Author response to Reviewer #2 comments

We sincerely thank the reviewer for the comments. Based on the comments we have carefully revised 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

This paper describes the impact of Australian extreme bushfire event during 2019/2020 on Antarctic PSC occurrence and subsequent ozone hole in 2020. It is a new idea to investigate the influence of extreme bushfire on Antarctic PSC formation. However, since the author discuss only one winter (2020), the discussion and conclusions that the authors mentioned in the paper is not very convincing. At least, the authors should analyze another year when the Antarctic ozone depletion is about the same magnitude as 2020 (for example, 2021 or 2022), and compare the results with those in 2020.

We thank the reviewer for the valuable comments and appreciation as well as the suggestion to extend the work to another year. But unfortunately, the CALIPSO PSC V2 product, which is used in the present work is available only till March 2021, and not beyond. As such, we could not carry out this study for 2021/ 2022.

However, now we have strengthened the discussion and conclusion section by performing the Lagrangian backward trajectory analysis to study the PSC formation pathways and also simulated the PSC formation using CLaMS microphysical box model (complying with the comments of reviewer #1) to validate the Microwave Limb Sounder (MLS) observed uptakes of HNO_3, and H_2O against the modelled uptake during formation of ice and liquid-NAT mixture PSC and included in the revised manuscript (also please see the responses to reviewer #1 in this regard). The details are as below:

First, we calculated the 48 h backward trajectories of ice and liquid-NAT mixture PSCs using CLaMS trajectory module with hourly ERA5 operational analysis meteorological data. Using the trajectories, we identified which type of PSC preceded the initial PSC (ice/liquid-NAT mixture) for which backward trajectory is estimated. The temperature history, uptakes of MLS HNO_3, H_2O mixing ratio during the formation of the ice/liquid–NAT mixture PSCs are studied. 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.

< Specific Comments >

1) P.3, L.72: MLS is a 5-band microwave radiometer, not a Fourier-Transform Spectrometer (FTS).

Thank you for pointing out this. We have made the necessary correction in the revised manuscript.

2) P.3, L.83: The authors used the terminology "Liquid Nitric Acid Trihydrate (LNAT)" throughout the paper. However, Nitric Acid Trihydrate is a solid PSC, not liquid at all. I assume that the authors are referring "Mix 1" or "Mix 2" PSCs in Pitts et al. (2009), which are the mixture of STS and NAT PSCs. The authors should not use the term "LNAT", but better use the term "Mix PSCs".

Thanks for pointing this out. We are sorry at using a confusing terminology (as also has been pointed out by the other reviewers) and have corrected it in throughout the revised manuscript. Now, instead of 'LNAT, we used 'liquid-NAT mixtures throughout the manuscript

3) P.7, L.204, Figure 3 caption: What is the definition of "standardized anomaly (Z)"? Please define Z by a equation.

The standardized anomaly (Z) is estimated as ratio between the anomaly (2020 – Background mean) and standard deviation estimated using background period. In the manuscript, it is given in equation no. 2 at line number 181.

4) P.8, L.214: What is the meaning of "additional"? In addition to what? Please explain.

Antarctic ozone depletion occurs during spring of every year and is attributed to the active halogens in lower stratosphere. Usually, the magnitude of ozone depletion (i.e. ozone loss) for a given year is estimated by subtracting ozone amount of September/October (i.e. spring period) from June of the same year. In addition to this usual normal ozone loss, additionally more ozone is lost during the year 2020 (Ansmann et al., 2022) and hence it is called as 'Additional ozone loss.'

Ansmann, A., Ohneiser, K., Chudnovsky, A., Knopf, D. A., Eloranta, E. W., Villanueva, D., Seifert, P., Radenz, M., Barja, B., Zamorano, F., Jimenez, C., Engelmann, R., Baars, H., Griesche, H., Hofer, J., Althausen, D., & Wandinger, U. (2022). Ozone depletion in the Arctic and Antarctic stratosphere induced by wildfire smoke. *Atmospheric Chemistry and Physics*, *22*(17), 11701–11726. https://doi.org/10.5194/ACP-22-11701-2022

5) P.8, L.222: What is the meaning of "additional"? In addition to what? Please explain.

Explained. Please see the response to comment #4

6) P.8, L.222; Fig S1: The year 2019 was a exceptional year when the Antarctic ozone loss was minimum (like the year 2002), due to the dynamical effect (split of polar vortex in early spring). The authors should not compare the year 2020 with 2019, but had better compare with another normal year (like 2021 or 2022). One suggestion is to show time-series plots not for 2019-2020, but show like 2020-2021.

