# We thank the reviewers 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.

#### Reviewer 1 Farahnaz Khosrawi

Nevison et al. present a follow-up study of their publication in ACP from 2011. This is quite interesting and valuable study and deserves to be published, however the current version of the manuscript needs significant improvements.

Please find my detailed review with comments and suggestions for improvement in the attachment. Since I submitted a community comment to their manuscript in 2011 and am now referee of this study I think I can skip being anonymous.

#### Attachment

Review on egusphere-2023-2877 Northern vs southern hemisphere differences in the stratospheric influence on variability on tropospheric nitrous oxide by Nevison et al. Nevison et al. present a follow-up study of their publication in ACP from 2011.

This comment was visible to the reviewers as part of a cover letter statement to the editors, which asked why the paper was appropriate for ACP. However, an additional part of that statement, which apparently was not visible to the reviewers, was, "*This paper was inspired by competing results in the literature, which find alternatively that stratospheric influences or El Nino Southern Oscillation (ENSO) cycles are the dominant mechanism driving variability in tropospheric N20. ... Within the literature that identifies the stratosphere as the dominant influence, there are further disagreements and hemispheric differences regarding whether the quasi-biennial oscillation (QBO) directly influences surface N20. These questions are relevant to our ability to interpret biogeochemical source signals in the tropospheric variability in N<sub>2</sub>O ..." Thus while we appreciate the suggestions of Reviewers 1 and 2 to remove the discussion of ENSO and QBO, this is difficult because this discussion provided some of the major motivation for the study. Since the Reviewers' suggestions were made in large part due to the need to reduce the length of the paper, we have addressed their comments by shortening the paper and rearranging the figures in a more logical order, while also providing an improved background and motivation for the paper.* 

In their current study they present simulations with a GCM using a tagged stratospheric tracer of N2O to investigate the stratospheric influence on the seasonal cycle of tropospheric N2O. Additionally, they use the QBO and ENSO index and the polar lower stratospheric temperature as proxy for atmospheric transport and correlate these parameters with the N2O atmospheric growth rate. They find that the atmospheric growth rate anomaly in the southern hemisphere is better correlated to the QBO index, than to polar lower stratospheric temperature and that this hemispheric difference is consistent with the current knowledge of hemispheric difference in atmospheric dynamics. This is a very interesting study and a valuable continuation of the previous work of Nevison et al. (2011). However, the current version of this manuscript is completely overloaded. There is too much analyses presented and a lot of necessary information missing, this is especially the case for the result section. You cannot expect the reader to read several papers additionally to your current manuscript to be able to follow the presentation of your current results. On the other hand, a very extensive discussion section is provided which rather confuses than helps to understand the dynamical processes behind. Thus, I would suggest

major revisions before publication in ACP based on the comments and corrections given below. General comment:

• My major comment and suggestion for improvement would be to remove the entire part with the ENSO and QBO index and correlation of these to the atmospheric growth rate.

#### Please see response above for why we have not removed these sections.

I would suggest to instead focus on the experiment with the tagged stratospheric tracer which is a really great and convincing tool to study the stratospheric influence on the tropospheric seasonal cycle of N2O. From the correlation analyses part you could make a second paper where you also use model experiments to better substantiate the dynamical processes behind. So far it has been done in a rather speculative way than providing real proof. With skipping that part you would still have 8 figures plus 2 from the supplement, thus 10 in total which would be a reasonable number.

#### We have clarified how the tagged tracer was calculated but have not

• In many occasions your provide already statements on the results before you have shown the respective figure/analysis. This is not a good way of writing and causes only confusion. Thus, I would suggest that you first describe what is shown in the figures/what analyses has been done and then provide the results you gain with exactly stating what leads you to your specific result/conclusion.

Although Reviewer 2 made a similar comment, some of us have been trained since graduate school NOT to present results this way! We were taught that it is better form to describe the point made by the figure rather than open the sentence by flatly stating what the figure shows, which in any case should be explained already by the figure caption. We have tried to compromise by citing the figure number early in the first sentence describing the figure, including a clause briefly describing what the figure shows, but also including the main point made by the figure in that opening sentence.

• A lot of explanation/description of the figures/analyses is missing in the result section. It is really hard to follow what exactly has been done and why. This definitely needs to be improved. This of course will make your manuscript somewhat longer, but more valuable for the scientific community. Further, to have a not too long paper, we are back at my first comment to then skip the part with QBO and ENSO index, because the results of the tagged tracer experiment provide enough content for one paper.

We have reorganized the figures in a more logical order and, as described above, rewritten the Results to present figure number early in the first sentence describing the figure. We have also rewritten the figure captions to explain more clearly what is shown in the figure and have removed all interpretative sentences from the figure captions and moved them to the text.

• In the submission form you stated that your paper is a follow-up study of Nevison et al. (2011), but unfortunately, nowhere in the paper you state what is the new analyses/finding compared to your previous study.

Added a paragraph to the Introduction: "This paper explores the causes of variability in both the seasonal cycle and the AGR of tropospheric N<sub>2</sub>O. It follows up on previous work

by *Nevison et al.* (2011), who inferred a stratospheric influence on surface atmospheric  $N_2O$  observations based entirely on correlations between interannual variations in stratospheric indices and detrended  $N_2O$  anomalies in months surrounding the seasonal  $N_2O$  minimum. In the meantime, altitude-latitude cross sections have become available from aircraft surveys that span a full seasonal cycle. In addition, advances in model development allow for explicit simulation of stratospheric  $N_2O$  tracers (*Ruiz et al.*, 2021; *Liang et al.*, 2022)."

• Figure captions should generally only describe what is shown in the figure not provide any statements on the results or method. This should be rather described in the respective sections. Thus, all figure captions need to be revised.

#### All interpretative statements have been moved from the captions to the text.

Specific comments:

P1, L20ff: The abstract needs to be revised. See ACP guidelines. You solely describe your results, but you do not provide any introduction to the topic (e.g. why is it important?). Further, the abstract should end with some sentences stating what the importance and implications of your results are.

### The abstract has been revised to begin with a motivating introduction and end with a forward-looking conclusion.

P3, L82-83: This is a result and should not appear in the introduction.

#### Removed

P3, 87: In the introduction I would have expected more information on the tagging experiment you have done. What is tagging and for what is it used. Not every reader is familiar with this technique. So a short explanation is needed.

We have revised the Introduction sentence to read, "(GEOSCCM) ... includes a tagged stratospheric N<sub>2</sub>O tracer that is transported individually in the model and distinguished from tropospheric tracers of fresh surface emissions (*Liang et al.*, 2022)." We also added a fuller description of the tagging process in the Methods paragraph just below, "Following the approach of *Liang et al.* (2008), the tropospheric tagged tracers become the tagged stratospheric tracer, N2O<sub>ST</sub>, when they are transported across the tropopause, and retain that identity even when N2O<sub>ST</sub> re-enters the troposphere, thereby providing a model estimate of the stratospheric influence on tropospheric N<sub>2</sub>O."

P4, L102: Add here some sentences stating how good the model is plus references. Are there evaluation studies that show the quality of the model simulations especially with respect to atmospheric dynamics? That many models have problems simulated a proper QBO is discussed e.g. in Khosrawi et al. (2013).

Added, "GEOSCCM has been evaluated extensively in multi-model assessments and shown to represent well the mean atmospheric circulation, the interhemispheric exchange rate, the mean age of air in the tropical and polar stratosphere, and the mean atmospheric lifetime of N<sub>2</sub>O (*Liang et al.*, 2022 and references therein)."

P5, L137: "local Kriging"? What to you mean with "Kriging"? I have never heard about this and I guess you should provide some more explanation. Further, the reference should appear without parenthesis (should be embedded in the text).

