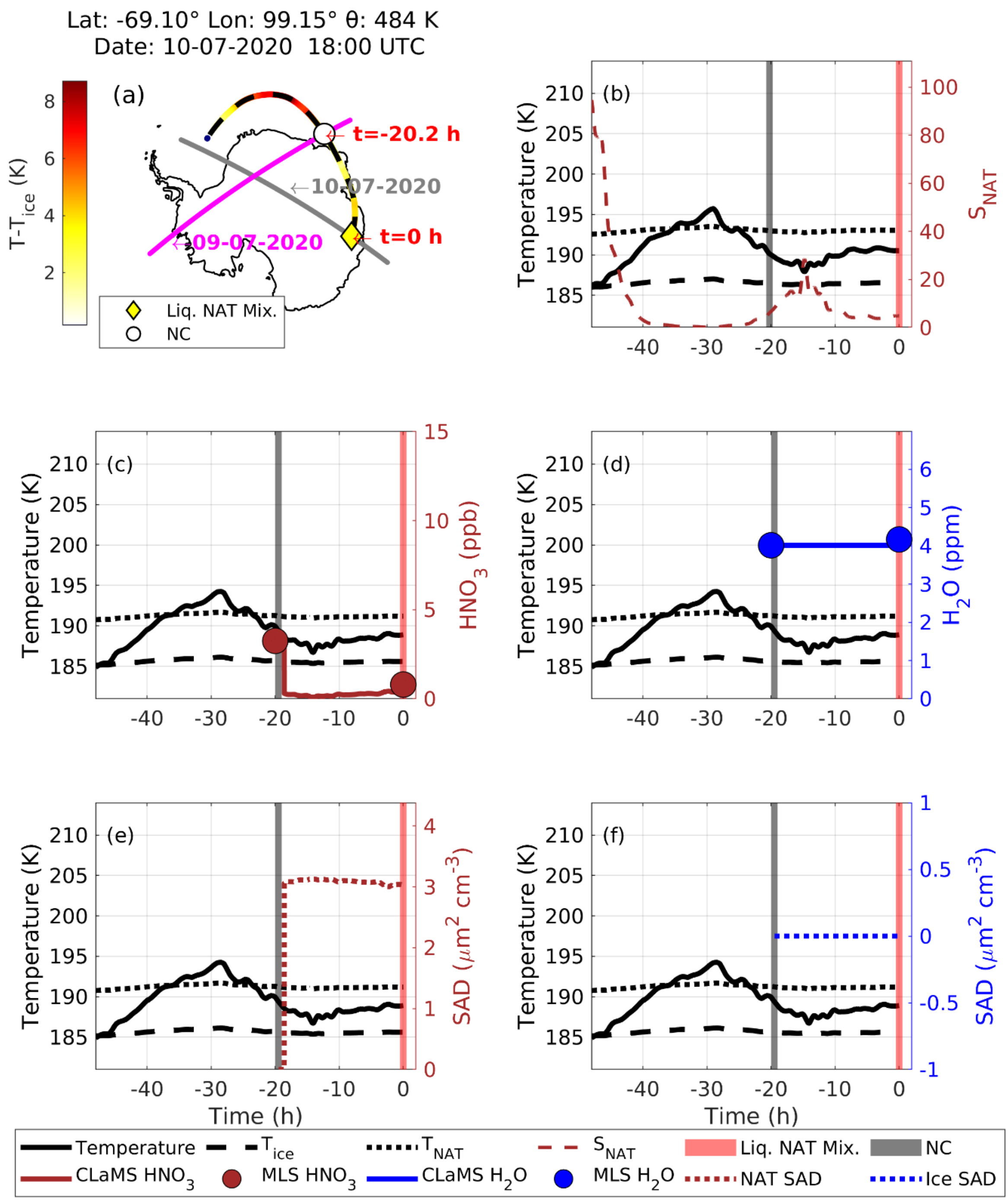

Figure 1. The lagrangian backward trajectory for a 48 h period starting at time, t = 0 h (corresponding to 18:00 UTC 10-07-2020) is shown. Here, the dashed black line is the backward trajectory and the color along this trajectory is the temperature at the T-Tice coordinate. The yellow diamond represents the observed liquid-NAT mixture from the CALIPSO scan track (solid grey line) corresponding to 10-07-2020. The complete coordinate of this liquid-NAT mixture is given in the title. The white circle represents the observed 'No Cloud (NC)' at the time, t = -20.2 h from the CALIPSO scan track (solid magenta line) corresponding to 09-07-2020. (b) shows the saturation ratio over NAT (S_{NAT}) (dashed brown line) and vertical bars mark the liquid-NAT mixture (red) and 'NC' (grey). (c) The brown circle marks the MLS HNO_3, and the solid brown line represents the CLaMS HNO_3. (d) The blue circle marks the MLS H2O, and the solid blue line represents the CLaMS H2O. (e) shows the NAT

surface area density (SAD) (dotted brown line). Panel (f) shows the ice surface area density (SAD) (dotted blue line).

On 10-07-2020, at 18:00 UTC, CALIPSO detected a liquid-NAT mixture at a latitude of -69.1° and longitude of 99.15°, with a potential temperature of 484 K. This observation is marked by a yellow diamond in panel (a) and corresponding CALIPSO scan track is shown as a solid grey line. The dashed black line in panel (a) represents the calculated 48 h backward trajectory of this PSC, with the color indicating the temperature history of the air parcel in ice coordinates. The temperature 'T' is obtained from ERA5 operational analysis, and T_{ice} is estimated using the ERA5 pressure, and mean MLS H_2O mixing ratio found along the trajectory following Marti and Mauersberger, (1993).

