We thank the reviewers and Dr. Šácha for their thorough and detailed reviews, which were very helpful in encouraging us to reorganize, clarify, and revise our manuscript. Please find the reviewers' comments below in plain text and our responses in bold font.

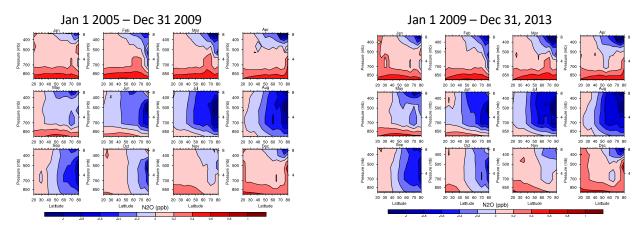
RNDr. Petr Šácha, Ph.D. Editor

Reviewer 1 - Dr. Farahnaz Khosrawi, after commenting on the significant improvements in the revised version, goes on to recommend acceptance after minor revisions. Still, Dr. Khosrawi highlights few outstanding minor issues, which are partly editorial in nature, but also lists some scientific critiques under the Specific comments. I urge you to take advantage of the editorial suggestions and to carefully reply to the scientific comments. Based on my own reading, I would like to add to the specific comment P19 Fig. 6 by Dr. Khosrawi on statistical significance a point, that the reader needs to see additional statistical information also around Figs. 1, 2, 3, 4 and 9a. Please add information here on the spread behind the mean seasonal cycles and on the representativeness of the mean monthly anomalies, considering also the uncertainties of the measurements where applicable.

We have added uncertainty on the mean seasonal cycles of GEOSCCM and NOAA data shown in Figure 1 and 9a. The method is explained in Section 2.3. "Mean seasonal cycles for NOAA surface  $N_2O$  observations and GEOSCCM  $N_2O$  tracers were estimated using a bootstrapping method in which 20% of the timeseries was randomly removed and the remaining 80% was fit to a 3<sup>rd</sup> order polynomial plus first 4 harmonics. These steps were repeated over 500 iterations to estimate the range of uncertainty in the harmonic components of the fit." This a a common approach for estimating uncertainty in NOAA data. The std deviation of multiple flask measurements for each NOAA surface N2O data point is reported but is generally similar throughout the time series for a given site. We therefore did not apply weights (e.g., the inverse square of the stddev) to each data point when computing the harmonic fits.

For the GEOSCCM contour plots in Figures 2-3 we apologize that we do not have an obvious way to estimate the uncertainty, nor was this done in the related publication Liang et al. 2022, which focused wholly on GEOSCCM and presented similar contour plots. However, we have emphasized that we are showing these plots to qualitatively illustrate the influence of the stratosphere, but it is beyond the scope of this paper to quantitatively calculate that influence. Similarly, we have used the NOAA empirical background qualitatively to illustrate the stratospheric influence on the troposphere. In addition, in Figure 4 we have expanded the number of years used to compute the climatology to 2005-2013 (previously we used 2009-2013). We also show for the editor's benefit below that the panels show a similar pattern regardless of which 5 year segment is used.

#### NOAA Empirical Background anomalies of N2O mixing ratio



Finally, Dr. Farahnaz Khosrawi has graciously offered a list of technical corrections which are necessary to improve the understanding, and therefore the impact, of what you are trying to say. We should both thank the reviewer for this as it improves the final product.

# We have added to the Acknowledgements, "The authors thank Dr. Farahnaz Khosrawi and 2 other anonymous reviewers whose detailed and helpful comments much improved the manuscript."

Recommendation for minor revisions is echoed also by Reviewer 2, who appears to remain less convinced about the quality and significance of your manuscript. In spite of this the reviewer first makes it clear that the manuscript has been improved. Importantly, the reviewer then brings to our attention that some of their original remarks have not been addressed. This includes possible important points as the solar cycle effect and relation to stratospheric N2O studies. I second the reviewer's request that all of their comments from the previous round need to be properly addressed. The reviewer also graciously offers a long list of minor and technical comments and suggestions, which we should again thank the reviewer very much for, because they are driven with the intention to improve the manuscript on all fronts. We appreciate the time that Dr. Khosrawi and Reviewer 2 have taken to provide very detailed and helpful comments, both here and in their previous reviews, and have done our best to respond to them.

Finally, I would like to add one additional comment based on my own reading and shear interest in your study. It seems to me that the positive anomalies shown across the figures are never discussed in the manuscript (except in the Supplement).

Do you assume the positive anomalies to be a poor consequence of your methodology and the existence of negative anomalies?

We have focused on the negative anomalies because they are more likely to come from the stratosphere, which is the main interest of this paper. However, we have added a paragraph in the Results discussing Figure 3. "The positive anomalies in Fig. 3 also differ between model and observations, with the summer-dominant soil source assumed in GEOSCCM appearing as surface contours at 40° and 50°N, while the NOAA empirical background shows positive anomalies at the surface in late winter and spring, likely reflecting North American agricultural sources (*Nevison et al.*, 2018). At 60° and 70°N, the stronger contrast between positive and negative anomalies in GEOSCCM compared to NOAA throughout the atmospheric column reflects the model's larger seasonal cycle, as seen also in Fig. 1."

