Author response to Reviewer #2 (Report #1) comments

We sincerely thank the reviewer for the valuable comments. Based on these we have made careful revisions, which has helped in improving the manuscript. Our point-by-point response to the review comments are given below. The comments are marked in bold blue font and our responses are marked in normal black font below each comment.

Reviewer #2

I felt that the authors have made a great effort to revise the manuscript. The quality of the paper has improved a lot after the revision, and the authors answered most comments which the reviewers raised. I think the paper is almost ready to be accepted, after some technical points, which are shown below, are corrected.

Thank you for your kind appreciation of our work and the efforts we have made in addressing the review comments.

< Specific Comments >

1) P.3, L.66: In the title of Section 2, "Polar stratospheric clouds" should be "polar stratospheric clouds" (lowercase).

Complied with.

2) P.3, L.70: making up 3-15%: this number doesn't match the previous 20-100 tons per day.

Thank you for pointing it out. The sulfur emission of 160 tons per day from the troposphere results in 650 tons per day production of aqueous sulfuric acid in the stratosphere. And extraterrestrial material influx of 20–100 tons per day contributes 3–15 % of total weight to the stratospheric aerosols i.e., 100x(20 tons/650 tons) = 3 % and 100x(100 tons/650 tons) = 15 %

Now, we have rewritten the corresponding lines as follows in page no. 3: L71 to L74

"*It receives an influx of 160 tons of sulfur per day from the troposphere, which corresponds to the production of 650 tons per day of aqueous sulfuric acid (Thomason and Peter, 2006). In addition to the sulfate aerosols, meteoritic dust particles of extraterrestrial origin contribute 20–100 tons per day (Cziczo et al., 2001), making up 3–15% of the total mass of stratospheric aerosols*"

3) P.4, L.106: Hoyle et al. 2013 --> Hoyle et al. (2013)

Complied with.

4) P.4, L.115: lidar on board, the HALO --> lidar on board the HALO

Complied with.

5) P.6, L.184: After the title: (i) Selection of ice and liquid-NAT mixture PSCs, insert a new line.

Complied with. A new line is inserted.

6) P.6, L.195; measured between 12:56 --> measured between 12:55 ?

Complied with. The time is corrected now.

7) P.7, L.229: by Hanson and Mauersberger (1988) --> by Marti and Mauersberger (1993)

Complied with. The citation is updated now.

8) P.10, L.291: discussed in Sect. 3.3 --> discussed in Sect. 4.3

Complied with.

9) P.13, L.361: discussed in Sect. 3.2 --> discussed in Sect. 4.2

Complied with.

10) P.14, L.377: from the Black Summer --> from the black summer

Complied with.

11) P.14, L.381: (Figure 7 caption) PSC monthly mean areal --> PSC areal

We have changed "PSC monthly mean areal coverage" to "PSC areal coverage"

12) P.14, L.385: methodology Sect. 2.3 --> methodology Sect. 3.3

Complied with.

13) P.14, L.388: Peak positive anomaly of up to 4 x 10^6 --> Peak positive anomaly of up to 6 x 10^6

The absolute value of ice PSC areal coverage is approximately 6×10^6 km^2 during second week of August. But the magnitude of increase in ice PSC areal coverage with respect to the background mean is 4×10^6 km^2.

14) P.15, L.398: Fig. 7 (e andf)) --> Fig. 7 (e and f))

Complied with.

15) P.15, L.416: case no.5 and 6 respectively --> case no.5 and 6, respectively

Complied with.

16) P.25, L.621: by (Luo et al., (2003) --> by Luo et al. (2003)

Complied with. The bracket before "Luo et al" has been removed.

17) P.35, L.828: 13.86 % --> 14%

Complied with.

18) P.35, L.829: 38.02 % --> 38%

Complied with.

19) P.35, L.831: 43.42 % --> 43%

Complied with.

20) P.35, L.832: 4.69 % --> 5%

Complied with.

21) P.35, L.840-844: Delete all the references here.

Complied with.

22) P.36: L.853: liquid-NAT mixture pathways --> liquid-NAT mixture PSCs

Complied with.

23) P.36, L.872: 13.86 % --> 14%

Complied with.

24) P.36, L.873: 38.02 % --> 38%

Complied with.

25) P.36, L.875: 43.42 % --> 43%

Complied with.

26) P.36, L.877: 4.69 % --> 5%

Complied with.

27) P.37, L.905: and Dr. Dr. Farahnaz --> and Dr. Farahnaz

Complied with. Thank you for pointing out the mistake.

Author response to Reviewer #4 (Report #2) comments

We sincerely thank the reviewer for the valuable comments. Based on these we have made careful revisions, which has helped in improving the manuscript. Our point-by-point response to the review comments are given below. The comments are marked in bold blue font and our responses are marked in normal black font below each comment.

Reviewer #4

The title claims that an injection of aerosol in the polar stratosphere, produced by bushfire in Australia, has an impact on the formation of ice PSCs. This is similar to the impact of aerosol released after volcanic eruptions. The authors then compare the observations in 2020 by OMPS and CALIOP with respect to the preceding years, defining anomalies and standardized anomalies (=anomalies divided by the standard deviations in the parameters). I wonder why the authors don't include the impact on STS PSCs as well in the title. Figure 7 clearly shows also positive anomalies for STS.

We agree with the reviewer's view. We have modified the title as follows.

"Australian Bushfire Emissions Result in Enhanced Polar Stratospheric Clouds"

They do not explain why NAT show predominantly negative anomalies.

The observed negative anomaly of the NAT is possibly due to two reasons: (i) conversation of NAT particles into ice PSCs by acting as ice nuclei, as shown in Fig. 14c in the revised manuscript, and (ii) severe denitrification which occurred during June and July 2020, as can be seen in Fig. 3 (a, c).

It would be also interesting to show the anomalies of all PSC types and of the early onset with respect to other years.

We have included the anomalies of 'all PSCs' in the supplementary section (Fig. S5) in the revised manuscript, as shown below.

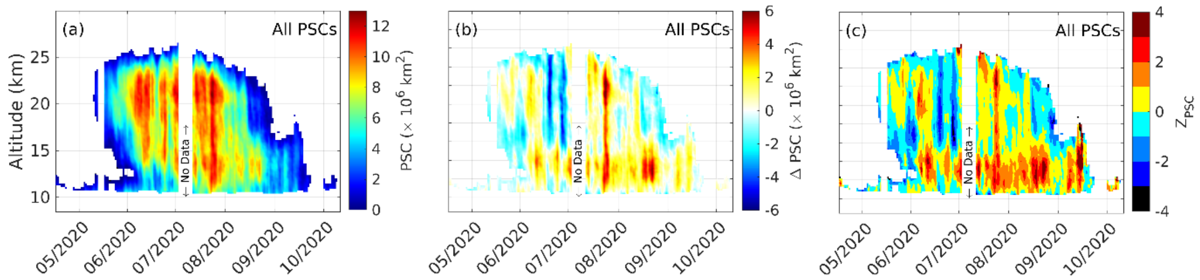

Figure. R1. CALIPSO Antarctic PSC areal coverage (panel a), anomaly (panel b), and standardized anomaly (panel c) for the year 2020.

For the onset of PSCs, we have included a table (Table S1) in the supplementary section which shows the onset of each PSC type from 2009 to 2020 (excluding 2015). The onset of each PSC is taken as the first day of their detection by CALIPSO. We did not observe an early onset of any PSCs during 2020. However, the onset of STS occurred on 26th May 2020, which is slightly later than the usual onset period of STS. Apart from this, there may be sub-visible PSCs which are not detected by CALIPSO, as the optical properties of those PSCs are below the CALIPSO detection threshold. Hence, the actual onset would be 1 to 3 days earlier than these dates. The sub-visible PSCs are often studied through the depletion of HNO_3 during early winter. Hence, we used MLS HNO_3 to check whether there is an early/late depletion in HNO_3, leading to an early/late onset of PSCs during 2020, at an altitude of 20 km (Fig. S4 in revised manuscript). The same plot is shown below in Fig. R2.