Thank you for the comment. In Fig. S1, we are showing the anomaly in Antarctic ozone from September 2019 to December 2020. This period is chosen to show the sudden increase in lower stratospheric aerosol and subsequent change in temperature followed by the black summer event which occurred at end of December 2019 through Fig. 1 and Fig. 2. Here, we are not comparing the year 2020 with 2019 but intended to show the sudden changes. To maintain consistency in chosen time period, we made the plot Fig S1 as well to start from September 2019 and end at December 2020.

7) P.11, L. 292: The authors claim that "increased H_2O is transport-related as the data are close to the linear fit". However, I felt that the 2020 H_2O data are also deviated upward from the linear fit.

Thank you for the comment. In case of HNO_3, the 2020 HNO_3 are extremely deviated from linear fit i.e., if we make another linear fit for 2020, it would be at least 40° with the linear fit line corresponding to background data. However, in case of H_2O, if we make another linear for 2020, this linear fit line would be less than 10° or less with the linear fit line corresponding to background data i.e., the deviation is not much stronger to conclude that H2O is chemically produced during 2020.

8) P.13, L. 331: The authors claim that "Followed by ice, the Supercooled Ternary Solution (STS) exhibited a high positive anomaly". However the presence altitudes of ice and STS are different (ice: 15-20 km, STS: below 15 km). STS are not "followed" by ice.

Thank you for the comment. By "Followed by ice", we tried to convey that ice PSC areal coverage increased to significantly with peak positive anomaly of 4×10^6 km^2. The magnitude of positive anomaly is higher than anomaly by other PSC. After that, STS PSC high positive anomaly of 3.5×10^6 km^2 as discussed in manuscript, L486 to L488, page no. 18.

9) P.13, L.334: The authors claim that "the STS areal coverage can … lead to additional ozone loss." However, since the appearance altitude of STS are mostly below 15 km, the additional ozone loss cannot be expected in such low altitudes.

It is true that most of the STS forms below 15 km altitude. But the STS PSC is also present at altitude of 16 to 25 km during early June 2020 (Fig. 7 (j–l)) which could provide surface for chlorine activation reaction leading to additional ozone loss. This is supported by results from Ansmann et al., (2022) who shown that around PSC height range of 14–24 km, additional ozone loss is observed in September–October 2020.

Ansmann, A., Ohneiser, K., Chudnovsky, A., Knopf, D. A., Eloranta, E. W., Villanueva, D., Seifert, P., Radenz, M., Barja, B., Zamorano, F., Jimenez, C., Engelmann, R., Baars, H., Griesche, H., Hofer, J., Althausen, D., & Wandinger, U. (2022). Ozone depletion in the Arctic and Antarctic stratosphere induced by wildfire smoke. *Atmospheric Chemistry and Physics*, *22*(17), 11701–11726. https://doi.org/10.5194/ACP-22-11701-2022

10) P.14, Figure 9: I am curious if the authors also show the similar plot to indicate the formation pathways of STS from other types of PSCs.

We appreciate the comment. In the lower stratosphere, it is the STS PSC which always forms first i.e., well before formation of ice/liquid-NAT mixture. Hence, in terms of STS formation pathways, it essentially does not require NAT/ice to act as nuclei for formation. But, it can form either homogenously or heterogeneously. For this reason, the formation pathways of STS are not discussed in the present study.

11) P.17, L.422: In the course of the formation of ice PSC, why uptake of HNO_3 occurs? Where the decreased HNO_3 goes to? Please explain.

It is explained as follows. Most of the ice PSCs form by nucleating on NAT particles as our study reveals. When temperature of the air parcel decreases, HNO_3 condenses forming STS first, followed by liquid-NAT mixture. This process leads to depletion of gas-phase HNO_3. Following the formation of the liquid-NAT mixture, if the temperature remains below T_{NAT} (the NAT existence temperature) or decreases further, the NAT particles continue to grow, leading to further depletion of gas-phase HNO_3. As temperatures decrease even further reaching below T_{ice}, these NAT particles act as nuclei for ice formation. Hence, even in the course of ice formation, depletion in HNO_3 is observed.