Especially based on Figs. 5 and 10 I would argue that the positive anomalies can stand for some physical process omitted in the discussion. For example, in Fig. 10 for 2017 (ATom2) except the existence of the positive anomaly centered around 55°S seemingly stemming from the tropics, the negative anomalies can be locally seen stronger than in 2016, which contradicts your discussion around this figure and the link with BDC you are trying to draw. We acknowledge in the presentation of Fig 10 in the Results that, "However, the positive anomaly in the mid-troposphere observed at 40-60°S during ATom-2, which may be a source plume from the Southern Ocean, tends to contradict the hypothesis that SH tropospheric N<sub>2</sub>O was lower overall in 2017 than in 2016."

We also discuss the limits of the QCLS data in the 3<sup>rd</sup> paragraph of the Discussion, including that, "QCLS data are measured across a narrow longitude band of the flight track for any given latitude on a limited number of days" and that "QCLS data are more likely to display synoptic-scale variability, such as the apparent surface source plume over the Southern Ocean seen in Fig. 10.."

Observational and model evidence for a prominent stratospheric influence on variability in tropospheric nitrous oxide by Nevison et al.

The manuscript has significantly improved and I appreciate the effort the authors have put in shortening and better structuring their paper. I have only some minor comments that are left and should be considered before publication.

Abstract: The abstract is much better now, but I still have some issues with it. For example the introductory two sentence you provide are somewhat independent from what you write in the next few sentences. To understand this connection the reader needs to be either an expert on the topic or have to read the paper first.

Some thoughts on the abstract:

- I would suggest to make the transition at line 25 a bit smoother. I think my major problem here is that you as motivation mention ENSO, but then discuss the results you get from the model without mentioning ENSO again until you come to the results concerning the correlations.
- Isn't the point here that you investigate the stratospheric influence on the seasonal cycle? Or are you investigating all processes that influence N2O cycle? This did not come really across in the abstract.
- ENSO is a tropical circulation, but then you discuss the influence of BDC and polar descent on the seasonal cycle how do then these processes fit together?
- My suggestion for the abstract would be: 1. Introductory sentence, 2. Data/model that are used, 3. There are hemispheric differences and then provide at the end your results and then the closing sentences.

Abstract rewritten to accommodate the suggestions above, "The literature presents different views on how the stratosphere influences variability in surface nitrous oxide (N<sub>2</sub>O) and on whether that influence is outweighed by surface emission changes driven by the El Niño Southern Oscillation (ENSO). These questions are investigated using a chemistryclimate model with a stratospheric N<sub>2</sub>O tracer, surface and aircraft-based N<sub>2</sub>O measurements, and indices for ENSO, polar lower stratospheric temperature (PLST), and the stratospheric quasi-biennial oscillation (QBO). The model simulates well-defined seasonal cycles in tropospheric N<sub>2</sub>O that are caused mainly by the seasonal descent of N<sub>2</sub>Opoor stratospheric air in polar regions with subsequent cross-tropopause transport and mixing. Similar seasonal cycles are identified in recently available N<sub>2</sub>O data from aircraft. A correlation analysis between the N<sub>2</sub>O atmospheric growth rate (AGR) anomaly in longterm surface monitoring data and the ENSO, PLST, and QBO indices reveals hemispheric differences. In the northern hemisphere, the surface N<sub>2</sub>O AGR is negatively correlated with winter (January-March) PLST. This correlation is consistent with an influence from the Brewer Dobson Circulation, which brings N<sub>2</sub>O-poor air from the middle and upper stratosphere into the lower stratosphere, with associated warming due to diabatic descent. In the southern hemisphere, the N<sub>2</sub>O AGR is better correlated to ENSO and QBO indices. These different hemispheric influences on the N<sub>2</sub>O AGR are consistent with known atmospheric dynamics and the complex interaction of the QBO with the Brewer Dobson Circulation. More airborne surveys extending to the tropopause, would help elucidate the stratospheric influence on tropospheric N<sub>2</sub>O, allowing for better understanding of surface sources."

#### **Specific comments:**

P1, L22: What forcing? This term is no longer used in the rewritten abstract -see above. P1, L23: Which issues? We now use the word "questions" which refer directly to the previous sentence, i.e., the "how" and "whether" clauses. P1, L28: What is meant with similar cycles? Have specified "similar *seasonal* cycles."

P2, L41: delete "ozone-depleting substance" since to my knowledge O3 is not directly reacting with N2O, but due to the conversion of N2O to the photolysis, ozone is destroyed by the resulting products and thus it is indirectly depleting ozone. If you would like to keep the sentence as is I would suggest to add the rereference of the paper by Ravinshankara et al. in Science (https://www.science.org/doi/10.1126/science.1176985). We have removed the ODS reference from the opening sentence, since it is discussed in a later sentence with mention of NOx as the actual catalyst of O3 destruction. We have also added the Ravishankara et al. 2009 reference to that latter sentence.

P4, L90: Here it is not clear from your sentence if Ruiz et al. (2021) did not find such an influence in the SH or if they did not investigate the SH. Thus, I would suggest to rephrase the sentence so that this becomes more clear. Reworded as, "Ruiz et al. (2021) found a direct correlation between the QBO and N<sub>2</sub>O photochemical loss rates in the tropical middle stratosphere but concluded that interannual variability in surface  $N_2O$  globally was governed more indirectly by QBO-related changes in the dynamical processes of the lowermost stratosphere. They showed evidence for a coherent influence of cross-tropopause transport on the surface N<sub>2</sub>O seasonal cycle in the NH but not the SH." Note: Ruiz et al. upon rereading are a bit ambiguous but seem to be referring to global patterns (see their supplementary Fig. S3). However, their methodology is complex, and involves creating a 28 month "composite QBO signal "in chemistry transport models and comparing to surface N2O from NOAA. They found that QBO-related loss rates of F11 and N2O are only weakly correlated in the tropical stratosphere but that the QBO composite surface signal of F11 and N2O are nearly 100% correlated, hence their conclusion. Their finding of a strong influence due to cross-tropopause transport in the NH but not the SH refers to the mean seasonal cycle rather than IAV.