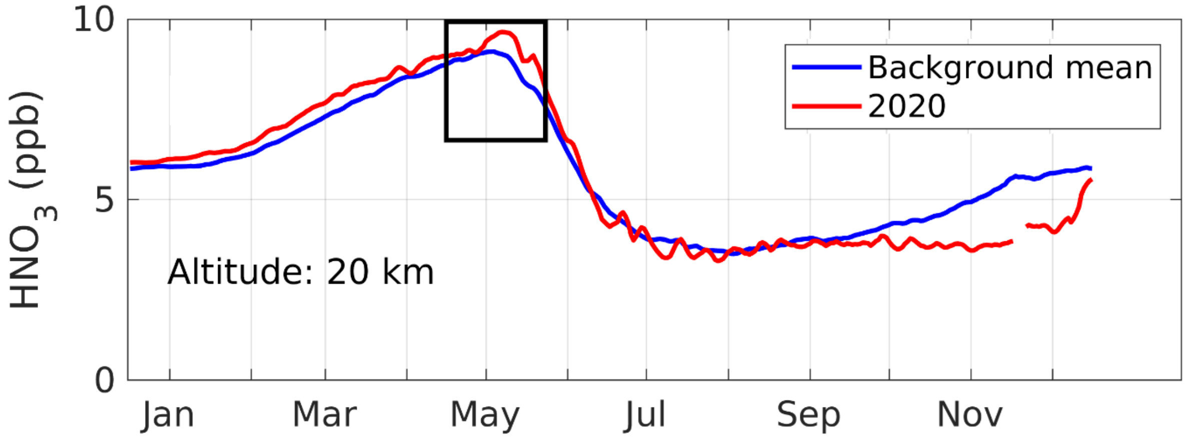

Figure R2. MLS obtained daily mean HNO_3 mixing ratio (ppb), averaged over the high latitudes (60°S to 90°S), is shown for the year 2020 (red line), and the background period of 2009-2019 (except 2015) (blue line) at the altitude of 20 km. The x-tick marks the middle of each month. The black rectangle box shows the period when depletion in HNO_3 occurs, leading to PSC formation every year.

The above figure shows that the depletion of HNO_3 occurs during the post-mid period of May every year at an altitude of 20 km and the pattern remains unchanged for the year 2020 as well, suggesting no signs of an early depletion of HNO_3. As discussed in Sect. 4.2 in the revised manuscript (Fig. 3a), the HNO_3 mixing ratio has increased by 1.5 ppb relative to the background mean within the altitudes from 20 to 30 km during 2020. In general, the NAT equilibrium temperature (T_{NAT}) is directly proportional to the gas-phase HNO_3 mixing ratio. Hence, the high availability of HNO_3 likely increased T_{NAT}, leading to the formation of PSCs at a relatively warmer temperature. To check the degree of impact of this increased HNO_3 on T_{NAT}, we used the theoretical gas-phase HNO_3 uptake model, assuming the thermodynamic equilibrium conditions for NAT. The uptake model revealed that under normal lower stratospheric conditions, withHNO_3 of 10 ppb and H_2O of 5 ppm at the pressure of 50 hPa (altitude ~20 km), T_{NAT} is 195.75 K. When the HNO_3 mixing ratio is increased by 1.5 ppb (as happened during early 2020), the T_{NAT} increased to 195.95 K i.e., an increase by 0.2 K. During May 2020, the mean T_{NAT} estimated using MLS-observed HNO_3, and H_2O mixing ratio is 195.5 K, which is 0.2 K higher than the background mean of 195.3 K, and consistent with the uptake model results (plot not shown). Similarly, an increase in the equilibrium temperature of STS by 0.2 K was also observed through both uptake models (Carslaw et a., 1995) and MLS observations.

The observational evidence from MLS HNO_3 and CALIPSO PSC suggests that there is neither an early depletion of HNO_3 nor an early onset of PSCs. Furthermore, the increase in T_{NAT} and T_{STS}, as a result of the high availability of HNO_3, is not significant. This suggests that even though the PSC equilibrium temperature increases owing to the increased production of HNO_3 through the N_2O_5 hydrolysis process on the bushfire aerosols, it is not significant enough to cause an early onset of the PSCs.

In the revised manuscript, we have included the interannual variations in the volume of all PSC types (Fig. 7b, in the revised manuscript). It shows that when we look at the PSC volume of 'total PSC' (i.e., a sum of all PSCs) during 2020, it is comparable to the background period. But if we check for each PSC type, the highest volume was recorded for ice, enhanced NAT mixture, and STS PSC during 2020 (shown below in Fig. R3).

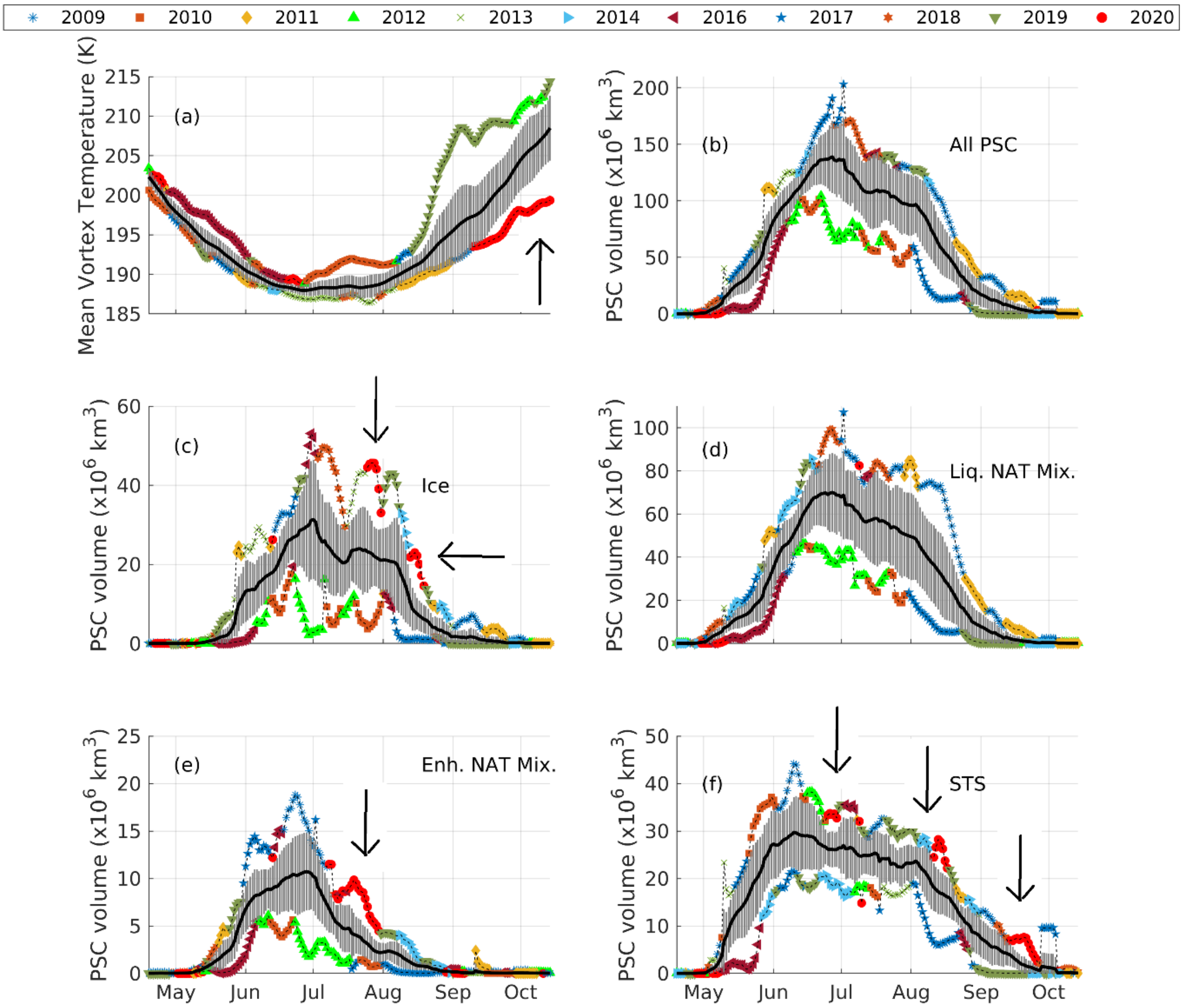

Figure R3: Time series of PSC volume obtained from CALIPSO over the Antarctic region from 2009 to 2020 (excluding 2015). Panel (a) shows the mean vortex temperature obtained from ERA5 operational analysis at a potential temperature of 450 K, and panels (b to f) show the volume of all PSC, ice, Liq. NAT Mix (Liquid-NAT mixture)., Enh. NAT Mix (Enhanced NAT mixture), and STS (Supercooled Ternary Solution). The black solid line and vertical lines represent the corresponding mean and standard deviations respectively. The daily maximum and minimum values are color-coded according to the year in which they occurred. The periods where anomalous high (low) value in PSC volume (vortex temperature) were observed in 2020 are marked with black arrows.