The Kriging reference has been deleted – it is not really relevant to this paper. Also, "(Hammerling et al., 2012)" was replaced with "Hammerling et al., (2012)". P5, L150: What is meant with "2-4"? Are these the campaign numbers? Please clarify. Rewrote the description of the ATom campaign with, "ATom consisted of 4 deployments over 3 years, with each deployment approximately 1 month long (*Thompson et al.*, 2022). QCLS N<sub>2</sub>O was measured during the second through fourth ATom deployments in January/February 2017, September/October, 2017 and April/May 2018, respectively (*Gonzalez et al.*, 2021). (Note: N<sub>2</sub>O measurements are not available from the first ATom

#### deployment in July/August 2016 due to technical problems.)"

P5, L152: What does this mean? Is thus data missing for this time/time period? Please clarify and rephrase sentence accordingly.

### Rewrote as (Note: N<sub>2</sub>O measurements are not available from the first ATom deployment in July/August 2016 due to technical problems.)

P8, L236: It would be better if you first would describe what is shown in the figure and what kind of analyses you have done before you make statements of what the result is. What exactly in this figures does point to your interpretation that there is a stratospheric influence?

#### We have removed the old Figure 1

P10, L251: Not stratospheric loss. I guess you mean here stratospheric air or air characterized by low N2O.

#### Replaced "loss" with "air depleted in N2O"

P10, L257: This deviation could have many reasons. Either the model has deficiencies in the sources and sinks of N2O or atmospheric transport. Not clear if stratospheric or tropospheric processes are the cause of the shift. It could be that N2OST is correct, but N2O not.

### Agreed. We have added some sentences in the Discussion about the questions raised by the model. "

P10, Figure 3: You show 6 stations for the NH, but only 3 for SH. Why do you show less for the SH than for the NH. How have these stations been selected? What where the criteria for choosing these?

#### There are 4 sites in the SH and 5 in the NH, chosen to span a full range of latitudes. Section 2.2.1 also now includes, "All of the NOAA sites considered in this study, including Alert, Canada; Summit, Greenland; Mace Head, Ireland; Cape Matatula, Samoa; Palmer Station, Antarctica and the HATS baseline sites listed above, are long-standing remote sites situated away from strong local anthropogenic sources."

P10, Figure 3: This figure is overloaded. You do two analysis in one figure. First, you use observations to evaluate the N2O seasonal cycle from the observations. Then you use N2OST and the other curves to investigate the processes that affect the seasonal cycle. Thus, I would suggest to split the figure. One where you compare N2Oobs with N2O model and one where you show N2OST, N2Osoil, N2Oocean and N2Oanthro. The complete figure showing all curves in one plot as done now, could be provided in the supplement.

To reduce the overload in the figure we have combined N2Osoil, N2Oocean and N2Oanthro into a single line representing tropospheric emissions. This seems like a better solution than adding a separate figure, given the clear message from the reviewers that the paper is too long already.

P10, Figure 3: The amplitude of the seasonal cycle is max +-0.6. Assuming a tropospheric mixing ratio of 330 ppbv, this corresponds to less than 1 %. Thus, I would state that this is a minimal change and have difficulties to understand why is nevertheless still important to investigate the seasonal cycle of N2O?

Good point. We've addressed this issue by expanding a paragraph in the Introduction, saying, "High precision measurements of N<sub>2</sub>O have revealed interannual variability in its atmospheric growth rate (AGR) and small-amplitude seasonal cycles in the range of 0.4-1 ppb (i.e., 0.1 – 0.3% of the background mixing ratio) (*Nevison et al.*, 2004; 2007; 2011; *Jiang et al.*, 2007; *Thompson et al.*, 2013). Spatial gradients in atmospheric N<sub>2</sub>O are also small, e.g., the northern hemisphere (NH) minus southern hemisphere (SH) difference is approximately 1 ppb (*Thompson et al.*, 2014b; *Liang et al.*, 2022). While strong local sources can create larger spatial and seasonal signals in atmospheric N<sub>2</sub>O at some sites, such as those influenced by agriculture or coastal upwelling (*Lueker et al.*, 2003; *Nevison et al.*, 2018; *Ganesan et al.*, 2020), 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). "

P10, Figure 3: Another comment on the figure or the discussion of this figure in the paragraph above. This figure indeed shows a stratospheric influence on the seasonal cycle of N2O, but generally, I think there is now requirement that the minimum in N2OST overlaps with the minimum of N2O. Especially, with the known atmospheric transport time scales I would rather expect a time shift between the minima.

P11, L270: That descent is underestimated in atmospheric models has been shown in e.g. Brühl et al. (2007) and Khosrawi et al. (2009, 2018).

#### These references have been added to the discussion in section 4.0

P11, Figure 4: Also here, first the figure should be described and then the results you get.

#### Please see our general response to the Major Comment on introducing figures.

P11, Figure 4: Looking at the scale of the figure, the variation is +-1 ppb. As stated above, for a species having a mixing ratio of a few hundred ppb this change could be rated as being neglectable. Why is it nevertheless important to investigate the seasonal cycle?

### This issue is addressed now in the Discussion, as described in the response to the similar comment above.

P11, Figure 4 and respective text parts: Why is the anomaly strongest in summer. Shouldn't that rather be the case for winter/spring? Why do you detrend the data by using the de-seasonalized time series from Mauna Loa? Why do you do this and why is Mauna Loa used and not one of the other stations? Why you do what should be clearly described in the manuscript.

### The Methodology for the empirical background now describes why and how the Mauna Loa fit is removed.

P12, L295: It is not clear why you use the HIPPO data here. You provide an explanation based on altitude, but although you show somewhat different figures than the previous one (latitude vs altitude instead of time vs altitude), the altitude region considered is the same. So the reasoning you provide is not clear at all.

#### HIPPO is the only one of the 3 QCLS campaigns (HIPPO, ORCAS, ATom) that provides data over a full seasonal cycle. ORCAS was focused on summer in the SH, while ATom intended to cover a full seasonal cycle but the N2O measurements from the first deployment in July/August are not available due to technical problems)

P12, L292: Why is here now also the Atom data used? Repeat once again what your intention is of additionally using data from aircraft campaigns.

This is moved to the Discussion, in the context of describing the synoptic influences on the QCLS

P12, Figure 5: Also here, it is not clear to me why you de-seasonalize the data with the N2O from Mauna Loa.

# This is now described in the Methods. It is a site that allows us to remove a mean value characteristic of the mid troposphere and present altitude-latitude contour plots of $N_2O$ as anomalies.

P13, Figure 6: Why does the HIPPO data show positive values  $\Delta N2O$  in the NH polar regions. I would have (at least in the winter months) expected negative values. I think I am now completely confused, because there is so much explanations/descriptions on your analyses missing.

The color scales of the N2O anomalies between Figure 4 (-1.0 to 1.0 ppb) and Figure 5 (-5 ppb to 5 ppb) are necessarily different, since HIPPO extends into the lower stratosphere where the depletion signals are stronger. HIPPO also represents a synoptic snapshot north-south across a single longitude where positive anomalies can occur due to plumes associated with strong surface emissions. The contours in the HIPPO plots are negative down to the surface in the August panel and are also negative through the mid troposphere in June.

P13, L300: The section title does not really reflect what you are actually showing in this section. You show here correlations between T and delta N2O and not the minimum in N2O or the seasonal variation of it.

We have renamed the section "Correlation analysis of N<sub>2</sub>O seasonal anomalies." As noted in the methods, PLST is the only correlate considered because "QBO and ENSO are monthly indices for which it is not straightforward to choose a representative month to correlate to the N<sub>2</sub>O anomaly, given that the anomaly might result from the cumulative effect over multiple months. PLST in contrast has one unique value each year that can be plotted against that year's N<sub>2</sub>O anomaly for any given month."

P13, L301: There is a correlation but I would not call it significant. Have you checked if it is statistically significant? Is that what you mean here?