The backward trajectory reveals that CALIPSO observed 'No Cloud (NC)' along this trajectory 20.2 hours earlier (at the time, t = -20.2 h), on 09-07-2020, marked by a white circle in panel (a). The temperature history shows that between these two observations, the temperature did not decrease below the T_{ice}, indicating that the condition is not conducive for ice formation. At the time of the NC observation, the temperature is ~189 K which is 2 K below the NAT temperature (T_{NAT}). During this time, MLS observed gas-phase HNO_3 and H_2O mixing ratios are 3.5 ppb and 4 ppm, respectively (panel (c) and (d)). Using these as initial conditions, a CLaMS box model run was performed from t = -20.2 h to 0 h, simulating the evolution from the NC to the liquid-NAT mixture. After 20.2 hours, the MLS HNO3 decreased from 3.5 to 0.5 ppb, with no significant change in MLS H_2O. The CLaMS modeled uptake of HNO_3, and H_2O agreed well with the MLS observations (panel (c) and (d)). Furthermore, the CLaMS box model run indicates that the NAT surface area density (SAD) increased to nearly 3 μm^2 cm^{-3} (panel (e)), while the ice SAD remained at 0 μm^2 cm^{-3}, confirming that no ice formation occurred before the observation of the liquid-NAT mixture. During the transition from 'No Cloud' (NC) to the liquid-NAT mixture, the saturation ratio over NAT stayed well below 30, further supporting the absence of ice involvement in the formation of the liquid-NAT mixture (panel b) (Luo et al., 2003; Voigt et al., 2005). It should be noted that, as the liquid-NAT mixture means the mixture of liquid STS and solid NAT, STS PSC should have formed between the observation of NC and liquid-NAT mixture and specifically before the formation of NAT.

L480: Stratospheric chemistry not shown and discussed in this study. However, a changed PSC occurrence will definitely affect stratospheric chemistry.

Thank you for the comment. We discussed the change in stratospheric trace gases in subsection 3.2, and their cause in subsection 3.3. The subsequent subsections discuss the influence of these changes on PSC occurrence. However, the change in stratospheric chemistry due to the change in PSC occurrence is not discussed. We acknowledge the reviewer view on this, and it will be helpful to extend the study in future.

L487: Based on which measurements?

Using Microwave Limb Sounder (MLS) measurement. We added this information in the L1085.

L491ff: High HNO3 and H2O and aerosol are not the sole reason for enhanced PSC occurrence. Also the temperatures need to be sufficiently cold. Was this a cold or warm winter? Other studies have not shown any influence so far.

We agree that temperature also needs to be sufficiently cold for enhanced PSC occurrence. But Fig. 3d and Fig. 4d show that, there is no significant negative temperature anomaly for 2020 winter i.e., the temperature during this period is comparable to the background period (2012-2019). But strong positive anomaly in HNO_3, and aerosol is observed. Hence, it is likely that increased HNO_3, and aerosols are primary reasons for enhanced PSC occurrence.

L494: You cannot make any statements on the formation mechanism. This is only guessing and should be more carefully expressed.

We understand the concern regarding the formation pathways retrieval. We revised the methodology and strengthened the discussion section to support the findings. Please kindly see the revised methodology section 3.3.

Technical corrections:

L36: Semicolon between "2020" and closing parenthesis obsolete.

Complied with.

L41: add "in the abundance" (or "the amount") so that it reads "changes in the abundance of various trace gas species".

Complied with.

L44: rephrase sentence.

Complied with.

L54: paramount -> change wording

Complied with.

L71: trace gases mixing ratio -> trace gas mixing ratios

Complied with.

L73: add "s" -> mixing ratios

Complied with.

L225: to the -> to a

Complied with.

L271: tracer-trace -> tracer-tracer

Complied with.

L276: Tracer-Tracer -> Tracer-tracer

Complied with.

L279 and 280: tracer gas -> trace gas

Complied with.

L285: Tracer-Trace -> Tracer-tracer

Complied with.

L312: and aerosol aging -> and "the" aerosol "ages" or "is aging".

Complied with.

L317: by -> to

Complied with.

L321: and discussed -> and is discussed

Complied with.

L317: add "on sulfate aerosols" so that it reads "results in the condensation of these trace gases on sulfate aerosols".

Thank you for the suggestion. We changed the L457 to "results in the condensation of these trace gases on stratospheric aerosols" as the condensation of the trace gases are not limited to the sulfate aerosols but also on other aerosols like meteoritic dust, volcanic ash, soot, or H_2SO_4 hydrates.

Figure 8 caption: Move "monthly mean" before "areal coverage" so that it reads "monthly mean areal coverage" or better write directly "monthly averaged areal coverage".

Complied with.

Figure 10 caption: redline -> red line

Complied with.

L497: anticipated -> expected

Complied with.

References:

Khosrawi, F., Urban, J., Lossow, S., Stiller, G., Weigel, K., Braesicke, P., Pitts, M. C., Rozanov, A., Burrows, J. P., and Murtagh, D.: Sensitivity of polar stratospheric cloud formation to changes in water vapour and temperature, Atmos. Chem. Phys., 16, 101–121, https://doi.org/10.5194/acp-16-101-2016, 2016.

Thölix, L., Backman, L., Kivi, R., and Karpechko, A. Yu.: Variability of water vapour in the Arctic stratosphere, Atmos. Chem. Phys., 16, 4307–4321, https://doi.org/10.5194/acp-16-4307-2016, 2016.