In terms of total PSC volume (i.e., the sum of the volume of all PSC types), there is no anomalous behavior in all PSC volume is observed which is also reported by Li et al., (2024). However, upon investigating the individual PSC volume, we found that ice, enhanced NAT and STS PSC exhibited anomalously high volume for certain periods during 2020. The period 8th to 14th August 2020 and 27th August to 9th September 2020 recorded the highest ice PSC volume among the years from 2009 to 2020, reaching up to 45×10^6 km^3 and 22×10^6 km^3 respectively. It suggests that the anomalous ice PSC areal coverage observed during the same period (shown in Fig. 6b, in the revised manuscript) is beyond the interannual variations. Similarly, the year 2020 recorded the highest enhanced NAT PSC volume during 21st July to 30th July 2020. For the STS PSC, the second week of July and the last week of August 2020 recorded the highest PSC volume. However, liquid-NAT mixture PSC volume did not exhibit significant anomalies relative to other PSC types.

Reference:

Li, D., Wang, Z., Li, S., Zhang, J., and Feng, W.: Climatology of Polar Stratospheric Clouds Derived from CALIPSO and SLIMCAT, Remote Sens., 16, 3285, https://doi.org/10.3390/rs16173285, 2024

Then a long discussion follows about formation processes. This is not included in the title, although it occupies most of the paper.

In response to this comment and the previous comment on the titles, we have updated the title.

The authors have done a lot of work to describe the presence of PSCs after the black summer 2020 in Australia. However, their analysis should also take into account interannual variations of the vortex temperatures as well.

Thank you for the suggestion. In the revised manuscript, we have studied the vortex dynamics using the ERA5 operational analysis product and compared it with the interannual variations in PSC volume. For this purpose, the essential statistics on the volume for each PSC type, observed from 2009 to 2020, along with their relationship with polar vortex dynamics, are studied in detail and discussed in Fig. 7,8,9 (sub-section 4.4.1 on Page 17 L448 to L555) and Fig. S6 in the supplementary section. We provide the brief of this analysis below.

The polar vortex is defined as the region enclosed by the 32 PVU (Potential Vorticity Unit) potential vorticity contours. The vortex area is quantified as the total area within this defined region, while the vortex temperature represents the mean temperature over the same area.

Figure S6 (panel a) from the supplementary section shows that during most of the days from late June to early August 2020, the Antarctic polar vortex area is recorded to be the highest, ranging between 23×10^6 km^2 and 32×10^6 km^2. However, this is not the coldest period, as revealed by the mean vortex temperature. We checked the role of this increased polar vortex area during 2020 to the increased PSC coverage by investigating the anomaly in the vortex occurrence frequency at a potential temperature level of 450 K and PSC volume from June to August 2020. For this purpose, a positive anomaly in the vortex occurrence frequency greater than one standard deviation with respect to the background period (single hatched region in Fig. R4) and negative anomaly greater than one standard deviation (cross-hatched region in Fig. R4) are overlaid on the monthly PSC volume anomaly.

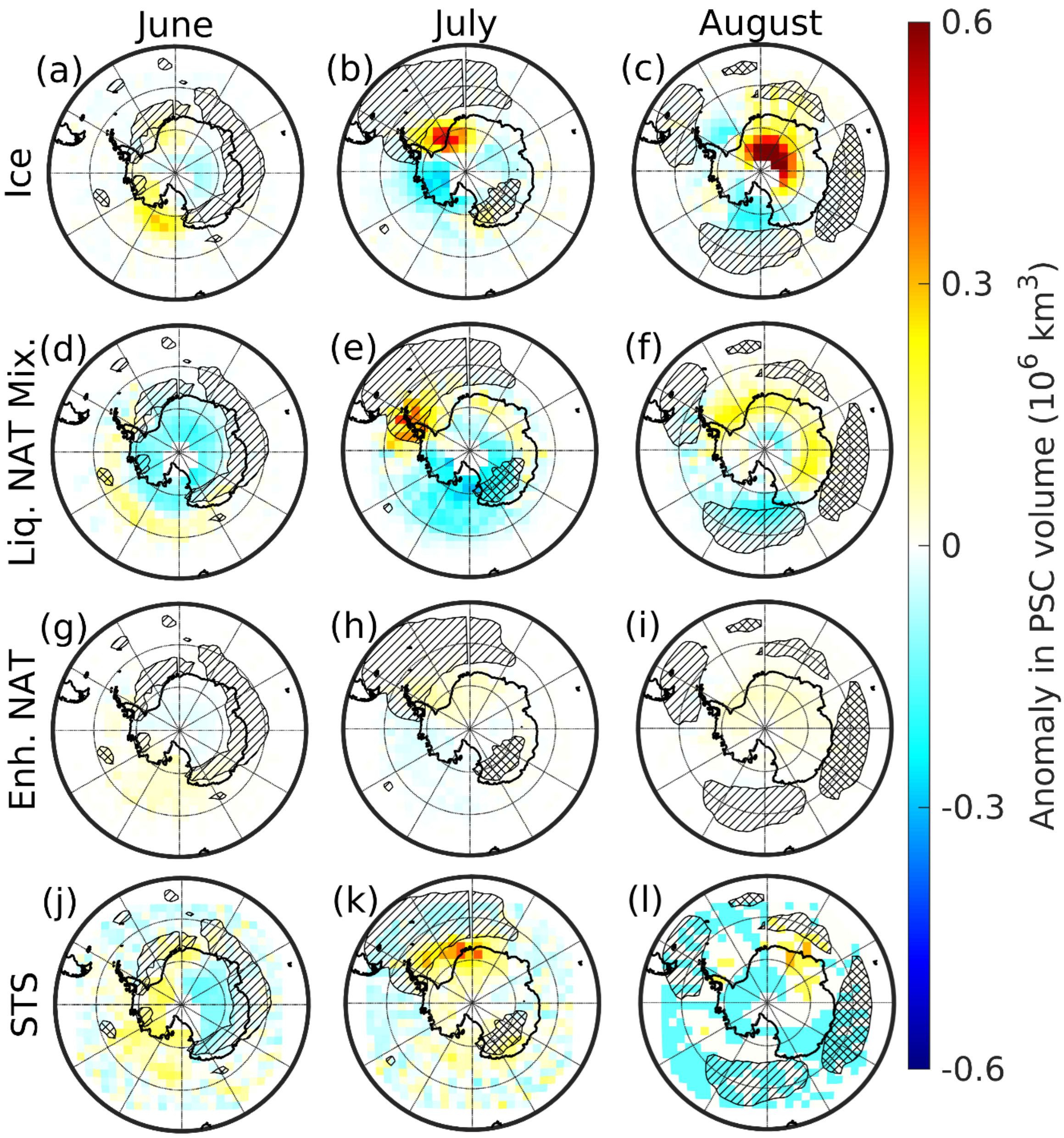

Figure R4: PSC volume anomalies for June (left column), July (middle column), and August (right column) are shown. Single-hatched regions indicate a positive anomaly in polar vortex occurrence frequency exceeding one standard deviation, while cross-hatched regions indicate a negative anomaly exceeding one standard deviation.