**The p value is now shown on the plot to support the claim that the correlation is significant.** P13, L304: In figure 4 however only the South Pole is shown.

It is noted in the text that South Pole is shown. Reviewer 3 requested that the number of panels in this figure be reduced to avoid showing largely redundant results.

P13, L310: What are Atom and ORCAS? Campaigns? Even though you have described your data in the method section it should be repeated here once again.

Introduced with a reminder as follows, "Figure 8, which compares February altitude-bylatitude QCLS N<sub>2</sub>O contour plots from the ORCAS and ATom airborne surveys, offers support for the observed surface correlations."

P13, L300ff: This is all too quick and condensed described. Figure 7 and 8 definitely need more description and explanation so that it is possible to follow your analyses and results.

Separate paragraphs are included for Figure 7 and 8 (now 6 and 7) and the description of them is expanded.

P14, L318-319: What does it mean that you derive only for Mace Head a significant correlation, but for all other stations not?

**It may not be meaningful and is moved to the end of the results and de-emphasized.** P14, Figure 7: You show here measurements and the results from the model, but why you show both and how they differentiate is not discussed in the text. Why then showing both? We have added a sentence to explain why both are shown, "The similarity of the observed and modelled correlations and the fact that they occur for  $N_2O_{ST}$ , which is not influenced by fresh surface emissions, as well as for total  $N_2O$  in GEOCCM, suggests they are caused by stratospheric influences."

P14, Figure 7: In this figure is too much information (see my previous comment). Also here it would be better to skip some of the subpanels or to split the figure into two figures.

#### The figure has been reduced from 6 to 4 subpanels.

P14, Figure 7 caption: "seasonal anomalies" since you look for minima I am confused, but the seasonal anomalies are also positive. Thus, as stated in my previous comment the section title is rather misleading.

#### The section title is changed to "Correlation analysis of N2O seasonal anomalies."

P15, Figure 8: In order that these figure panels can be more easily compared with each other, they should 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.

That 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.

P15, Figure 8 caption: The figure caption text is really confusing and needs more structure. Describe what is shown on the left and right (stick to the facts) and put everything else (results, reasoning etc.) into the main text.

The detrending and binning information has been moved to the Methods and all reasoning has been moved into the text.

P15, L339: Here, you should repeat again what your intention is with using the QBO and ENSO index.

We have added sentences at the beginning of Section 3.2 and 3.3 that state the intention of the correlation analyses for N2O AGR and monthly anomalies, respectively.

P16, Figure 9 and P17, Figure 10: Add a legend? What is shown by the different colors and line styles?

**The colors of the left and right Y axes are now matched to the respective lines on the graph.** P16, L345: Why is this correlation significant? Is it just statistically significant? From the correlation coefficient itself I would say that there is no or only a low correlation found.

The Introduction now includes some sentences acknowledging that the variability in N2O is very small, but that, "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)." Due to the small variability, the correlations between N2O anomalies and causal indices are generally weak, but still statistically significant, as indicated by p values < 0.05.

P18, L375: Finishing reading this section I was wondering why these correlations are shown at all. It seems that there is no correlation or anti-correlation found. Why then discussing the correlation between N2O and QBO and ENSO?

We also now begin Section 3.2 by stating the intent of the correlation analyses, "In this section NOAA surface  $N_2O$  AGR anomalies from 1998-2020 are plotted against polar lower stratospheric temperature (PLST) and QBO and ENSO indices, with varying lag times as described in the Methods. The analysis focuses on the NOAA global, NH, and SH mean products, with the premise that a significant correlation between the interannual

### variability in the N<sub>2</sub>O AGR and the stratospheric or ENSO indices can be interpreted to support a causal influence of the latter on the N<sub>2</sub>O AGR."

P18, L380ff: Here you start to describe the atmospheric processes in detail, but that should be rather done somewhere in the paper between method and result section or embedded at the respective places in the result section. In the discussion you should just put your results rather in broader context of the scientific field and discuss uncertainties, agreements or disagreements with previous studies etc.

The Discussion has been overhauled to remove this material. The BDC and QBO are now described more briefly in the Introduction.

P19, L385: I do not see this.

Replaced with, "These complex dynamics may explain why the N<sub>2</sub>O AGR in the NH correlates best with lower stratosphere QBO indices, why the correlation has a negative rather than positive sign (due to the vertical reversal of the sign of QBO with altitude) and why the correlations is weaker than in the SH."

(The wording in question was a relic from an earlier version of the paper that looked at specific NOAA surface sites.)

P19, L398: Why slow? Couldn't also fast descent bring N2O depleted air down? Wouldn't it be that the amount of N2O depleted air would differ dependent on slow or fast descent? **Deleted "slow"** 

P21, L456: What exactly is different in the NH compared to the SH? Wave activity is also high in the SH, so that alone cannot be responsible for the differences in dynamics between the hemispheres.

Have rewritten as, "In the NH, some of the same mechanisms and interactions between the QBO and BDC occur, but they are more difficult to isolate than in the SH due to the larger wave activity in the NH compared to the SH (Scaife and James, 2000, Kidston et al., 2015). "

P21, L473: I am not sure if the differences in dynamics between the southern and northern hemisphere as described in Holton et al. (1995) are still valid. This should be checked. There have been many studies and new findings on atmospheric dynamics in the recent years.

We are not familiar with a better reference than Holton et al. 1995 to support this point, but would be happy to include additional references if the reviewer has some specific ones in mind.

P23, L521: The discussion section is much too long, too detailed and too speculative. This section should be significantly shortened and rewritten so that it is more concise. Focus on what is really important and directly related to your study and your results.

#### The Discussion has been overhauled and shortened from 2430 words to 1679 words.

P23, L526: I would not say that the air accumulates there. The air descends during winter in the stratosphere and is then transported to the troposphere.

#### Replaced "accumulates" with "descends ... into"

P23, L526: When exactly the air crosses the tropopause has not been shown and also no references are provided. How do you know when and to what amount (e.g. N2O fluxes) N2O is transported down to the troposphere? You get some information on this from the tagged tracer in your model simulation experiment, but that should be clearly discussed here.

There are no references because this is a concluding statement but references are provided in the Discussion about how signals from the stratosphere are transmitted to the surface in GEOSCCM (Liang et al., 2008, 2009).

P23, L526: Another question on this text line is, what is happening with N2O on its way? It is not chemically reacting in the troposphere, but the N2O depleted air will mix with the tropospheric N2O rich air on its way that will result in that the stratospheric signal will get less and less as longer N2O is on its way to the surface.

#### Have added, "transmitting a diluted but still coherent signal."

P23, L533: Stratosphere-troposphere exchange and how it is linked to the BDC is not shown and also no references have been provided.

### All references to STE have been removed from the paper and replaced with a more general term, cross-tropopause transport.

References: The formatting of the references is not correct. See ACP manuscripts/guidelines. Further, there are several citations in the list that are not used, as e.g. Khosrawi et al. (2008) and Glatthor et al. (2007).

#### These references have been deleted.

Technical corrections:

P5, L134: magl -> m AGL

Done

P5, L149: four ~ month-long -> ~ four month-long

**Rewrote the description of the ATom campaign with, "ATom consisted of 4 deployments over 3 years, with each deployment approximately 1 month long** (*Thompson et al.*, 2022). P7, L194: The abbreviation PLST has not been introduced.

Defined as polar lower stratospheric temperature in the previous paragraph

P7, L198: Add which months you consider as winter months and which as spring months.

Added, "Winter months (January-March) were averaged in the NH and spring months (September-November) in the SH..."

P15, L333: Figure 8 caption: First sentence not correct. Please rephrase.