P5, L118: add "can be" so that it reads "can be distinguished from tropospheric tracers......" **Done** 

P5, L122: Is the aircraft data only used/available for the NH? Thus, SH solely based on model data? Please state more clearly what data has been used for which hemisphere. We have rewritten the sentence as, "The study also examines atmospheric N<sub>2</sub>O data collected in the NH by the National Oceanic Atmospheric Administration (NOAA) during routine aircraft monitoring, as well as N<sub>2</sub>O data measured by recent global airborne surveys spanning both hemispheres." This reordering clarifies that the NOAA data are in the NH only but that the airborne surveys (referring to HIPPO, ORCAS, ATom) span both hemispheres. (It is a somewhat lengthy process to spell out all the acronyms and describe the QCLS instrument, so that text is deferred to Section 2.2.3 in the methods.)

P5, L124: reflects? I would rather write "is used as tracer for". Substituted, "is used as a tracer for"

P6, L160: Again I have to ask why Mauna Loa is used. You provide an answer in your reply to my comments, but you have not added a reasoning here. I would suggest to add here a short explanation why you picked Mauna Loa and not one of the other stations. Added, "The N<sub>2</sub>O time series at MLO is a good proxy for the global N<sub>2</sub>O trend and thus its subtraction provides a convenient, single-station approach for calculating anomalies of the N<sub>2</sub>O mixing ratio for contour plots." (Note: by "convenient, single-station" we mean that the reason to use one station and not a combination of many is that it's a lot easier to extract matching values and make the calculations for model output as well as observations.)

P7, L178: you mean "approach the end of their lifetime" or are these instruments just getting old? In the latter case I would rather write "as the instruments are aging" than "as the instruments approach their lifetime". **Replaced with "as the instruments age."** 

P7, L187: Listed above? Which ones? Are these the same ones as for NOAA? Please rephrase the sentence to be more clear with which stations you mean. The 5 HATS sites are named in the preceding paragraph. We modified the next paragraph to read, "This study used the NOAA combined HATS/CCGG  $N_2O$  product from 1998-2021, which is based on monthly medians from the CATS *in situ* program (at the 5 HATS baseline sites) and monthly means from the CCGG flask program at a selected subset of 12 of the ~55 total sites (https://doi.org/10.15138/GMZ7-2Q16; Hall et al., 2007). All of the NOAA sites considered in this study are long-standing remote sites situated away from strong local anthropogenic sources. They include Alert, Canada; Summit, Greenland; Mace Head, Ireland; Trinidad Head, California, Cape Kumakahi, Hawaii, Cape Matatula, Samoa; Palmer Station, Antarctica, and the 5 HATS baseline sites (at which CCGG also makes overlapping flask measurements). In addition to these 12 individual sites, global, NH and SH means are estimated from the latitude-binned and mass-weighted means of the combined monthly means for the 12 sites (*Hall et al.*, 2011). The combined monthly data are first aggregated at the measurement program level for each sampling location. At sites where both HATS and CCGG measure, a weighted mean is calculated based on the programs' monthly uncertainties."

P7, L208: See my comment a tP6, L160. Again here you mention that Mauna Loa has been used for detrending the data, but without given a reason. You should at some point in the manuscript provide one sentence why Mauna Loa is used and not another station. **See response above re: MLO.** 

P14, L336: What one can clearly see from the cross sections is the downward transport of the air. But I have difficulties to see horizontal transport and mixing. Since this are known transport processes I would suggest to add here some adequate references. We have added the references latitudes (*Liang et al.*, 2009; 2022), which describe horizontal transport and mixing for GEOSCCM.

P17, Figure 5: Why do you use for the aircraft data a different color scheme than for the other datasets? The aircraft panel figures were created by Dr. Stephens while the other panels were created by Dr. Nevison using different software.

P19, Figure 6: Not for all panels the p value has been added.

We have added p values for all the AGR plots which involved correlating monthly N2O AGR to monthly QBO and ENSO indices. These correlations required special treatment because they have a variable N due to the autocorrelation that is introduced in part by the 12-month running mean used to deseasonalize N<sub>2</sub>O to compute the AGR. To account for autocorrelation in each time series, we used an effective N (N<sub>eff</sub> = N/), which is described in the new section S2 in the Supplementary Materials.

P29, 618ff: You are not really providing here a summary at least not in the sense what was the aim of the study and what has been done. Further, the order as you discuss the results in your summary and conclusion is somewhat weird. Why do you start with the aircraft data instead with the model and NOAA data which are the main data sets of your study? Further, the discussed results cannot only be derived with aircraft data. This can be achieved with other measurement data sets as well.

We have reversed the order of the first 2 sentence, presenting the GEOSCCM results first, then the aircraft results. In the penultimate sentence, we have changed "To further refine" to "To help refine" which doesn't exclude other measurement data sets (e.g., from satellites) but focuses on summarizing the data sets presented in the current study and how our analysis can be improved going forward.