During June 2020, the vortex occurrence frequency exceeded one standard deviation over the eastern Antarctic and northeastern regions (marked with single hatching in Fig. R4, panels in the left column), indicating a significant expansion of the polar vortex in these areas. However, no positive anomalies in any PSC types were observed over the same region, suggesting that the increased vortex area during June 2020 did not influence PSC formation (Fig. R4, left column). In July 2020, the polar vortex shifted off the South Pole toward the Antarctic Peninsula, resulting in a significant positive anomaly in vortex occurrence frequency over the northwestern region (marked with single hatching in Fig. R4, middle column) and a negative anomaly over the southeastern region (marked with cross-hatching in Fig. R4, middle column). Over the Antarctic Peninsula, a positive anomaly in liquid-NAT mixture PSC volume slightly overlapped with this stretched vortex area, while no other PSC types exhibited anomalous behaviour (Fig. R4, middle panel). By August 2020, the vortex expanded further, stretching toward the Antarctic Peninsula while compressing over the eastern Antarctic region (Fig. R4, right column). Although a significant increase in PSC volume was observed during this month, particularly for ice and liquid-

NAT mixtures, these PSCs remained confined close to the South Pole. This again suggests that the increased polar vortex area did not influence PSC formation or contribute to the observed positive anomalies in PSC volume.

After this, we investigated the role of vortex temperature in PSC anomaly. For this purpose, we analyzed the periods during 2020 when the volume of each PSC type reached its highest decadal values and compared them with the corresponding vortex temperature, as shown below in Fig. R5.

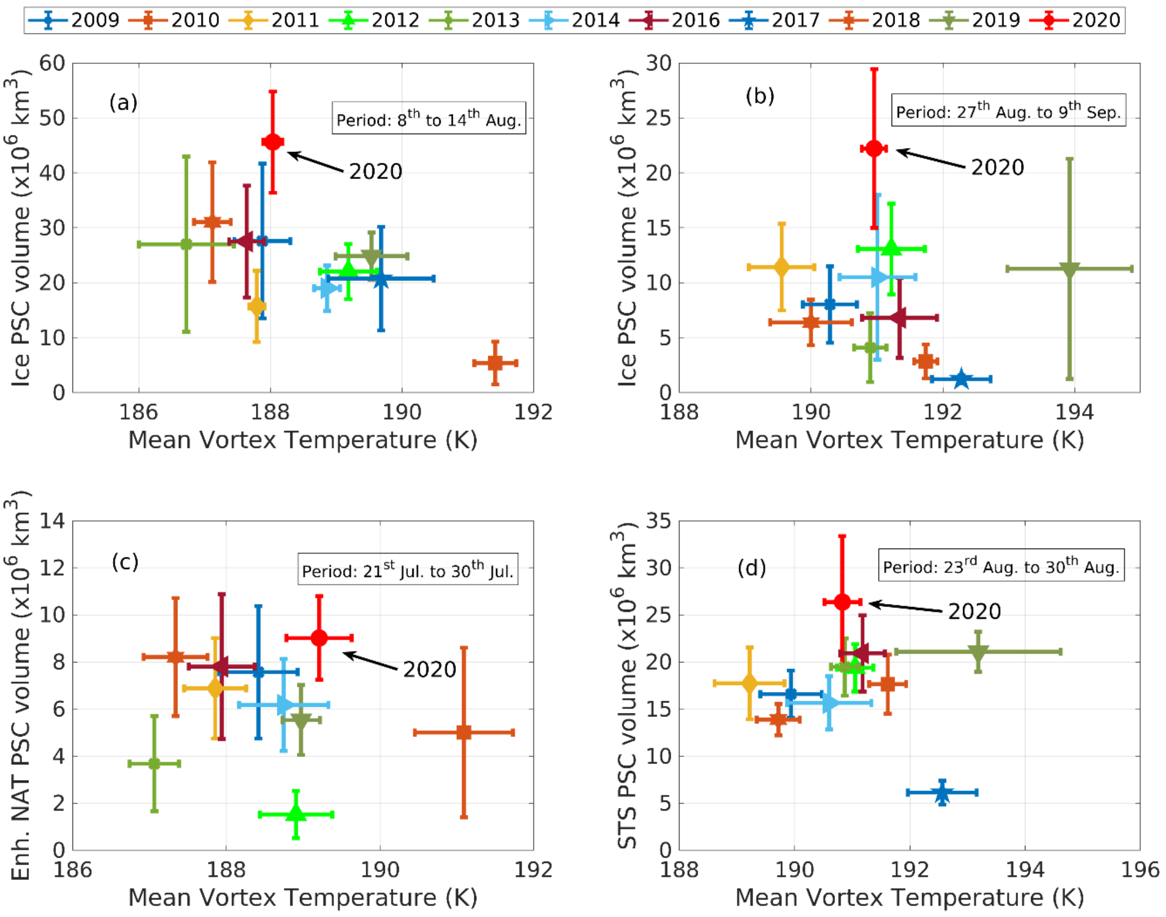

Figure R5: The relationship between mean vortex temperature and ice (panels a and b), enh. NAT (panel c), and STS (panel d) PSC volume are shown. The center of each marker represents the mean value, while the vertical and horizontal lines indicate one standard deviation of PSC volume and mean vortex temperature for each period (shown in the inset) respectively.

We compared the mean vortex temperature with the ice PSC volume during 8th to 14th August 2020 (Fig. R5a) and 27th August to 9th September 2020 (Fig. R5b), enhanced NAT mixture during 21st to 30th July 2020 (Fig. R5 c), and STS during 23rd to 30th August 2020 (Fig. R5 d) with previous years. The mean vortex temperature from 8th to 14th August 2020 is ~188 K which is considerably warmer than previous years (such as 2011, 2016, 2017, 2018, and 2019). Despite this relatively warm condition, the mean ice PSC volume is observed to be 46×10^6 km^3 (Fig. R5 a) which is significantly higher than in previous years. Similarly, from 27th August to 9th September, the mean vortex temperature is ~191 K. Once again it is not the coldest period when compared to years from 2009 to 2019. Yet, the mean ice PSC volume peaked at 22.5×10^6 km^3, which is two-fold higher than the previous recorded values. Similar observations are made in the case of enhanced NAT mixture and STS PSC volume (Fig. R5c, d), where a significant increase in their respective PSC volume is observed despite the vortex temperature being comparable to the previous years. This analysis indicates that the increased PSC areal coverage and volume cannot be attributed to the vortex dynamics, such as the variations in vortex area or temperature. Instead, we attribute these anomalous high PSC areal coverage and thus their volumes to the intrusion of bushfire aerosols from the black summer event.

The findings from this analysis are listed below:

1) **Polar vortex area:** The polar vortex area during June, July, and August 2020 is the largest in the years from 2009 to 2020 (excluding 2015). However, this additional vortex area did not contribute to additional PSC volume, i.e., the region where the vortex enlarged did not exhibit a significant positive anomaly in the volume of PSC.

2) **Polar vortex temperature:** Despite having a large area, the vortex temperature is not the coldest but comparable to previous years. However, anomalous increase in ice, enhanced NAT mixture, and STS PSC volumes are observed in some periods during the Austral winter of 2020. It suggests that the increased PSC volume is not attributed to the vortex temperature.

3) Since the increased PSC areal coverage and volume cannot be attributed to vortex dynamics, we attribute these anomalous high PSC areal coverage (and thus their volume) to the intrusion of bushfire aerosols from the black summer event.

The synthesis of previous works on the formation processes is not accurate and citations are obviously not correct (see below).

Thank you for the suggestion. We have updated the references related to the literature review of the PSC formation process.

The approach of studying many cases with Lagrangian backward trajectories is interesting, although some serious doubts remain concerning the CLaMS box model. It is not clear why the H2O gas phase as observed by MLS is not in agreement with the box model. It might be good to add some more information about the box model and the input parameters of the model.

In the CLaMS box model run, for simplicity, other chemical reactions (e.g., N_2O_5 hydrolysis, HCl null cycles) which involve H_2O are not considered, as discussed in Sect. 3.4 in the revised manuscript. Hence, even if such reactions occur and deplete/produce H_2O in the real atmosphere, the CLaMS box model run does not account for them. This simplified box model run likely contributes to the weak correlation between H_2O from CLaMS and MLS observations. A similar reasoning could apply to HNO_3; however, since the HNO_3 depletion is primarily driven by the NAT formation, the omission of other chemical reactions in the simplified box model does not significantly affect the correlation between the HNO_3 mixing ratio from CLaMS and MLS.