**Rewrote** N<sub>2</sub>O in ppb as a function of altitude and latitude from ORCAS (Jan.-Feb. 2016) and ATom-2 (Jan.-Feb. 2017). As in Figure 6, flight track data were interpolated onto a 5 degree latitude by 50 hPa grid using the akima package in R (Akima, 1978) and a deseasonalized fit to the NOAA time series at Mauna Loa was subtracted from all data.

P15, L334: I guess here it should read ORCAS and not Atom/Atom-2.

#### NA after rewording.

P15, L336-337: Sentence not clear. This needs more explanation. However, this should not be done in the figure caption, but in the main text.

#### Moved the interpretative part of the caption to the text.

P15, L338: Check title, some of the words should be rather starting with a small letter than a capital letter.

### ATom is the correct acronym for Atmospheric Tomography Mission, so the T is intentionally capitalized.

P17, L367: August-July -> July- August

This section is describing the 12 month intervals over which the N2O AGR was averaged in order to compare to PLST (which has only 1 annual value). Thus August-July is correct (i.e., beginning in August of a given year and extending through July of the following year. We have shortened and reworded the caption to avoid confusion.

P22, L505 and 506: The abbreviations OMZ and ETSP have not been introduced. **They were introduced in the old line 504 immediately above.** 

References:

Brühl, C., Steil, B., Stiller, G., Funke, B., and Jöckel, P.: Nitrogen compounds and ozone in the stratosphere: comparison of MIPAS satellite data with the chemistry climate model ECHAM5/MESSy1, Atmos. Chem. Phys., 7, 5585–5598, https://doi.org/10.5194/acp-7-5585-2007, 2007.

Khosrawi, F., Kirner, O., Stiller, G., Höpfner, M., Santee, M. L., Kellmann, S., and Braesicke, P.: Comparison of ECHAM5/MESSy Atmospheric Chemistry (EMAC) simulations of the Arctic winter 2009/2010 and 2010/2011 with Envisat/MIPAS and Aura/MLS observations, Atmos. Chem. Phys., 18, 8873–8892, https://doi.org/10.5194/acp-18-8873-2018, 2018.

Khosrawi, F., Müller, R., Urban, J., Proffitt, M. H., Stiller, G., Kiefer, M., Lossow, S., Kinnison, D., Olschewski, F., Riese, M., and Murtagh, D.: Assessment of the interannual variability and influence of the QBO and upwelling on tracer-tracer distributions of N2O and O3 in the tropical lower stratosphere, Atmos. Chem. Phys., 13, 3619–3641, https://doi.org/10.5194/acp-13-3619-2013, 2013.

#### **Reviewer 2 Anonymous**

Review of "Northern vs. southern hemisphere differences in the stratospheric influence on variability in tropospheric nitrous oxide" by Nevison et al. This manuscript is a follow-up study of Nevison et al., (2011) published in ACP. In this study, the authors use simulations of nitrous oxide (N2O) from a Chemistry-Climate model (GEOSCCM) together with ground-based and airborne observations of N2O to evaluate the impact of the stratospheric N2O on its concentrations in the troposphere. Both models and observations agree that N2O-poor air accumulates in the wintertime stratospheric polar regions and moves downward and equatorward, reaching the surface by early-autumn. The authors evaluate the impact of the ENSO, the QBO and the Brewer-Dobson Circulation (BDC, here quantified by the polar lower stratospheric temperatures) on the atmospheric growth rate of the surface N2O. They find that, in the Northern Hemisphere, the surface N2O atmospheric growth rate is negatively correlated with the polar lower stratospheric temperatures of the previous winter, and they attribute this correlation to the BDC. They also find that, in the Southern Hemisphere, the correlation between the surface N2O atmospheric growth rate and the polar lower stratospheric temperatures is not significant, but the correlations between the surface N2O atmospheric growth and the QBO and ENSO are significant. They argue that their findings are consistent with the current literature on the hemispheric differences in stratospheric transport and dynamics. The manuscript presents a coherent analysis and is generally clearly written. The authors use a large number of datasets that makes this comparison really valuable and worth publication in ACP. However, I have the impression that the manuscript needs some adjustments before final publication in ACP. My main concern is that manuscript misses to appropriately highlight "what's new" and does not

provide a possible outlook for future work. Therefore, I recommend publication in ACP once my comments below are addressed.

#### Major Comments

• Abstract. I suggest adding one/two sentences on "why this study is relevant". As it is, the authors mention right away what they did without providing information on why they did it. Similarly, I feel that the end of the Abstract lacks one/two conclusive sentences that highlight the main message of the paper and possible future paths to continue the work. Added, "The literature presents differing views on how variability in surface nitrous oxide (N<sub>2</sub>O) is influenced by the stratosphere and whether forcings associated with the El Niño Southern Oscillation (ENSO) outweigh those influences. These issues, which are relevant to interpreting biogeochemical source signals in tropospheric N<sub>2</sub>O, are investigated using...

• Introduction. The Introduction lacks a paragraph briefly describing the datasets used in the study: the GEOSCCM model and ground-based and air-borne observational datasets. Such paragraph could be added around P3L70. Concerning the novelty of the study, the current paragraph (P3L71-76) could be improved by explicitly stating that his study is a follow-up of Nevison et al., 2011 (this info is available only for reviewers, is there a particular reason?) and that the novelty arises from 12 years of model development and acquisition of additional observations.

Added a paragraph describing the datasets and another saying, "This paper explores the causes of variability in both the seasonal cycle and the AGR of tropospheric N<sub>2</sub>O. It follows up on previous work by *Nevison et al.* (2011), who inferred a stratospheric influence on surface atmospheric N<sub>2</sub>O data based entirely on correlations between interannual variations in stratospheric indices and detrended N<sub>2</sub>O anomalies in months surrounding the seasonal N<sub>2</sub>O minimum. In the meantime, altitude-latitude cross sections have become available from aircraft surveys that span a full seasonal cycle. In addition, advances in model development allow for explicit simulation of stratospheric N<sub>2</sub>O tracers (*Ruiz et al.*, 2021; *Liang et al.*, 2022)."

The Brewer Dobson Circulation (BDC) is mentioned a few times throughout the manuscript, but it is (briefly) described only in the Discussion section (Section 4.1). I think that such an important aspect of the manuscript deserves a few sentences in the Introduction (see also my minor comment about this below).

These sentences have been added to the Introduction: "The BDC is a planetary-wave-driven, large-scale meridional circulation that transports ozone, greenhouse gases, and other constituents poleward and maintains the thermal structure of the stratosphere (*Butchart*, 2014; *Minganti et al.*, 2020). As part of this transport, the BDC brings warm, N<sub>2</sub>O-poor air from the tropical middle and upper stratosphere into the polar lower stratosphere in the winter hemisphere (*Liang et al.*, 2008; 2009; *Nevison et al.*, 2011)."

The QBO also is described only in Section 2.4.2 and is hardly mentioned in the Introduction. Again, a few sentences about the QBO should appear in the Introduction (again, see also my minor comment about this below).

# This sentence has been added to the Introduction, "The QBO is a tropical, stratospheric, downward-propagating zonal wind variation with an average period of ~28 months that dominates the variability of tropical lower stratospheric meteorology (*Baldwin et al.*, 2001; *Butchart*, 2014)."