Additional comment: Concerning my comment on P21, L473 of the previous version of your manuscript concerning if the reference of the text book by Holton (1995) is still valid for the hemispheric differences

in the BDC. First of all, here you should rather cite the 2<sup>nd</sup> edition of the text book published in 2006 or check the following papers by Garny et al. (2013), Butchart et al. (2014) or Fu et al. (2019).

For the discussion of the NH vs. SH difference in Section 4.2 we believe that Holton 1995 (which is a paper in Reviews of Geophysics), while old, is still a good reference for the assertion that the BDC is stronger in the NH than the SH (see, e.g., their Table 1). We have cited it along with Scaife and James, 2000 and Kidston et al., 2015 and Butchart 2014. We have additionally cited the Holton et al. 2004 textbook earlier in Section 2.4.1 as a one of the references describing why we use PLST as a proxy for the strength of the BDC. We appreciate the Garny, and Fu references but our reading suggests that those papers are focused mainly on recent changes in the strength of the BDC rather than the fundamental NH vs. SH difference. However, Butchart 2014 mentions the stronger BDC in the NH so we have cited it.

**Technical corrections:** 

P7, L191: Empirical background -> empirical background Changed to lower case

P8, L211: Abbreviation HIPPO not introduced. It's done on L219, but this should appear at the first instance where the abbreviation is used. **Changed HIPPO to "first of the airborne surveys described below" and spelled out High-performance Instrumented Airborne Platform for Environmental Research (HIAPER)** Pole to Pole Observations (HIPPO) in the preceding section 2.2.3 describing the airborne surveys.

P8, L223: Abbreviation ORCAS has not been introduced. O<sub>2</sub>/N<sub>2</sub> Ratio and CO<sub>2</sub> Airborne Southern
Ocean (ORCAS)
P17, L390: "an annual sequence" appears twice. One is thus obsolete. Deleted extra "annual sequence"

P17, L395: Add here the section number. Added Methods (Section 2).

P19, L425: Since all the correlations you consider are rather week I would suggest to omit the term "strongest". I would rather use the term "highest". Further, when the correlation is negative you should either clearly state that this correlation is negative or call it an anticorrelation. Rewritten, "In contrast to the SH, the NOAA surface N<sub>2</sub>O AGR in the NH is anticorrelated significantly to winter PLST (R = -0.67), with an optimal correlation for the 12-month period from July-June encompassing the January-March PLST average (Fig. 7b). A similar anticorrelation is found between the GEOSCCM PLST and NH N<sub>2</sub>O AGR (Fig. 7d). Also in contrast to the SH, the NOAA NH N<sub>2</sub>O AGR is correlated only weakly to the QBO index at all altitudes, with a negative sign. The highest correlation in the NH occurs for 50 hPa QBO (R= -0.23, p>0.1) with a 10-14 months lag (Fig. 7a). GEOSCCM also predicts an anticorrelation (R = -0.47, p < 0.05) between the GEOSCCM QBO and the NH N<sub>2</sub>O AGR, which also is optimal around 50 hPa with 10-14 month QBO lag (Fig. 7c)."

P20, Figure 7: Also here in some of the panels the p-value is not given. We have added p values to Fig. 7 – see response above for Fig.6.

P21, Figure 8: The grey lines are hardly visible on a printout version of your manuscript. Please use a somewhat darker grey for these lines. We have used a darker grey for the ENSO lines.

P25, L515: The reference Khosrawi et al. (2009) is missing in the reference list. Instead you still have there the Khosrawi et al, (2013) reference which is actually not cited. We have removed Khosrawi et al. 2013 and added Khosrawi et al. 2009 to the References.

P26, L541: Didn't you state before that the strongest correlation for the QBO is found at 50 hPa? Please check the numbers and levels if everything is correct and consistent discussed. We have clarified by adding "... consistent with Ray et al. (2020) (who only presented results for QBO = 50 hPa)." In other words, we are pointing out that our lag time is consistent with Ray et al. 2020 at 50 hPa, which was the only pressure at which they considered the QBO. (We considered the QBO index at a range of pressures from 100 hPa to 10 hPa.)

P29, L617: "Summary and" should also be in bold face. Done

P31, L665: Check the formatting of the references. Indents for the consecutive lines of each reference are missing and different style for the references is used. This should be done in a uniform style and according to the ACP guidelines. We have indented the references and put them in ACP format.

P33, L754: Reference Khosrawi et al. (2013) appears twice, but has not been cited in the manuscript. Further, Khosrawi et al. (2009) which has been cited in the manuscript is not listed in the reference list. We have removed Khosrawi et al. 2013 and added Khosrawi et al. 2009 to the References.

Note: Figure should appear as Fig. in the text, except at the begin of a sentence (see ACP manuscript preparation guidelines). Changed Figure to Fig. throughout the text.

Supplement, 2<sup>nd</sup> page, 2<sup>nd</sup> paragraph: What do you mean with "a year prior"? Do you mean "a prior year"? **Changed to "one year earlier."** 

Supplement: Figure captions -> remove Supplement before S1, S2, and S3. Done

#### **References:**

Butchart N., The Brewer-Dobson circulation (2014), Reviews of Geophysics, 52 (2), pp. 157 - 184, DOI: 10.1002/2013RG000448.

Fu Q., Solomon S., Pahlavan H.A., Lin P.: Observed changes in Brewer-Dobson circulation for 1980-2018, Environmental Research Letters, 14 (11), 114026, DOI: 10.1088/1748-9326/ab4de7, 2019.