But in the case of ice formation, the correlation between H_2O from CLaMS and MLS has improved (R^2 = 0.57; Fig. 19 in the revised manuscript). This is because the H_2O depletion is primarily driven by ice formation in this case and CLaMS is able to simulate the ice formation. This has been added in the revised manuscript on page no. 31, L764 to L771.

More details about the CLaMS model run are added to Sect. 3.4 (Page no. 7, L238 to L260).

It appears that the box model is mainly used to assign a degree of confidence to the main formation pathways.

The main objective of the box model run is to test the reliability of the methodology employed to retrieve the formation pathways, as suggested by Reviewer #1 (report #1). The good agreement between the MLS observed uptakes of HNO_3, and H_2O increases the reliability of the methodology and thus the retrieved pathways. Therefore, the box model run is not used to assign the degree of confidence of the formation pathways, but the methodology used to retrieve them in the present study. The degree of confidence of each formation pathways are assigned as follows:

For liquid-NAT mixture formation pathways: The degree of confidence is made using the minimum temperature of the air parcels along the trajectories (obtained through the CLaMS trajectory module). The trajectories are estimated using CLaMS trajectory module with ERA5 operational analysis as input. In addition to estimating the trajectories, the CLaMS trajectory module also interpolates the ERA5 temperature along the trajectory coordinates. Hence, it is more appropriate to say that the degree of confidence for liquid-NAT mixture formation pathways is made using the ERA5 temperature history.

Ice-assisted pathways with high confidence: Direct CALIPSO observations of ice PSC along the backward trajectory starting from liquid-NAT mixture and temperature of the air parcel does not exceed the T_{ice}.

Ice-assisted pathways with low confidence: In this case, there is no direct observation of ice by CALIPSO, but ice could have possibly formed as the temperature of the air parcel decreased 1.5 K below T_{ice} before the observation of liquid-NAT mixture along the backward trajectories.

Ice-free pathways with high confidence: In this case, neither a direct observation of ice by CALIPSO nor the temperature of the air parcel decreased 1.5 K below T_{ice}, before the observation of liquid-NAT mixture.

For ice formation pathways: Here, the presence of liquid-NAT mixture and availability of MLS HNO_3 along the backward trajectory of ice PSC (i.e., before the formation of ice) is used to assign a degree of confidence.

NAT-assisted pathways with high confidence: In this case, there is direct CALIPSO observation of liquid-NAT mixture along the backward trajectory of the ice PSC and temperature of the air parcel does not exceed T_{NAT}.

NAT-assisted nucleation process with medium confidence: Here, there is no direct observation of NAT, but significant availability of gas-phase MLS HNO_3 above 3 ppb during the observation of NC or STS along the backward trajectory suggests the possible formation of NAT before ice formation.

NAT-assisted formation with low confidence: In this case, MLS HNO_3 mixing ratio during NC or STS observations lie between 0.5 and 3 ppb. While NAT formation cannot be ruled out, the lower HNO_3 mixing ratio suggest only a low likelihood of the involvement of NAT. Hence, we classified these types of cases to NAT-assisted nucleation with low confidence.

NAT-free formation with medium confidence: In this case, there is no direct observation of NAT along the backward trajectory of ice PSC. In addition, a very low MLS HNO_3 (<0.5 ppb) is observed along the backward trajectories, creating conditions where NAT formation is unlikely, and ice must have formed without the presence of NAT. Collectively, CALIPSO, MLS, and ERA5 data are used to assign a degree of confidence for the different formation pathways.

In conclusion, the presented results are interesting by themselves, but:

• the title does not cover the contents of the paper

We have updated the title to accurately cover the contents of the paper.

• the description of the formation processes and citations of previous literature is not accurate

Thank you for pointing out. We have added more relevant references in the revised manuscript along with a detailed description of the formation process.

• the CLaMS box model should be better described

More details are added to the Sect. 3.4 in the revised manuscript, where an introduction to the CLaMS and inputs to the CLaMS box model run are explained (Page no. 7, L230 to L243).

• interannual variations of the polar vortex should be included in the discussion

The interannual variations of the polar vortex and PSCs have been included in the discussion of the results in the revised manuscript (sub-section 4.4.1 on Page 17 L448 to L555).

• many questions remain unanswered (e.g. why NAT has negative anomalies, why the H2O concentration calculated with the CLaMS box model does not agree with MLS)

The reason for the NAT negative anomaly can be found in the response to the second comment. The reason for the disagreement in H_2O concentration between CLaMS and MLS can be found in the response to an earlier comment concerning the same issue.

• the discussion of the formation processes is presented as a general study, but it is valid only for the 2020 data and might be different for other years. As this might be the case, the findings of this paper might be only relevant for the 2020 data. The authors might want to comment on this. I recommend a major revision of the paper.

This study identifies the dominant formation pathways contributing to the formation of liquid-NAT mixtures and ice PSCs, quantified in terms of the relative percentage contributions of various pathways. The magnitude of the percentage contribution may also have interannual variations similar to the polar vortex and PSCs. But we believe that the main picture about the formation pathways (i.e., the pathway which contributes the most in forming ice/liquid-NAT mixture) remains the same for other years too. In the case of NAT formation pathways, our analysis revealed that the majority of the liquid-NAT mixture formed through ice-free formation pathways. This is logical given the fact that the NAT formation temperature is higher than the ice formation temperature and the high abundance of solid stratospheric aerosols. Even though bushfire aerosols have increased the abundance of stratospheric aerosols, they did not alter the formation temperature of PSCs itself. Hence, the results on NAT formation pathways can be applicable to other years.

Our analysis revealed that the ice formation pathways also depend on the availability of gas-phase HNO_3 i.e., under extremely denitrified scenarios, ice PSC forms through NAT-free formation processes, and under normal conditions, ice forms through NAT-assisted pathways. Since denitrification happens every year and there is no highly anomalous denitrification scenario observed during 2020, the results on ice formation pathways also can be extended to other years.

Some remarks:

Line 83 : always exist in the solid state, maybe better “Nat are solid particles….”

Thank you for the suggestion. Changed to “The Nitric Acid Trihydrate (NAT) are solid particles with a proportion of one HNO_3 and three H_2O and form at T_{ice} + 4 K.”

Line 84: Tice +7 K is an equilibrium temperature, formation may occur below this temperature, usually NAT forms 3-4 K below TNAT.

Complied with.

Line 89: “But the solid PSC like NAT grows large in size, typically > 10 μm”. Actually a NAT radius of about 1 micron is quite usual, large PSCs are seldom observed, due to their low number density.

Thank you for pointing it out. These lines are now removed from Section 2.1 to reduce the length of the introduction section as suggested by reviewer #5. Please also see the authors' response for reviewer #5 (report #3).

Line 95 : the term kinetically suppressed was not found in Koop, 1995 … Koop’s paper is mainly about STS droplets, and claims slow nucleation rates for homogeneous freezing. The authors should better explain what they mean with kinetically suppressed.

By ‘kinetically suppressed’, we mean the slow nucleation rate of the NAT. We used the term kinetically suppressed following Tritscher et al., (2019). In Koop et al., (1995), under the section “Application to the Stratosphere”, the authors argued that NAT could not form by nucleating on ternary solution due to a slow nucleation rate. But when the temperature drops below the frost point, ice forms first and it acts as nuclei for HNO_3 hydrate or NAT i.e., the formation of NAT requires solid nuclei.

References:

- 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.
- Tritscher, I., Pitts, M. C., Poole, L. R., Alexander, S. P., Cairo, F., Chipperfield, M. P., Grooß, J. U., Höpfner, M., Lambert, A., Luo, B., Molleker, S., Orr, A., Salawitch, R., Snels, M., Spang, R., Woiwode, W., and Peter, T.: Polar Stratospheric Clouds: Satellite Observations, Processes, and Role in Ozone Depletion, Reviews of Geophysics, 59, e2020RG000702, https://doi.org/10.1029/2020RG000702, 2019.