• Conclusions. The Conclusions lack to (re-)emphasize the novelty of the study and (more importantly) to highlight some strong forward-looking points of conclusions. As it is, the Conclusions reads more like a summary of what has been done. **Changed the name of Section 5 to "Summary and Conclusions"** 

To start, I suggest changing the title of the section from "Conclusions" to "Summary and conclusions". Then, I suggest reducing the repetition of information compared to the previous sections and provide a more detailed summary of what has been done (mention the model, observations, period, methods, a few numbers, ...). Finally, I suggest adding some additional points of conclusions, here's a few ideas:

The reviewer's suggestions are valuable but are beyond the scope of the authors' expertise, which is based more in aircraft data than satellite data. Satellite observations of N2O at least historically have mainly been used at ~ 100 hPa and above but do not resolve the smaller differences in the lower stratosphere and upper troposphere as well as aircraft data. We have therefore added some foreword-looking sentences about the importance of global airborne surveys to the Summary and Conclusion section.

o The authors find that the surface N2O growth rate presents hemispherical differences 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). o It would be interesting to add one/two sentences on the possible impact of the solar 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. o I suggest mentioning the possible added/complementary value of satellite measurements of tropospheric (and stratospheric) N2O (e.g., the IASI, MLS, ACE-FTS instruments). The authors could also mention the use of Chemistry-Transport Models driven by different dynamical reanalyses. Since N2O has a very simple chemistry, such analysis would provide some insights about the dynamics of each reanalysis. In addition, maybe it could be worth mentioning the possible extension to additional CCMs?

o The authors could mention the possibility to perform sensitivity tests with GEOSCCM. For example, an experiment with the QBO switched off (if possible) would isolate the patterns due only to the BDC.

o The authors mention a few times the "stratosphere-troposphere exchange". I suggest dropping that terminology in the manuscript (as it opens a whole research subject) but highlighting the importance of further investigating the cross-tropopause transport/mixing of N2O and its seasonality.

We have removed the term STE and used the reviewer's suggested term instead.

• ENSO. I have the feeling that the discussion about the impact of the ENSO is out of scope in this study for two reasons: o First, the title of the manuscript states "stratospheric influence on....". The ENSO is primarily a tropospheric mode of variability that ultimately impacts the stratosphere. Because of that, I do not think that the ENSO belongs in a manuscript that addresses the impact of purely stratospheric processes such as the QBO and the BDC on the surface N2O AGR. o Second, the impact of the ENSO on the N2O AGR is not as direct to understand as for the QBO and the BDC. In Section 4.4, the authors argue that the correlation between N2O AGR and ENSO are mostly due to tropospheric meteorology and biogeochemical processes (first and second paragraphs). In addition, the authors declare that the impact of ENSO on the N2O AGR is hard to evaluate because of possible complicated mechanism difficult to capture (third and fourth). The difficulty to provide a convincing discussion for the impact of the ENSO on the N2O AGR (contrarily to the QBO and BDC where the discussion is satisfying) is a sign that the ENSO should be removed from the manuscript (and maybe mentioned as possible future work in the Conclusions?). Hence, I suggest removing the ENSO parts of the manuscript as it is not a purely stratospheric mode of variability, and its impact of the N2O AGR is difficult to interpret compared to the QBO and BDC. In addition, the removal of the ENSO discussion will imply the removal of Fig. 11 and the related discussion together with Sections 2.4.3 and 4.4. I think that this would be beneficial for the manuscript as it will allow more space for possibly enhancing the discussion about the PLST and the QBO.

We appreciate this suggestion but are reluctant to eliminate the discussion of ENSO, since one of the main motivations in writing this paper was to address the relative importance of the stratosphere vs. ENSO in determining variability in surface N2O. The recent IPCC chapter on N2O, for example, emphasizes ENSO as the main driver and we want to present a different opinion. Instead of eliminating the ENSO section we have shortened other parts of the Discussion and have rewritten the Introduction as well as the Abstract to provide more background and motivation for why we are addressing this issue.

• Results. The Results section suffer from a flaw that makes the manuscript a little hard to follow: the Figures are never properly introduced. Each Figure is always mentioned between parentheses after one/two sentences describing the results in that figure (e.g., P8L236-238). This is confusing as the reader does not know where to look (and what to look at) when going through the text. I suggest introducing the Figures at the beginning of every paragraph where each Figure is discussed – something like "Figure 1 shows latitude-altitude cross sections of N2O anomalies for GEOSCCM for January to December...".

Although Reviewer 1 made a similar comment, some of us have been trained since graduate school NOT to present results this way! We were taught that it is better to describe the point made by the figure rather than open the sentence by flatly stating what the figure shows, which in any case should be explained already by the figure caption. We have tried to compromise by citing the figure number at the beginning of the sentence that first references the figure, including a clause briefly describing what the figure shows, but also including the main point made by the figure in that opening sentence. On a related note, we have revised our captions to remove all interpretative material in response to another request made by the reviewers.

Minor Comments

• P1L20 – as mentioned in my major comment above, I suggest starting the Abstract with one/two sentences on why this study is important.

Keeping in mind ACP's 250 word limit on the abstract we have started with a brief motivatin statement, "The literature presents diverging views on how the stratospheric quasi-biennial oscillation (QBO) influences surface N<sub>2</sub>O and has found distinct differences between the northern and southern hemispheres. These issues, which are relevant to the interpretation of biogeochemical source signals in tropospheric N<sub>2</sub>O, are investigated..."

• P1L25 – "... N2O atmospheric growth rate anomaly ..." could the authors specify the months when this anomaly occurs?

Rewrote as, "In the northern hemisphere, the annually averaged surface N<sub>2</sub>O atmospheric growth rate anomaly derived from long-term monitoring data is negatively correlated with winter (January-March) polar lower stratospheric temperature."

• P1L25 – "... winter's polar lower stratospheric temperature." Could the authors specify these winter months? As the seasons could be austral or boreal, I think that specifying the month(s) avoids possible confusion (even when the Hemisphere is mentioned).

Please see rewording in response to previous comment.

• P2L41 – I suggest adding the reference to Tian et al., (2020) together with Canadell et al., (2021).

#### Added Tian et al., 2020.

• P2L62 – the QBO here should be briefly explained/introduced. See also my major comment above.

Added here (moved from Methods), "The QBO is a tropical, stratospheric, downwardpropagating zonal wind variation with an average period of ~28 months that dominates the variability of tropical lower stratospheric meteorology (*Baldwin et al.*, 2001; *Butchart*, 2014)."

 $\bullet$  P2L65 – I suggest removing the acronym STE as it is barely mentioned in the rest of the manuscript.

#### Removed

• P3L71 – I suggest adding a paragraph introducing the different datasets used in this study: the GEOSCCM model and the airborne and ground-based observations. See also my major comment above.

Second to last paragraph in Intro: "This study uses the NASA Goddard GEOS-5 Chemistry-Climate Model (GEOSCCM), which includes a tagged stratospheric N<sub>2</sub>O tracer that is transported individually in the model and distinguished from tropospheric tracers of fresh surface emissions (*Liang et al.*, 2022). The study also examines atmospheric N<sub>2</sub>O data measured by recent global airborne surveys spanning both hemispheres and collected by the National Oceanic Atmospheric Administration (NOAA) during routine aircraft monitoring in the northern hemisphere. Finally, atmospheric N<sub>2</sub>O data from global ground-based NOAA monitoring sites are used in a correlation analysis of the N<sub>2</sub>O AGR against ENSO and QBO indices as well as polar lower stratospheric temperature (PLST), which reflects the influence of the BDC."

• P3L71-76 – This paragraph about the importance/novelty of the study should be improved. I suggest explicitly stating what is the added value of the study and that this is a follow-up of Nevison et al., (2011). See my major comment about this above.

Added a paragraph to the Introduction: "This paper explores the causes of variability in both the seasonal cycle and the AGR of tropospheric N<sub>2</sub>O. It follows up on previous work by *Nevison et al.* (2011), who inferred a stratospheric influence on surface atmospheric N<sub>2</sub>O

observations based entirely on correlations between interannual variations in stratospheric indices and detrended N<sub>2</sub>O anomalies in months surrounding the seasonal N<sub>2</sub>O minimum. In the meantime, altitude-latitude cross sections have become available from aircraft surveys that span a full seasonal cycle. In addition, advances in model development allow for explicit simulation of stratospheric N<sub>2</sub>O tracers (*Ruiz et al.*, 2021; *Liang et al.*, 2022)." • P3L77-87 – This paragraph should describe the structure of the manuscript, but it also introduces the GEOSCCM model (P3L80-82, this belongs to the paragraph describing the datasets mentioned above) and provides some anticipation of the results (P3L82-83 and P3L85-87). I suggest shortening this paragraph by moving the GEOSCCM part to the paragraph describing the datasets and removing the anticipation of the results. Furthermore, I suggest rephrasing the paragraph like: "Section 2 describes the data and methods used in this work, Section 3 ...."