Garny H., Bodeker G.E., Smale D., Dameris M., Grewe V.: Drivers of hemispheric differences in return dates of mid-latitude stratospheric ozone to historical levels, Atmospheric Chemistry and Physics, 13 (15),7279 - 7300, DOI: 10.5194/acp-13-7279-2013, 2013.

### **Reviewer 2**

I appreciate the thorough work of the authors: the quality of the manuscript is highly improved, and the results are clearer and the reading smoother. I have the feeling now that my problem with the ENSO discussion was the lack of a strong introduction and motivation.

However, a few of my comments were only partially answered or not discussed at all, and the revised version of the manuscript brought to the surface an issue concerning the discussion about the impact of the BDC. In addition, I have some minor/technical corrections that should be addressed.

I recommend publications after the points below are addressed.

### Partially answered/unanswered comments.

In the following, I copy my comments of the previous review in italic.

• Conclusions. The authors highlight the relevance of airborne measurements for the current study in comparison with satellite data and that is a perfectly fair point. However, the authors do not provide any comments regarding the following possible points of discussion that I raised in the first review (pasted below). If the authors decide to disregard these suggestions, I would be interested to know why.

o TheauthorsfindthatthesurfaceN2Ogrowthratepresentshemisphericaldifferences in the response to the impact from the QBO (strongest in the SH) and the BDC (strongest in the NH). Minganti et al., (2022) found hemispherical differences in the N2O trends in the stratosphere (positive in the SH and negative in the NH) in satellite observations and reanalyses. I wonder if these hemispherical differences in the stratospheric trends can be related to the differences in the surface N2O growth rate (or just mentioned).

We have changed the Summary and Conclusions to Summary and Outlook. We include these sentences in the Outlook portion, "Another important issue for future research is the impact on N<sub>2</sub>O of climate change driven increases in the strength of the BDC (*Garny et al.*, 2013; *Butchart et al.*, 2014; *Fu et al.*, 2019). Of particular relevance to the results presented here are studies based on

# ground or satellite-based N<sub>2</sub>O observations that find a decrease in the N<sub>2</sub>O lifetime (*Prather et al.*, 2023) and interhemispheric differences in stratospheric N<sub>2</sub>O trends (*Minganti et al.*, 2022)."

o Itwouldbeinterestingtoaddone/twosentencesonthepossibleimpactofthesolar activity on the N2O growth rate. The major chemical destruction of N2O occurs in the tropical upper stratosphere, so I do not expect large impact on the surface growth rate. However, some signal could still reach the troposphere and certainly an additional proxy for solar activity would help to better understand the N2O changes in the stratosphere.

We have also added to the Summary and Outlook, "The solar cycle is an additional influence on variability in N<sub>2</sub>O that may be worth investigating in future studies. While previous studies have estimated a relatively small effect over the 2000s and 2010s due to low solar activity (Ruiz et al., 2021; Prather et al., 2023), solar cycle-driven changes in the UV flux affect N2O photolysis both directly and indirectly through the impact stratospheric O3."

o TheauthorscouldmentionthepossibilitytoperformsensitivitytestswithGEOSCCM. For example, an experiment with the QBO switched off (if possible) would isolate the patterns due only to the BDC.

We appreciate the suggestion but feel that it is beyond the scope of our study. While it is possible to switch off QBO in the GEOSCCM model simulation as a sensitivity study, it is computationally expensive to do so. And since GEOS is free running GCM, switching off QBO will likely lead to other changes that complicate the interpretation.

- Results. I appreciate the compromise of the authors, but Figures 7 and 8 do not seem to meet this compromise (respectively, P19L421 and P20L439). We have rewritten these paragraphs to introduce Fig. 7 and 8 by first describing what they show and then presenting the salient results. (We also did this for Fig. 6.)
- In Figure 8, the authors compare different observational datasets (Atom and ORCAS) for different periods (2016 and 2017). In my opinion, this makes the discussion difficult to follow. I suggest using only the ORCAS dataset for Figure 8. This would allow more room for discussion about the ORCAS dataset (maybe separating January and February?) and remove the asymmetry in Figure 8.

Can the authors clarify on why this comment was not answered?

We apologize for overlooking this comment. We did however respond to a related comment in the first review that requested that the ORCAS and ATom panels in Figure 8 be plotted in the same manner. "Thus, the left panel should also be plotted from -70 S to 80 N, but masking the parts of the data that are not considered as white area."

Our response then and now is that this would leave a lot of blank space on the ORCAs panel. We've added a sentence to remind readers that ORCAS focused on the Southern Ocean and was restricted to the extratropical SH, which is why there is an asymmetry in the two panels. The purpose of Fig. 8 is to provide a comparison of aircraft data in the extratropical SH in two successive years with opposite extremes in PLST and notable differences in their Feb. seasonal anomalies (as shown in Fig 9). While the comparison is qualitative and is complicated/undermined by synoptic scale features in the ATom panel, which we now acknowledge, we believe that this plot is important and relevant to our paper. P1L32. "warm". I think the "warm" here (and throughout the manuscript) arises from some confusion. The N2O-poor stratospheric air that is transported by the BDC over that region is not necessarily warm. The downwelling (i.e., the downward transport) due to the BDC at high latitudes during winter warms up the lower stratosphere because it's a diabatic process. Because of that, the PLST is used as a proxy of the strength of the BDC, i.e., warmer PLST indicate stronger downwelling due to stronger BDC.