105 “Ground-based lidar observations by Biele et al. (2001) provided evidence of the existence of such STS with foreign nuclei inclusion” I did not find anything in Biele et al (2001) which confirms this statement. The paper is mainly dealing with mixtures of NAT and liquid droplets. It refers to a possible nucleation of NAT on ice [Koop et al.. 1995].via heterogeneous nucleation on hitherto unknown nuclei [Drdla, 1993]

Thank you for pointing it out. We are sorry for that mistake and have rectified it in the revised manuscript.

113 eliminate “the”

Complied with.

121 the black summer event

Complied with.

123 the percentage

Complied with.

136 through a

Complied with.

138 study.

141 .. eliminate one .

Complied with.

142 The

Complied with.

143-144 clouds (Pitts et al 2007).

Complied with.

150 “The total areal coverage of the PSCs such as liquid─NAT mixtures, STS, Ice, enh. NAT and MWI contribute to 48 %, 24.7 %, 21.4 %, 5.8 %, and 0.1 %”. This is not a universal truth, it depends on where and when. Strong interannual variations have been observed in PSC coverage. (see e.g. Pitts 2009)

Thank you for raising this point and we agree with this comment. The percentage contribution of each PSC to the total PSC is based on the climatology. As, there will be interannual variation, this value is not absolute. We have clarified this point in page no. 5 (L155) in the revised manuscript.

155 PSCs PSCs

Complied with.

157 5 km long the flight track, 300 m perpendicular to the track

Complied with.

177 The CLaMS model is introduced without any reference. (e.g. McKenna et al., 2002),

Complied with, by adding references and an introduction to the CLaMS model in Sect. 3.4 (page no. 7 L238 to L260) in the revised manuscript.

202-204 :” 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 to 5×10-5 cm-3, within ~19 h (0.8 days) the NAT particles exceed CALIPSO perpendicular backscatter threshold and becomes detectable (Lambert 2012). “ In Lambert 2012 no such statement can be found….

Thank you for pointing it out. The correct reference should be Lambert et al., (2016), and has been corrected in the revised manuscript.

226 The CLaMS microphysical box model is mentioned without any further information. It would be desirable to know which parameters have been used in the model.

Complied with. Under Sect. 3.4, we have added the parameters used for the CLaMS box model run to page no. 7 L238 to 260.

3.4 Not well explained

Following the reviewer's suggestion, Sect. 3.4 ((page no. 7 L238 to L260), has been revised carefully. We sincerely believe that it will enhance clarity about the box model run.

Figure 7. How is the standard deviation calculated for the monthly mean areal coverage?

Daily mean areal coverage is shown in Fig. 6. But in the caption, it was mistakenly written as monthly mean. We apologize for the error and have corrected it in the revised manuscript. The standard deviation for the daily mean areal coverage is estimated for the period 2009 to 2019 (excluding 2015).

One should bear in mind that there is a large interannual variation of the PSC coverage, meaning that several years would have a significant anomaly with respect to the averaged values.

We agree that PSC could exhibit large interannual variations. We have added a new sub-section 4.4.1 on Page 17 L448 to L555. In this section, we have discussed the interannual variation of PSC and polar vortex in detail. The brief about the discussion is also given in earlier response.

How significant is the anomaly of 2020 with respect to the anomalies of other years?

We have added a new sub-section 4.4.1 on Page 17 L448 to L555. This section discusses the interannual variability of PSC providing a comprehensive picture of anomalies of each PSC type.

How much of this anomaly can be attributed to the increased aerosol load and how much to other factors, such as the temperature?

Thank you for raising this interesting question. With the available satellite and reanalysis data, we are able to quantify the PSC anomalies during 2020. But to delineate the cause of these anomalies to aerosols and temperature, rigorous modelling studies are required. We feel that it would be beyond the scope of this paper but will be considering to answer this question in the future studies.

408 4.5. Formation pathways of liquid–NAT mixture and ice PSC

Here 6 case studies are discussed. How representative are these 6 cases ?

We considered a total of 8,841 trajectories. Out of these, 3792 are backward trajectories of the liquid-NAT mixture and the rest 5049 belong to ice. For NAT formation pathways, 2996 trajectories correspond to the liquid-NAT mixture formed through the ice-assisted formation pathway and 796 trajectories to the ice-free formation pathway. For ice formation pathways, 4796 trajectories correspond to the ice formed through the NAT-assisted formation pathway and only 253 trajectories to the NAT-free formation pathway. The discussed five cases (case no. 2 is moved to a supplementary section in response to the reviewer's suggestion) are representative of 8841 trajectories.

In the revised manuscript, we added the above information on page no. 7 L228 to L229, page no. 33 L826 to L829, and page no. 41 L978 to L980

Author response to Reviewer #5 (Report #3) comments

We sincerely thank the reviewer for the valuable comments. Based on these we have made careful revisions, which has helped in improving the manuscript. Our point-by-point response to the review comments are given below. The comments are marked in bold blue font and our responses are marked in normal black font below each comment.

Reviewer #5

The paper by Prasanth et al. on the influence of Australian bushfire aerosols on the formation of polar stratospheric clouds has improved significantly. The authors have addressed all previous reviewer questions and incorporated suggested methods to enhance the data analysis, such as examining the temperature history along the Lagrangian backward trajectory and incorporating the CLaMS microphysical box model into the analysis. The new methodology is thoroughly explained, with case studies added for each PSC formation pathway discussed in the paper. In essence, the updated paper is almost like a new manuscript.

I appreciate that the authors took the reviewers' concerns seriously and improved the paper considerably. I recommend its publication in ACP.

We sincerely thank the reviewer for the comments and for appreciating our efforts made in addressing the review comments.

My only suggestion would be to integrate Section 2 into the introduction, while also condensing to shorten Section 2.

Thank you. We have condensed Sect. 2.1 while ensuring the key points remain intact. Section 1 provides a literature review of the black summer event, while Sect. 2 focuses on the literature regarding PSC, especially its formation pathways. Given that these sections address distinct topics, we humbly feel that keeping them separate would provide more clarity to the reader. We would hence raise a request to let us retain them as separate sections.

The paper is quite long, maybe some of the case studies could be moved to the supplement.

Thank you for the comment. We have moved the NAT formation pathway case no. 2 to the supplementary section (Fig. S8). The case no. 2 discusses the 'ice-free formation pathway of liquid-NAT mixture' scenario where STS precedes the liquid-NAT mixture along the backward trajectory. We have discussed a similar scenario through case no.1 (Page no. 22, L585 to L627), where NC precedes the liquid-NAT mixture. Hence, moving case no.2 to supplementary section still retains the valuable key points that we make through this study.

We have already also moved another case of 'NAT-free ice formation pathway' to supplementary (Fig. S16) during the second submission. We believe moving these to the supplementary has reduced the length of the manuscript but retains the essential portions required to discuss the different formation pathways.

Author response to Reviewer #3 (Report #4) comments

We sincerely thank the reviewer for the valuable comments. Based on these we have made careful revisions, which has helped in improving the manuscript. Our point-by-point response to the review comments are given below. The comments are marked in bold blue font and our responses are marked in normal black font below each comment.

Reviewer #3

The paper by Srinivasan Prasanth and colleagues addresses a very interesting topic: The impact of bushfire events on Antarctic PSC formation, specifically the impact of the extreme Australian bushfire event in 2019/2020 on the occurrence of Antarctic PSC in 2020. The study "aims to investigate the anomalies in stratospheric chemistry and PSC dynamics caused by the Black Summer event" and attempts to recover and quantify PSC formation pathways. After reviewing the first version of the manuscript, I have come to the conclusion that this study cannot be published. I see and appreciate that a lot of work and effort has been put into the second version and that many suggestions for improvement have been implemented. However, I still have problems with this publication. The following comments from me are one-sided. I will focus my critic on the PSC part since this is my expertise. However, even if I only consider this part, I recommend to reject the paper for the following reasons:

Thank you for summarizing our manuscript.

• If the authors want to quantify PSC formation pathways, they first need to do a proper literature review of what is already known. As mentioned in my first review, I have the impression that the authors themselves are not fully aware of all PSC background literature.