All results have been removed from the final introductory paragraph, which now read as, "The paper is organized as follows: Section 2 describes the data and methods used. Section 3 presents the results, beginning with a subsection examining climatological mean seasonal cycles and latitude-altitude cross sections of N<sub>2</sub>O from GEOSCCM and aircraft data. Section 3.2 examines correlations between variability in the N<sub>2</sub>O AGR from NOAA long-term surface monitoring data, PLST, and indices of QBO and ENSO, with the premise that significant correlations offer evidence of causation. Section 3.3 examines correlations between PLST and variability in monthly N<sub>2</sub>O anomalies near the month of seasonal minimum. Sections 3.2 and 3.3 include parallel correlation analyses of variability in GEOSCCM N<sub>2</sub>O sampled at NOAA surface sites and model-derived QBO and PLST indices. Section 4 interprets and discusses the results. Section 5 concludes with a summary and conclusion."

 $\bullet$  P3L84 – The ENSO is already defined, no need to repeat the meaning of the acronym.

#### Removed

• P3L90 – The GEOSCCM acronym is already defined.

#### Removed

• P3L93-95 – Could the authors provide more details on the implementation/meaning of these four N2O tracers?

Rewritten as, "In addition to the standard total  $N_2O$  tracer, four additional  $N_2O$  tracers were included to track: 1) aged air from the stratosphere ( $N2O_{ST}$ ), and 2) soil, 3) ocean, and 4) anthropogenic sources freshly emitted in the troposphere. Following the approach of *Liang et al.* (2008), the tropospheric tagged tracers become the tagged stratospheric tracer,  $N2O_{ST}$ , when they are transported across the tropopause, and retain that identity even when  $N2O_{ST}$  re-enters the troposphere, thereby providing a model estimate of the stratospheric influence on tropospheric  $N_2O$ ."

• P3L96 – "... while a total N2O tracer ...." Is this "total N2O" the result of the sum of the four tracers mentioned before (N2Ost, soil, ocean, anthropogenic source)? If yes, I suggest saying it explicitly.

See above. Yes, effectively total N2O is the sum of the four tracers, but total N2O is also its own explicit tracer.

• P3L97-98 – "... 2000-2019 ..." could the authors mention the spin-up time for this run? The full answer to this question is somewhat complicated and described in detail in Liang et al., 2022. We have added a few sentences outlining the details, "The full GEOSCCM

simulation spanned 1980-2019, but this study focuses on the final 20 years from 2000-2019... As described in detail in *Liang et al.* (2022), the GEOSCCM N<sub>2</sub>O lifetime decreased slightly after 2000 (to  $116 \pm 2$  yr in 2010s down from  $119 \pm 2$  yr in the 1990s) and model emissions were gradually increased to keep model atmospheric N<sub>2</sub>O in balance with observations.

• P3L98 – "... 5 years of simulation ..." why did the authors select the last 5 years of the simulation for the seasonal cycle and not the last 10? A longer period would smooth the internal variability of the model.

This was an incorrect holdover from Dr. Liang's isotopic work (the isotopic cycles tended to blow up if run too long) and has been removed. In fact, the 20-year simulation from 2000-2019 was used to calculate the mean seasonal cycle of N2O and N2Ost.

• P4L107 – I suggest changing "(NOAA/HATS) (Thompson et al., 2004)" to "(NOAA/HATS, Thompson et al., 2004)".

HATS and CCGG have different references. NOAA/HATS and NOAA/CCGG have been changed to HATS (Thompson et al) and CCGG (Lan et al).

• P4108 – "(NOAA/CCGG)" is it possible to provide a reference here?

Lan et al. 2022 added.

• P4L109 - "... 5 baseline sites." I suggest mentioning the location of the sites.

Added: at Barrow, Alaska; Niwot Ridge, Colorado; Mauna Loa, Hawaii; Cape Grim, Tasmania; and South Pole, Antarctica.

• P4L115 – I suggest replacing "near" with "approach".

Done

• P4L118 – Is it possible to provide a reference instead of (or together with) the link?

Added Hall et al. 2007 (which is the reference given on the website).

• P5L137 – What is "Kriging"? I suggest explaining it or re-phrasing.

#### The Kriging reference has been deleted – it is not really relevant to this paper.

• P5L137 – I suggest changing "(Hammerling et al., 2012)" with "Hammerling et al., (2012)". **Done** 

• P5L138 - I suggest replacing "QCLS" (not defined yet) with "airborne".

Renamed the section as N2O data from global airborne surveys

• P5L143 – Could the authors provide a reference for the HIPPO project?

Yes, Wofsy et al., 2011

• P5L151-152 – "(Note: ....)" I suggest removing this sentence as the Atom-1 deployment is not mentioned further in the manuscript.

Rewrote the description of the ATom campaign with, "ATom consisted of 4 deployments over 3 years, with each deployment approximately 1 month long (*Thompson et al.*, 2022). QCLS N<sub>2</sub>O was measured during the second through fourth ATom deployments in January/February 2017, September/October, 2017 and April/May 2018, respectively

(Gonzalez et al., 2021). (Note: N<sub>2</sub>O measurements are not available from the first ATom deployment in July/August 2016 due to technical problems.)" It is relevant to note why ATom 1 N2O data are not available, so we prefer not to delete. Since ATom did not complete a full seasonal cycle for N2O, HIPPO is the only one of the 3 QCLS campaigns to

do so, which is why it is feature in Figure 5. This is noted in the text.

• P6L166-167 – "which was plotted .... As described below." This sentence does not really belong here as it already mentions plotting and proxies. I suggest removing it.

The sentence was moved to the beginning of Section 2.4.

• P6L180 – "BDC" the acronym was not introduced before. It should appear in the Introduction (where the BDC should be first described), see my major comment on that above.

#### The BDC acronym is now defined in the Introduction

• P6L183-185 – "(Note: ...)" This is a good point and I suggest keeping it as a normal sentence (i.e., without brackets and "Note"). Something like "We highlight that strong local...."

### Moved out of parentheses and reworked into an introductory paragraph describing the variability in N2O.

• P6L188 – "... the latter..." Do the authors mean the QBO and ENSO indices here? If yes, I suggest specifying it by adding "... the latter two indices..." or something similar.

Changed to, "The monthly N<sub>2</sub>O anomaly correlation analysis was applied only to the PLST BDC proxy, because QBO and ENSO are monthly indices for which it is not

#### straightforward to choose a representative month to correlate to the N2O anomaly"

• P7L193-194 – "... in winter/spring ... (September-November) in the SH ..." I suggest rephrasing this part to something like "... in January-March (winter/spring in the NH) and September-November (spring in the SH) ...".

#### Done

• P7L194 – Could the authors provide the meaning of the MERRA-2 acronym?

### Spelled out as the Modern-Era Retrospective Analysis for Research Applications, Version 2 (MERRA-2)

• P7L195 – "... stratospheric downwelling" I suggest adding "... stratospheric downwelling due to the BDC (e.g., Holton, 2004)."

#### Added

• P7L198 – I suggest adding the months between parentheses after "Winter months" and "spring months". Something like "Winter months (January-March) .... spring months (September-November)". I think it is better to have the explicit months together with the season when discussing the Northern and Southern Hemispheres.