In a nutshell, the BDC does not bring "warm air" to the lower stratospheric high latitudes, the air over that region is warm because of the BDC (e.g., Holton et al., 1995).

Thank you for this correction. We rewrote as, "The mean PLST in each hemisphere was treated as a proxy for the integrated strength of the BDC, which brings  $N_2O$ -poor air from the middle to upper tropical stratosphere into the polar winter lower stratosphere through diabatic descent. This warms up the lower stratosphere, with warmer PLST corresponding to stronger descent."

This comment does not change the conclusions of the authors: warmer PLST indeed indicate stronger BDC, but I suggest removing the "warm" before "N2O-poor air" throughout the manuscript as it might generate confusion.

#### Minor/technical comments.

- P1L15. Izana -> Izaña. Done
- P1L24. I suggest removing "atmospheric". **Done**
- P1L32. circulation -> Circulation (throughout the manuscript). **Done**
- P2L42. I suggest removing "(GWP)" as the abbreviation is not used further in the manuscript. **Done**
- P2L50. I suggest changing "ppb" with "ppbv" throughout the manuscript.

Our NOAA and QCLS co-authors Dr. Lan and Dr. Kort encounter this issue frequently but are firm that ppbv should NOT be used in place of ppb. Briefly, NOAA and QCLS are not measuring or reporting the N2O mole fraction in ppbv. The mole fraction they report is defined as the number of molecules of N2O in any given air parcel divided by the total number of all molecules (except water) in that parcel. For N2O it is usually expressed as parts per billion, abbreviated as ppb. To make values in mole fraction (in ppb) the same as those in ppbv, one would have to assume ideal gas law works, which is not the case in reality, and Dr. Lan and Dr. Kort don't make such approximations during their measurements or reporting. For more details please see section 7 of

https://gml.noaa.gov/aftp/data/trace\_gases/n2o/flask/surface/README\_n2o\_surface-flask\_ccgg.html

And also the WMO guidelines <u>https://library.wmo.int/records/item/57135-20th-wmo-iaea-meeting-on-carbon-dioxide-other-greenhouse-gases-and-related-measurement-techniques-ggmt-2019</u>

• P2L52. N is not defined her, but it is defined at the line below (L53). I suggest moving

"nitrogen (N)" to L52. Done

• P3L61-65. "While larger ... (Nevison et al., 2018)." I suggest re-phrasing this long sentence

into two sentences separated by a period. Rewritten, "Larger spatial and seasonal signals in atmospheric  $N_2O$  have been observed at sites influenced by strong local agricultural or

coastal upwelling sources (*Lueker et al.*, 2003; *Nevison et al.*, 2018; *Ganesan et al.*, 2020). However, at sites remote from local sources even variations of 0.2 ppb in estimated background N<sub>2</sub>O levels can significantly affect the magnitude of N<sub>2</sub>O emissions inferred from atmospheric inversions (*Nevison et al.*, 2018)."

• P3L75-76. "... phases in the eastern tropical Pacific (ETP.)". I suggest being more specific here

and mention sea surface temperature - something like: "... phases in sea temperatures over

the eastern tropical Pacific (ETP)". Done

- P3L82. "northern hemisphere" and "southern hemisphere" are already defined. Thank you for catching this, replaced here with "NH and SH."
- P4L90. I suggest changing "dynamics" with "dynamical processes". Done
- P4L113. "altitude-latitude cross sections" I suggest specifying that you are talking about

measurements here. Added "observed altitude..."

• P5L130-131. "with the premise .... of causation". This sentence belongs more to the Methods

section. Also, could you provide your reasoning (or some reference) regarding why

correlation is evidence of causation for this case? **Rewritten as**, "...with the assumption that significant correlations offer support, although not proof, of causation" We elaborate on this statement in Section 3.2 (see below) but include it briefly here in response to earlier reviewer comments that requested a clearer blueprint in the Introduction of why the correlation analysis was included.

• P6L154. I suggest swapping "116+-2" with "119+-2" since the authors highlight that the

lifetime is decreasing after 2000. Replaced with "from  $119 \pm 2$  yr in the 1990s down to  $116 \pm 2$  yr in the 2010s"

• P6L158. "they". Do the authors refer to the QBO and temperature here? If yes, could they

specify it? Yes, replaced with, "However, GEOSCCM QBO and PLST were computed in the same way as the observed indices..."

- P6L166. (HATS) (Thompson et al., 2004) -> (HATS, Thompson et al., 2004). Done
- P6L167. (CCGG) (Lan et al., 2022) -> (CCGG, Lan et al., 2022) Done
- P7L174. Is the "X2006A" necessary here? It sounds strange to someone not familiar with this

## terminology like me. Our NOAA co-author and lab expert Dr. Lan prefers to keep the name of the WMO scale, since it is meaningful to experimentalists.

- P7L182. Is 13 a subset of the ~55 sites mentioned before? If yes, could the authors specify it? Added, "at a selected subset of 13 of the ~55 total sites"
- P8L211. "HIPPO" not yet defined here. "the (HIAPER) Pole to Pole Observations (HIPPO) project" (HIPPO is an acronym within an acronym, taking its HI from HIAPER)

- P8L219. "HIAPER" not yet defined here. "the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER)"
- P8L223. "ORCAS" not yet defined here. O<sub>2</sub>/N<sub>2</sub> Ratio and CO<sub>2</sub> Airborne Southern Ocean (ORCAS)
- Section 2.3.2. This section contains only one paragraph and I suggest merging it with Section

2.3.1 to retain only Section 2.3. We have eliminated the subheadings 2.3.1 and 2.3.2 and relabeled 2.3 as Interannual variability in surface N<sub>2</sub>O for the correlation analysis.