Taking cue from the review comments, we have revised Sect.1 (introduction), Sect.2 (literature review), and the discussion of the results, which we believe has strengthened the manuscript.

• In my opinion, the second part about PSC nucleation pathways is not well connected to the Australian bushfire event. The authors show that there is an enhancement of PSC observations in the Antarctic winter of 2020 compared to the "background mean". Later, they try to explain/determine PSC formation pathways in general. They could do the analysis for any winter.

We appreciate the reviewer's view. Despite the high availability of HNO_3 during 2020, liquid-NAT mixture did not exhibit any anomalous behaviour whereas ice PSC did. To explain this observation, we made a hypothesis that majority of the ice PSC was formed via nucleating on NAT particles. Hence, the entire analysis on the PSC formation pathway is carried with an objective to check our hypothesis. We believe this methodology to retrieve formation pathways can be extended to other years too but would be beyond the scope of this paper.

• A possible attempt to connect these two parts could be Figure 7. Figure 7 shows that there is an enhancement in PSC observations compared to the "background mean". Looking at the seasonal evolution of the Antarctic PSC areas observed by CALIPSO for more than one year (e.g. Li et al., 2024, https://doi.org/10.3390/rs16173285), we can see that there is also quite a large variability from year to year in Antarctica, and that 2020 was not outstanding in terms of PSC area coverage. Is the "background mean" a useful parameter for comparison?

We studied the interannual variation of PSC and a detailed discussion has been added to the revised manuscript in sub-section 4.4.1 in Page 17 L448 to L555, as shown below.

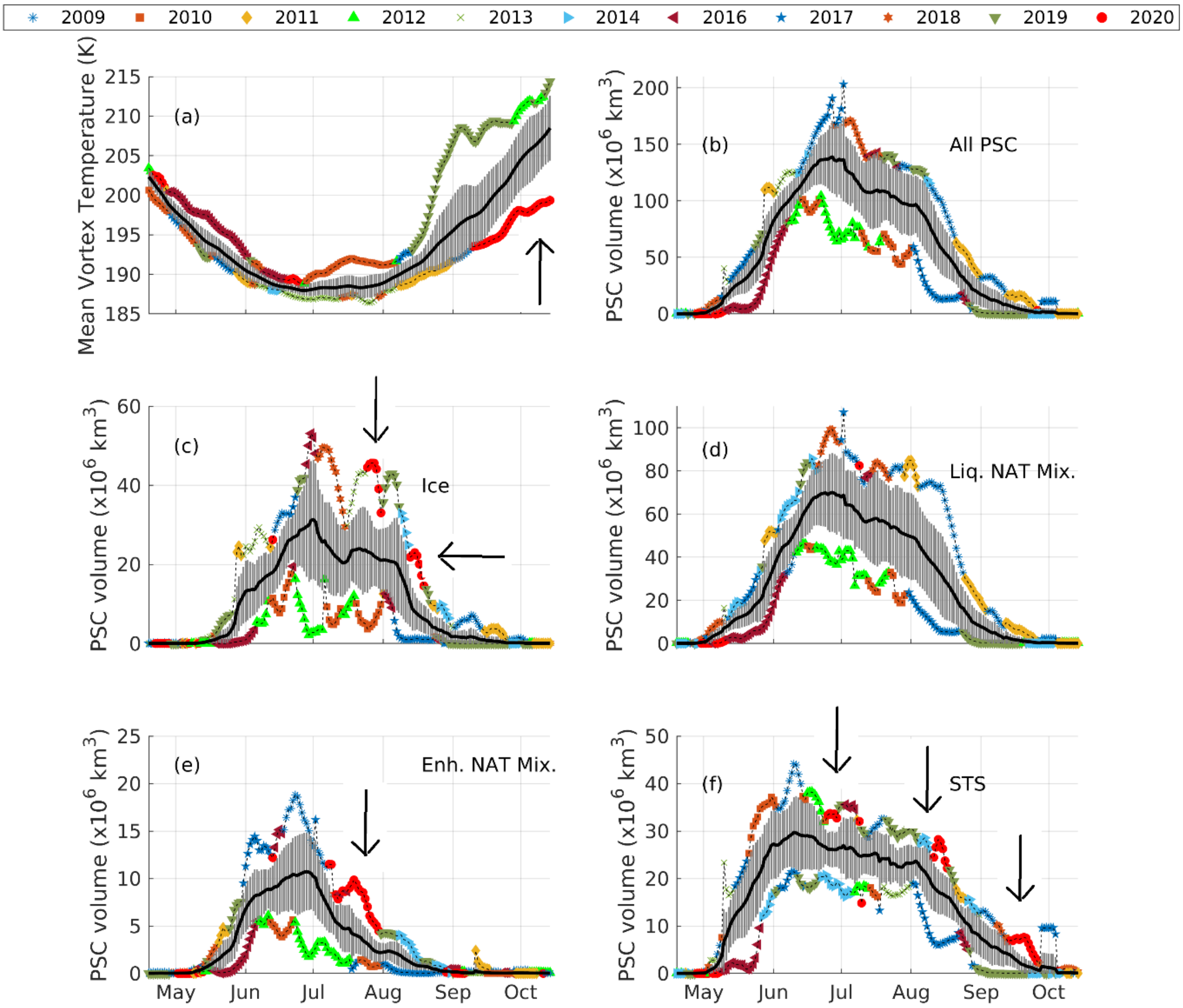

Figure R1: Time series of the CALIPSO obtained PSC volume over Antarctica from 2009 to 2020 (excluding 2015) are shown. Panel (a) shows the mean vortex temperature obtained from ERA5 operational analysis at the potential temperature of 450 K, and panels (b to f) show the volume of all PSC, ice, Liq. NAT Mix., Enh. NAT Mix, and STS. The black solid line and vertical lines represent corresponding mean and one standard deviation respectively. The daily maximum and minimum values are color-coded according to the year in which they occurred. The periods where anomalous high (low) values in PSC volume (vortex temperature) during 2020 are marked with black arrows.

In terms of all PSC volume (i.e., a sum of the volume of all PSC types), there is no anomalous behavior observed in all PSC volume, as has also been reported by Li et al. (2024). However, upon investigating the individual PSC volume, we found that ice, enhanced NAT, and STS PSC exhibited anomalously high volume for certain periods during 2020, exceeding one standard deviation as well as remaining recorded highest. The period 8th to 14th August 2020 and 27th August to 9th September 2020 recorded the highest ice PSC volume among the years from 2009 to 2020, reaching up to 45×10^6 km^3 and 22×10^6 km^3, respectively. It suggests that the anomalous ice PSC areal coverage observed during the same period (Fig. 6b in the revised manuscript) is beyond the interannual variation. Similarly, the year 2020 recorded the highest enhanced NAT PSC volume during 21st to 30th July 2020. For STS PSC, the second week of July and the last week of August 2020 recorded the highest PSC volume of 29.5×10^6

km^3 and 8.2×10^6 km^3, respectively. However, liquid-NAT mixture PSC volume did not exhibit significant anomalies relative to other PSC types.

We also analyzed the interannual variability of polar vortex dynamics, as suggested by Reviewer #4 as well, and found that the increase in PSC areal coverage and volume is not related to the vortex area and temperature. Figures 8 and 9 and the associated discussion in the revised manuscript (sub-section 4.4.1 on Page 17 L448 to L555) provide details on this aspect.

• Again Figure 7, the authors write: "It is evident from Figure 7 (b, h, and k) that the positive anomalies in areal coverage of PSCs such as ice, STS, and enhanced NAT exceed three standard deviations with respect to the background mean. The increase in HNO3-containing PSCs: STS and enhanced NAT can be attributed to the increased surface area provided by the wildfire aerosols and the production of HNO3 by the N2O5 hydrolysis process. In contrast, the areal coverage of another HNO3-containing PSC, i.e. liquid-NAT mixtures, showed strong negative anomalies (Fig. 7 (e and f))". A comparison with the literature states that liquid NAT mixtures in particular are formed by heterogeneous nucleation of NAT on foreign nuclei, and that enhanced NAT is a product of NAT nucleation on ice. This does not fit with this study. At least it needs to be discussed.