#### Done

• P7L204-205 – This sentence is very pertinent for introducing the QBO. I suggest moving it to the appropriate new paragraph in the Introduction were the QBO and the BDC are first mentioned. See my major comment about this above.

#### This sentence was moved to the Introduction.

• P7L208 – Is it possible to have a reference together with the link?

### This is a product created by co-author P. Newman but does not have a dedicated published reference.

• P7L215 – The ENSO acronym was already defined before, there's no need to define it again. **Removed definition** 

• P7L216-217 – "El Nino is ... Kelvin waves (...)" this sentence should be moved to the Introduction where the ENSO should first be mentioned. However, given my major comment about removing the ENSO discussion, this point can be disregarded.

#### Moved to the Introduction with some rewording

• P8L231-233 – "(Note: ....)" This sentence about ENSO can be removed (if discussion about ENSO is removed) or re-phrased to avoid the "Note:" (if ENSO discussion is kept).

#### Rephrased to avoid "Note:"

• P8L236 – Figure 1 needs to be introduced. See my major comment above.

### We have decided to move this Figure to the Supplement. The old Figure 3 is now Figure 1 and leads off the Results section.

• P8L236-238 – "GEOSCCM simulates ... (SH) (Figure 1)" Could the authors specify the months of winter ("during winter in the polar lower stratosphere") and springtime/early summer ("in springtime (early summer)")?

#### Done

• P8L240 – "the surface minimum ... hemispheres" please rephrase with something like "the minimum in the lower troposphere does not reach the surface until ..."

#### Done

• P8L240 – please introduce Figure 2.

#### Please see our general response to the Major Comment on introducing figures.

P9, Figure 1 -Since the tropopause is mentioned in the text, it would be nice to show the tropopause level in Figure 1.

### This would be quite difficult to do so we request to be excused from this request. Also, Figure 1 has been moved to the Supplementary Materials.

• P9, Figure 1 - I suggest providing the units of the N2O anomalies on the right of the colorbar, instead of repeating them at the top right corner of each panel.

#### Done.

• P9, Figure 1, caption – "GEOSCCM N2O anomalies" please specify the units of the N2O anomalies.

#### The caption has been changed to, "Figure 1: GEOSCCM monthly N<sub>2</sub>O anomalies in ppb as a function of latitude and altitude extending from the surface up to 200 hPa (about 12 km). From left to right: January, February, March, April (top row); May, June, July, August (middle row); September, October, November, December (bottom row)."

• P9, Figure 1, caption – please replace "in a monthly sequence" with "as a function of". • P9, Figure 1, caption – "up to 30 hPa (24 km)" you state that you show from the surface to 30 hPa (24 km), but the top level of the panels of Figure 1 is 200 hPa (corresponding to 12 km). Could the authors explain that?

#### See rewording of caption above.

• P9, Figure 1, caption – please insert something like "From left to right: January, February, March, April (top row); May, ... (middle row); September, ... (bottom row)". **Done** 

### • P9, Figure 1, caption – "The GEOSCCM N2O .... the anomalies" this sentence belongs more to the text. I suggest moving it to the part describing Figure 1.

### See rewording of caption above and note below about moving Mauna Loa detrending to the methods Section 2.1.

• P9, Figure 1, caption – "Mauna Loa" the authors never mentioned Mauna Loa before in the text. I suggest moving this part to the text and mention the Mauna Loa sampling in the Methods section. There are other occurrences of the Mauna Loa sampling in the caption of other figures, therefore I suggest moving the Mauna Loa sampling description into the appropriate Methods section and not repeating it again.

We have included this statement in Section 2.1 and removed it from the Figure 1 caption: "The GEOSCCM  $N_2O$  fields were saved as monthly means and were detrended and converted to anomalies by subtracting a deseasonalized fit to the model time series sampled at Mauna Loa." • P9, Figure 2 – Also here, please provide the units on the right of the colorbar.

#### Done

• P9, Figure 2 – the bottom right panel (50S) has an additional label on the y axis (1000) that is not present in the other panels. I suggest removing it.

#### Removed

• P9, Figure 2 caption – please replace "N2O anomalies vs month" with something like "N2O anomalies as a function of month and altitude".

#### Done

• P9, Figure 2 caption – please rephrase "in the northern (top row) and southern (bottom row) hemispheres" with "in the NH (top row) and in the SH (bottom row)".

#### Done

• P9, Figure 2 caption – please specify the meaning of the columns. Something like: "From left to right:  $70^{\circ}$ ,  $60^{\circ}$  and  $50^{\circ}$ ".

#### Done

• P9, Figure 2 caption – "The GEOSCCM N2O fields ... relative to the NH" I suggest moving (and appropriately re-phrasing to fit the context) this sentence to the text where Figure 2 is introduced.

Moved into the text, "In Figure 2, the SH panels are plotted with a 6 month shift to help visualize the earlier seasonal phasing of the stratospheric influence in the NH relative to the SH."

• P10L252 – please introduce Figure 3.

### We have moved the old Figure 3 to Figure 1 because it most clearly illustrates that N2Ost dominates the total N2O seasonal cycle at most surface sites in GEOSCCM.

• P10L252 – I suggest replacing "stratospheric loss" with "N2O-depleted air". **Done** 

• P10L254 – I suggest adding "for all sites" after "N2Ost minimum".

#### Added, "... minimum at all extratropical sites" (MLO and SMO are exceptions).

• P10L255 – I suggest replacing "Southern Hemisphere" with "SH".

#### Done

• P10L256 - I suggest replacing "northern latitudes" with "latitudes in the NH".

#### Done

• P10, Figure 3 – I suggest swapping the panels for Summit (70N) and Barrow (71N). Barrow is northern than Summit and it would be consistent with the current layout of the panels (north to south from top left to bottom right).

#### Done

• P10, Figure 3 – I suggest removing the IDs of the stations on the bottom left corner of the panels (alt, sum, brw, ...) because the names of the sites are already in the titles of the panels. **Done** 

• P10, Figure 3 caption – please mention the names of the sites after "… 9 surface sites". Something like "Top row from left to right: Alert (Canada), …; middle row from left to right: Mace Head (Ireland), …; bottom row from left to right: Cape Grim (Country?), …". **Done** 

• P10, Figure 3 caption – "The black heavy ... observed N2O" should come before "the red line ...".

#### Done

• P10, Figure 3 caption – I suggest re-phrasing "The red line ... tracer N2Ost" with something like "For GEOSCCM, the total N2O from all forcings is in red, and the stratospheric tracer N2Ost is in dashed red".

#### Done

• P11L264 – please introduce Figure 4.

#### Please see our general response to the Major Comment on introducing figures.

• P11L264-265 – "When viewed as a ... up to 8 km" This sentence (properly re-phrased to correctly describe a figure) should be included in the description of Fig. 4.

#### Done

• P11L266 – please replace "is felt in" with something like "reaches".

#### Done

• P11L268 – again, please introduce Fig. 5 here.

#### Please see our general response to the Major Comment on introducing figures.

• P11, Figure 4 caption – as for Figures 1 and 2, the authors mention Mauna Loa in the caption with no reference in the text. I suggest moving (and expanding) this part to the Methods section. **Moved to Methods** 

• P12L285 – please introduce Figure 6.

#### Please see our general response to the Major Comment on introducing figures.

• P12L289 – I suggest removing "April".

#### Done

• P12L289 – "By June it has descended..." I find difficult to see that the anomaly has descended into the troposphere by June. I suggest re-phrasing with something like "By June it starts descending into ...".

#### Done

• P12L291-292 – "Notably, ... (Supplementary Figure 1)" this sentence states that the fuller dataset does not provide results as clear as the subset shown in Fig. 6. Do the authors know why? It would be interesting to provide a few words of explanation.