- P10L265. PLST is already defined here. **Replaced with "plotted against PLST as described below."**
- P10L269-270 "PLST reflects .... (Holton, 2004)". This sentence belongs more to the

Introduction where the authors first talk about the PLST. Consolidated and rewrote as "The mean PLST in each hemisphere was treated as a proxy for the integrated strength of the BDC, which brings  $N_2O$ -poor air from the middle to upper tropical stratosphere into the polar winter lower stratosphere through diabatic descent. PLST represents the cumulative effect of descent throughout fall and winter, with warmer PLST corresponding to stronger descent (*Holton*, 2004; *Nevison et al.*, 2007; 2011)." We introduce the BDC in the Introduction but feel that these sentences are more appropriate here because we are describing why PLST is used as a proxy for the BDC. To go into this level of detail in the Introduction would potentially confuse and distract readers. Note, with this rewording we are also addressing the reviewer's request to remove references to the BDC bringing warm air into the lower stratosphere.

- P11L295. "0.4 degrees C" -> "0.4 degrees °C". **Replaced**
- Figure 1 caption. I suggest changing "...atmospheric N2O modeled...." with "... N2O mixing ratios [ppbv] modeled ...". For the captions of Figures 2, 3 and 4, I suggest something like "Anomalies of N2O mixing ratios [ppbv] ....". **Done for Figs 1-4 captions.**
- P15L364. I suggest adding "in the NH" after "altitude-latitude contours". Done
- Figure 5 caption. "... pressure-latitude contour plots arranged to form ....". I suggest re-

phrasing with something like "... pressure-latitude contours of anomalies of N2O mixing

ratios [ppbv] to form ....". Rewrote, "Sequence of five HIPPO pressure-latitude contours of anomalies of  $N_2O$  mixing ratio (ppb)..."

• P17L393-394. Given that the manuscript has shortened and become clearer, I would remove

the additional definition of PLST here. We can remove it if necessary, but given that it will be an unfamiliar acronym for many and this is its first mention in the Results, we would prefer to err on the side of repeating it here.

- P17L395. I suggest adding "(Section 2)" after "Methods". Done
- P17L397. Again, why do the authors assume that significant correlation between N2O AGR

and one index implies causal influence of that index on the N2O AGR? We have reiterated from the Introduction that correlations have limited usefulness and don't prove causation, but do provide reasonable support for causation, especially in the context of other evidence

from GEOSCCM and aircraft observations. "a significant correlation between the interannual variability in the N<sub>2</sub>O AGR and one or more of the indices can be interpreted to support a causal influence of the latter on the N<sub>2</sub>O AGR. However, correlation does not prove causation and cannot distinguish possible confounding effects, such as the influence of ENSO on both interhemispheric transport and surface sources."

• Figure 6. I suggest swapping panels 6b and 6c. This way, the discussion can focus on the QBO

first and then on the PLST (i.e., first discuss 6a and 6b, then 6c and 6d). We have rearranged the discussion to discuss 6a,b,c, and d in order, as for Fig. 7 below. We have also introduced Fig. 6 by describing what it shows and then presenting the salient results.

- Figure 6 caption. Can the authors specify the units of the AGR in the caption? Added "atmospheric growth rate (AGR in ppb/yr)"
- P19L421. I suggest introducing Figure 7 as was done for the previous figures. Rewritten as, "Figure 7, which presents the corresponding correlations for the NH surface N<sub>2</sub>O AGR, shows that,..."
- Figure 7. I suggest re-arranging the discussion of Figure 7 so it would smoothly describe 7a,

7b, 7c and 7d. In alternative, the authors could keep the discussion as it is and re-arrange the panels in Figure 7 accordingly (panels a and b for temperature and c and d for QBO). I find the current discussion of Figure 7 (7b, 7d, 7a, 7c) rather counter-intuitive. We have followed the first suggestion to rearrange the discussion.

- Figures 7 and 8 captions. As for Figure 6, I suggest specifying the units of the AGR in the caption. ? Added "atmospheric growth rate (AGR in ppb/yr)"
- P20L439. Also here, I suggest introducing Figure 8 as for the previous figures. Rewritten as "Figure 8, which shows correlations between the Niño 3.4 index and the surface N<sub>2</sub>O AGR, shows that that the two are significantly anticorrelated (R = -0.5, p < 0.05) for both the global and SH N<sub>2</sub>O AGR, …"
- P20L440. I suggest re-phrasing "... with little to no ...". Changed to "little or no"
- P21L450. I suggest re-phrasing "... of the seasonal cycle .... NOAA sites." with "... of the

seasonal cycle for NOAA sites at remote mid and high latitude." **Replaced with, "for NOAA** sites at remote mid and high latitudes."

• P22L458. "January". Why do the authors mention January if panel 9b and its caption say

February? Rearranged the sentence as, "This correlation is shown for February at South Pole in Figure 9b and is observed in both January and February at several extratropical southern NOAA sites including Cape Grim, Tasmania, Palmer Station, Antarctica, and South Pole."