We discussed this in detail through a case study (Case no. 4; Page no. 26; Fig. 11). In this case, we show that, initially temperature of the air parcel is above T_{NAT} and no PSC is formed by this time as confirmed by CALIPSO observation. When temperature decreased well below T_{NAT}, NAT particles appeared first as revealed by CLaMS box model run (Fig. 11 (e)). As the temperature decreased below T_{ice}, ice PSC formed (Fig. 11 (f)). During this ice formation process, ~5 ppb of HNO_3 is depleted.

12) P.18, L.457: The authors claim that "explain the high anomalous ice areal coverage." However, not reference nor supporting material/figure are shown to support the "high anomalous ice areal coverage" in 2020. Please show anything to explain that ice areal coverage in 2020 was anomalously high compare with other years.

Thank you for the comment. In the manuscript, at page no. 18 Fig. 7 shows the CALIPSO observed PSC areal coverage (first column), corresponding anomaly with respect to background period (second column), and standardized anomaly (third column). In this figure, ice PSC areal coverage (Panel a), anomaly (Panel b), and standardized anomaly (Panel c) are included to show the high positive anomaly of ice PSC.

13) P.19, L. 468: The authors claim that "anomalously high PSC areal coverage was observed." However, not reference nor supporting material/figure are shown to support the "anomalously high PSC areal coverage" in 2020. Please show anything to explain that PSC areal coverage in 2020 was anomalously high compare with other years.

Replied under comment #12

< Grammar/Typos >

14) P.2, L.36: Last ";" after Selitto et al. is not needed.

Complied with.

15) P.4, L.120: orbit the Earth ~15 times ---> orbit the Earth ~14 times

The section 2.5 which contains this line is completed removed, as we changed the methodology to retrieve the formation pathways using by studying the temperature history, MLS HNO_3, H_2O along with CLaMS box model simulation of PSC formation.

16) P.6, L.185: $\Delta T = T_n - T_{n+2}$ ---> $\Delta T = T_{n+2} - T_n$

Thank you for pointing out this. The section 2.4 containing this equation no. 3 is completely removed. As we are studying the complete temperature history of the air parcel containing ice/liquid-NAT mixture now (a more vital information than ΔT), we have not considered ΔT in this revised manuscript.

17) P.6, L.186: $\Delta HNO_3 = HNO_{3n} - HNO_{3n+2}$ ---> $\Delta HNO_3 = HNO_{3n+2} - HNO_{3n}$

The section 2.4 containing this equation no. 4 is completely removed. However, we considered the uptake of MLS HNO_3 during PSC formation and compared with CLaMS box model result. This scatter plot of observed MLS HNO3 uptake and CLaMS modelled uptake is shown in Fig. 12 and Fig. 18.

18) P.6, L.188: obtained from MLS and MERRA-2 ---> obtained from MERRA-2 and MLS

Thank you for pointing out. We completely removed section 2.4. This time, we considered temperature from ERA5 operational analysis instead of MERRA-2 as ERA5 is more reliable than MERRA-2.

19) P.9, L.240: in k_{ext} ---> in Δk_{ext}

Complied with.

20) P.9, L.241: the result of mesospheric air ---> the result of the descent of the mesospheric air

Complied with.

21) P.10, L.276: (such as convection or advection. ---> (such as convection or advection).

Complied with.

22) P.10, L.281: chemical production of ---> chemical production or destruction of

Complied with.

23) P.11, L. 298: the same period (Fig. 7a) ---> the same period (Fig. 7c)

Complied with. As the Fig. 1 and Fig. 2 which was demonstrating the methodology which employed to retrieve PSC formation pathways are changed, Fig. 7 became Fig. 5 now. Hence, changed Fig. 7a to Fig. 5c.

24) P.13, L.328: mid-April itself (Fig. 7c) ---> mid-April itself (Fig. 7b)

Complied with.

25) P.15, L.375: ΔHNO_3 = -0.8 ppbv ---> ΔHNO_3 = -1.0 ppbv

The section 3.5.1, which contains this line is removed. However, we presented the change in HNO3 (ΔHNO3) in the form of case studies in revised manuscript. Furthermore, the MLS observed ΔHNO3 is validated against CLaMS modelled ΔHNO3 and shown in Fig. 12 (for liquid-NAT mixture) and Fig. 18 (for ice).

26) P.16, L.399: ΔT = 0.8 K ---> ΔT = 1.0 K

The section 3.5.1 which contains this line is removed.

27) P.18, L.439: ΔT and ΔHNO_3 of -0.7 K, and -0.9 ppbv ---> ΔT and ΔHNO_3 of -1.1 K and -1.1 ppbv

The section 3.5.1 which contains this line is removed.