Thank you for the comment. In the present study, we show that NAT nucleating on ice also resulted in the liquid-NAT mixture (please see the case no. 2 and Fig. 11 in the revised manuscript). It is possible that enhanced NAT could be a result of NAT nucleation on mountain wave ice. However, since the ERA5 operational analysis product does not capture gravity wave, it is difficult to discuss the enhanced NAT anomaly in detail as we did for liquid-NAT mixture and ice PSC cases.

• The paper lacks a proper description of the tools and methods used. The CLaMS model was never introduced. What does the acronym CLaMS mean is the least question. The authors never introduce the box model at all. To run the box model, you have to define many more parameters than just H2O and HNO3 concentrations. MLS does not measure all the quantities you need to define before you can run the model. In addition, you need to predefine thresholds and pathways for PSC formation. For example, at what supersaturation does NAT/ice form in the model?

In Sect 3.4 in the revised manuscript (page no. 7 L238 to L260), we have added more details on the CLaMS model and the information about the input parameters, as shown below

"*During the formation of ice and liquid-NAT mixture PSCs, uptake of gas-phase HNO_3, and H_2O mixing ratio occurs. We used the Chemical Lagrangian Model of the Stratosphere (CLaMS) microphysical box model to validate the MLS observed uptake in these gases with CLaMS-modeled uptake during the formation of the PSCs. CLaMS is a chemical transport model developed by Research Centre Jülich, Germany (McKenna et al., 2002a, 2002b). It has several modules such as chemistry (McKenna et al., 2002b), mixing (McKenna et al., 2002a), trajectory (Konopka et al. 2004), sedimentation (Grooß et al. 2005; Grooß et al. 2014; Tritscher et al. 2019), etc. which collectively helps in simulating simulate stratospheric chemistry, PSC formations, and ozone depletion.*

In the present study, we utilized the trajectory module and microphysical box model from the chemistry module. First, a backward trajectory is estimated as discussed in Sect 3.3 (ii), and the intersection of the trajectory with the CALIPSO scan track is identified as discussed in Sect 3.3 (iii). Since the CALIPSO PSC product comes with MLS observations interpolated to the CALIPSO grids, the beginning and end of the trajectory have MLS HNO_3 and H_2O. Intersection of the backward trajectory with the CALIPSO scan track is considered as a beginning point and fed into the CLaMS microphysical box i.e., the trajectory starts at time, $t < 0$ hr and ends at time = 0 hr. The inputs for the box model run are trajectory coordinates (time, latitude, longitude, and potential temperature), temperature and pressure along the trajectories, and MLS observed HNO_3 and H_2O mixing ratio at the beginning *of the trajectory. The mixing ratios of the rest of the gases are set to '0', hence other chemical reactions such as chlorine activation, HCl null cycles, and N_2O_5 hydrolysis are not considered. In the box model run, T_{ice} is estimated following equations provided by Marti and Mauersberger (1993), and T_{NAT} is estimated following Hanson and Mauersberger (1998).*"

• Even though the reviewed version of the paper now includes backward trajectories, looking at the temperature history, I still miss a lot of details. How many trajectories did the authors calculate to reach their conclusion? Are 6 case studies enough? How does the entire PSC look like? Why do the authors not show plots of the selected CALIOP PSC orbit curtains?

We considered a total of 8,841 trajectories. Out of these, 3792 are backward trajectories of the liquid-NAT mixture and the rest 5049 belong to ice. For NAT formation pathways, 2996 trajectories correspond to the liquid-NAT mixture formed through the ice-assisted formation pathway and 796 trajectories to the ice-free formation pathway. For ice formation pathways, 4796 trajectories correspond to the ice formed through the NAT-assisted formation pathway and only 253 trajectories to the NAT-free formation pathway. The discussed five cases (case no. 2 is moved to a supplementary section in response to the reviewer's suggestion) are representative of 8841 trajectories.

In the revised manuscript, we added the above information on page no. 7 L228 to L229 , page no. 33 L826 to L829, and page no. 41 L978 to L980

Furthermore, for each case study, we have added the CALIPSO PSC orbit curtains plots in the supplementary section (Fig. S7, S9, S10, S14, and S15)

• I like the "match approach", but the time in between the two CALIOP observations has not been examined closely enough in my opinion. Especially when looking at ice formation, half a degree K can make a big difference. A resolution of 1 x 1 degree does not include small scale gravity waves. Looking at Figure 9: There is the Antarctic Peninsula between the two CALIOP observations. It could well be that ice PSCs formed in between, producing wave ice.

We agree with the reviewer's view that wave ice could have formed due to temperature fluctuations caused by gravity which acts as nuclei for NAT formation. It could also lead to uncertainty in the final estimated percentage contribution of formation pathways. In the revised manuscript, we clarify this in page no. 42 (L1006 to L1008).

"*The inability of the ERA5 operational analysis to capture the small-scale temperature fluctuation caused by the gravity waves and the possible bias in ERA5 temperature may lead to uncertainties in the percentage contributions of the formation pathways."*

However, please note that there are several cases where NAT was observed to form over regions less prone to gravity waves. Moreover, the corresponding temperature history revealed that the temperature of the air parcel never decreased below T_{ice}, suggesting no involvement of ice in NAT formation.

• "It could be due to a bias in the temperature from the ERA5 operational analysis used in this paper. The ERA5 operational analysis temperature is consistently 1 K lower than the radiosonde measured temperature (Engel et al., 2013)." This is incorrect. ERA5 did not exist at that time.

Thank you for pointing it out. In Engel et al., (2013), the author mentioned that the ECMWF operational analysis has cold bias of 1 K. But in the present study, we use ERA5 operational analysis. Hence, we have removed this sentence from the manuscript.

• As mentioned in my first review, the role of STS is still confusing. The NC -> PSC formation mechanism

makes no sense. STS droplets do not form suddenly, they gradually increase in size from the background stratospheric sulphuric acid aerosols by absorbing HNO3 and H2O, and at a certain size they can be detected by CALIOP. NAT and ice particles are always mixed with the background aerosols or, at a certain size, the so-called STS droplets.

The 'NC' flag in CALIPSO refers to the "No Cloud" conditions but may contain stratospheric aerosols such as sulphuric acid droplets and/or meteoritic dust particles or sub-visible PSCs (Lambert et al., 2012;2016). In 'NC -> PSC' formation, we also say that due to a decrease in temperature condensation of gases such as HNO_3, and H_2O occurs forming PSCs (see case no. 1 in page no. 22 L585 to L627).

• The authors only consider selected PSC classes. What about enhanced NAT and wave ice? They did not even talk about it.

The anomaly in the areal coverage and volume of enhanced NAT was already given in Sect 4.4. However, their formation pathways are not discussed as the ERA5 reanalysis data does not capture gravity wave which plays a crucial in forming enhanced NAT. For the same reason, the formation pathways of wave ice are not investigated. The contribution of the wave ice to the total PSC areal coverage is negligible (~0.1 % to the total PSC) (Pitts et al., 2018). Hence, wave ice is not even discussed in the present study.

• Bimodal distribution of the depolarization ratio: What does depolarization mean? Why do the authors look at depolarization? I miss such kind of explanations.

The reason for investigating the depolarization ratio was given in page no. 37 L844 to L851 in the second submission.

• I found some careless mistakes like "In the box model run, Tice is estimated following the equations provided by Hanson and Mauersberger (1988), and TNAT is estimated following Hanson and Mauersberger (1998)".

Thank you for pointing out. We have corrected it as:

"In the box model run, T_{ice} is estimated following equations provided by Marti and Mauersberger (1993), and T_{NAT} is estimated following Hanson and Mauersberger (1998)"

To summarize: The paper is not well structured and not easy to understand. The paper still lacks a proper literature review. The study of PSC formation requires much more in-depth analysis. Understanding ice formation requires much higher resolution of all parameters in space and time. There are unacceptable errors in the manuscript and the conclusions drawn by the authors are questionable in my opinion. I still cannot recommend that this manuscript be published in its current form.

Thank you for the comments. We sincerely believe that the revised version of the manuscript has improved in terms of structure and scientific arguments, as has been pointed out by the other reviewers. We once again express our heartfelt thanks to the reviewers for the constructive comments.