• P22L464. Similarly to the comment above, why do the authors mention March if panels 9c,d

and their captions say February? **Reworded as, "GEOSCCM simulates similar correlations between PLST and austral summer N<sub>2</sub>O anomalies at these sites, both for N<sub>2</sub>O<sub>ST</sub> and total N<sub>2</sub>O in February (Figure 9c,d), and also March, i.e., the correlations are delayed by about 1 month relative to NOAA surface observations."** 

• Figure 9 caption. I suggest re-phrasing "a) ... mean seasonal cycle in N2O ..." with "a) ... mean

seasonal cycles in N2O mixing ratios [ppbv] for the NOAA surface station observations (Obs), and the GEOSCCM total N2O (N2Otot) and stratospheric N2O (N2Ost) ...". Also, I suggest re-phrasing "b) NOAA surface N2O seasonal anomalies ... " with "b) NOAA surface seasonal anomalies of N2O mixing ratios [ppbv] ....".

I also suggest a similar re-phrasing for "Bottom row shows seasonal anomalies for ...." with

"Bottom row shows seasonal anomalies of N2O mixing ratios [ppbv] for ...". Fig 9 Caption reworded as, "Figure 9: Top row shows a) mean seasonal cycles in N<sub>2</sub>O for NOAA surface station observations (Obs) and GEOSCCM surface total N<sub>2</sub>O (N<sub>2</sub>O<sub>tot</sub>) and stratospheric N<sub>2</sub>O (N<sub>2</sub>O<sub>ST</sub>) and b) NOAA surface seasonal anomalies of N<sub>2</sub>O mixing ratio (ppb) in February at South Pole spanning 1998-2020, plotted vs. mean lower stratospheric MERRA-2 temperature at 100 hPa averaged over 60-90°S over the previous spring (September-November). The labeled anomalies in 2016 and 2017 correspond to the year of the ORCAS and ATom-2 aircraft surveys, respectively. Bottom row shows GEOSCCM surface seasonal anomalies of N<sub>2</sub>O mixing ratio (ppb) for c) N<sub>2</sub>O<sub>ST</sub> and d) N<sub>2</sub>O<sub>tot</sub> in February at South Pole spanning 2000-2019, plotted vs. mean GEOSCCM lower stratospheric temperature, which is sampled the same way as the MERRA-2 temperature."

- P23L476. The authors mention "February" but the ORCAS dataset also covers January. Figure 9 originally showed panels for both January and February (NOAA) and February and March (GEOSCCM), (which has a delayed stratospheric signal at the surface relative to observations). In the first round of reviews, Reviewer 3 said that the extra panels (January and March) were redundant and should be deleted. Thus, February was the logical choice as the optimal single month to be featured in Figure 9.
- Figure 10 caption. I suggest changing "N2O anomalies in ppb ...." with "Anomalies of N2O

mixing ratios [ppbv] ....". Replaced with "Anomalies of N2O in ppb" -see comment 5 response for explanation of why we prefer ppb to ppbv.

• P24L490-494. I feel that this paragraph gives too much importance to a figure that is not

shown in the main manuscript. I suggest reducing this paragraph to a sentence that captures

its essence. Reduced to one sentence, "In contrast to the SH, PLST in the NH from the previous winter is not correlated significantly to N<sub>2</sub>O monthly anomalies at extratropical surface sites for either NOAA or GEOSCCM in any of the months surrounding the NH N<sub>2</sub>O seasonal minimum, with the exception of Mace Head, Ireland, where a negative correlation is found in July in GEOSCCM (Supplementary Figure S3)."

- P25L500. I suggest replacing "shows up" with "enters". Done
- P25L512. I suggest re-phrasing "...simulates too long a delay ..." with "... simulates a too long

delay ....". Done

• P25L513. "The rate of descent". Can the authors specify what is descending? **Replaced with** "diabatic descent"

P25L517. "... may be overestimated" why do the authors think that the summer soil emission might be overestimated? Was that a conclusion of Liang et al., 2022? Added some text and 2 references to better explain this point "summer soil emissions are from a soil biogeochemistry model and may be overestimated, leading to unrealistic surface maxima in July (*Saikawa et al.*, 2013; *Nevison et al.*, 2018; *Liang et al.*, 2022)." The model is from Saikawa et al. and is mentioned in Liang et al. The latter doesn't directly state the overestimate although Nevison et al., which used the Saikawa soil source, found that it overestimated summer emissions in North America.

- P26L534. Given my comment above about the warming due to the BDC, I suggest removing "warm". **Done**
- P26L539-540. I suggest moving the reference to Ray et al. 2020 at the end of the sentence and put it between parentheses (Ray et al., 2020). **Done**
- P27L562. "Paradoxically". Why do they authors say that? Did they expect something different? Added (i.e., when photochemical destruction is highest). The paradox being that less N2Opoor air is transported toward the poles during this configuration.
- Section 4.2. Very interesting section. However, I think it can be summarized and merged with Section 4.1 to highlight their main points. When doing that, I suggest clearly separating the discussion between the SH and NH (you could even have a subsection for the SH and one for the NH). We have consolidated 4.1 and 4.2 into one section. We did not find a good way to separate the SH and NH into subsections since the discussion moves back and forth between the two, comparing and contrasting.
- P28L589. I suggest removing "ENSO". Removed
- P29L611. I suggest re-phrasing "tease out". Replaced with "infer."
- All figures. Please replace the occurrences of "ppb" with "ppbv". See response above to comment 5 for explanation of why we stick with ppb.