# The QCLS data represent snapshots that contain synoptic features (e.g., plumes of strong surface sources) that may obscure the stratospheric depletion signal, which as Reviewer 1 points out, is small < 1ppb against a 330 ppb background.

• P13, Figure 6 – The August panel of Figure 6 (for the NH) does not agree well with the corresponding August panel of Figure 4 (considering the same latitudes and altitudes). Do the authors know why? Maybe because the periods are different?

This is addressed now in the Discussion. The color scales are different, since HIPPO extends into the lower stratosphere where the depletion signals are stronger. HIPPO also represents a synoptic snapshot north-south across a single longitude. In contrast, the NOAA empirical background data are averaged as monthly means across 170W-50W. • P13, Figure 6 caption – could the authors explain the meaning of "transects"?

A transect according to the dictionary is a "line that cuts through a natural landscape (in the case of the figure by latitude) so that standardized observations and measurements can be made. In the case of the HIPPO and other QCLS data, it is not a line but rather a 2-dimensional plan spanning both latitude and altitude. We have added, "Each panel represents a north-to-south transect across with vertical profiling from the surface to 14 km."

• P13, Figure 6 caption – ".... an annual sequence." I suggest saying explicitly what contains each panel. Something like "an annual sequence; from left to right: January, March, June (top row) and August, November (bottom row)."

Done

• P13, Figure 6 caption – "HIPPO data extend ... influence on tropospheric N2O" This sentence (properly re-phrased to blend into the text) belongs to the text in the introduction of Figure 6.

• P13L301 – please introduce Figure 7.

Please see our general response to the Major Comment on introducing figures.

• P13L301 – "(PLST)" PLST is already defined – no need to define it again here.

Since most readers are probably not familiar with PLST as an acronym, and since it hasn't been mentioned since Section 2, it seems better to reiterate what it stands for rather than risk confusing readers.

• P13L303 – "seasonal minimum" I suggest adding the reference to the specific panel of the Figure (Fig. 7a).

#### Done

• P13L304 – I suggest removing the IDs of the stations (CGO, PSA, SPO).

Done

• P13L304 – I suggest changing "(Figure 7)" to something like "(Fig. 7b,c)".

Done

• P13L308 – I suggest changing "(Figure 7)" to something like "(Fig. 7e,f)".

#### Done

• P13L308 – "In support of ...." before this sentence please introduce Figure 8.

#### Please see our general response to the Major Comment on introducing figures.

• P13L308 – P14L313 – 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.

• P14L312 – I suggest changing "(weak Brewer Dobson circulation)" with "(weak BDC)". **Done** 

• P14L313 – I suggest changing "(strong Brewer Dobson circulation)" with "(strong BDC)". **Done** 

• P14L315-319 – this paragraph refers to generally unsignificant correlations for a Figure in the Supplement. I suggest reducing the importance of this paragraph to one/two sentences and move it to the end of the discussion of Figure 7.

#### Moved to the end of the section and trimmed.

• P14, Figure 7 – I suggest removing the bottom left panel (GEOS  $\Delta$ N2O vs GEOS Strat T) for two reasons: 1) it is not explicitly mentioned in the text and 2) it shows N2Otot from GEOSCCM that is not the most relevant tracer for this comparison.

Figure 7 has been reduced to 4 panels as per the request of a different reviewer and the text now explicitly mentions N2Ost, which is relevant because it suggests that the correlation shown is due to the influence of PLST on N2Ost and not fortuitously caused by surface surface sources.

• P15, Figure 7 caption "The correlation between ... its seasonal minimum" I suggest moving this sentence to the text.

#### Done

• P15L339 – please introduce Figure 9.

Please see our general response to Major Comment on introducing figures.

• P15L339 – P18L371 – Discussion of Figures 9 and 10. As it is, the discussion forces the reader to swap back and forth between Fig. 9 and Fig. 10 to follow the structure of the text (currently describing first the QBO in the SH and NH and then the PLST in the SH and NH). I suggest keeping the structure of the text as it is and re-arranging the figures accordingly: Fig. 9 should contain the QBO for both SH and NH, and Fig. 10 the PLST for SH and NH.

### We have restructured the text to first discuss the SH, both QBO and PLST correlations, referencing Figure 9 (now 8) and then the NH, referencing Figure 10 (now 9).

• P16L353 – please remove "of the weak".

#### Done

• P16L353 – please replace "correlations" with "correlation".

#### Done

• P17L364 - please add "(Fig. 9b)" after "NOAA".

#### Done

• P17L365 – please replace "(Figure 9b,d)" with "(Fig. 9d)".

#### Done

• P18L373-375 – based on my previous comment of the ENSO part, this discussion (and Figure 11) should be removed. However, if the authors decide to keep it, please introduce Figure 11.

#### Please see our general response to Major Comment on introducing figures.

• P18L382 – "vertical profiles" there is no vertical profile in the manuscript (for me vertical profiles are line plots with the values on the x axis and the altitude/pressure on the y axis). I suggest replacing "vertical profiles" with something like "latitude-pressure cross sections". **Done** 

• P18L382 – please re-phrase "big picture" with something like "broad".

#### **Broad-scale**

• P19L387 – please replace "Brewer Dobson circulation" with "BDC".

#### Done

• P19L394-395 – "which transports warm ... in the winter hemisphere" The impact of the BDC is not limited to the transport of N2O-poor air: the BDC also transports ozone, GHGS and CFCs and maintains the thermal structure of the stratosphere (Butchart, 2014, Minganti et al., 2020). I suggest re-phrasing this sentence to include also these important effects. In addition, based on my major comment above, the BDC needs to be described in the Introduction, so I suggest moving this description to the appropriate part in the Introduction.

Rewritten as, "The mechanistic pathway by which the stratosphere imparts a distinct seasonal signature to surface N<sub>2</sub>O is linked to the Brewer Dobson circulation (BDC), which transports ozone, greenhouse gases, and other constituents downward and poleward and maintains the thermal structure of the stratosphere (*Butchart*, 2014; *Minganti et al.*, 2020). As part of this transport, the BDC brings warm, N<sub>2</sub>O-poor air from the tropical middle and upper stratosphere into the polar lower stratosphere in the winter hemisphere (*Liang et al.*, 2008; 2009; *Nevison et al.*, 2011)."

• P19L394 – please replace "N2O-depleted" with "N2O-poor".

#### Done throughout

• P19L396 – "a large seasonal amplitude in the polar lower stratosphere" could the authors specify the amplitude of what?

### Rewritten as, "...leads to a large seasonal amplitude in the N<sub>2</sub>O mixing ratio in the polar lower stratosphere"

• P19L405 – I suggest replacing "upwells" with something like "is transported".

#### Done

• P19L410 – I suggest replacing "(~32 km) (Strahan et al., 2021 ....)" with "(~32 km, Strahan et al., 2021 ....)".

### Rewritten as, "...transports more N<sub>2</sub>O to its peak loss region around 32 km (*Strahan et al.*, 2021; *Ruiz et al.*, 2021)."

• P20L423 – I suggest replacing "in the July-September period" with "in July-September". **Done** 

• P20L430 – I suggest removing the quotes from surf zone. • P20L433 – I suggest removing "relatively".

#### Done

• P20L436 - also here, I suggest removing "relatively".

#### Done

• P20L436 – I suggest replacing "is felt at" with "reaches the".

#### Done

• P20L437 - I suggest replacing "permits" with "results in".

#### Done

• P21L456 – I suggest being more specific and replace "more complex atmospheric dynamics in the NH stratosphere" with something like "larger wave activity in the NH compared to the SH (Scaife and James, 2000, Kidston et al., 2015)".

#### Done

• P21L461 – "mixing of air" I suggest adding a reference here. For example, Shepherd, 2007. **Done** 

• P21L466 – I suggest removing the link because the text between parentheses is not a citation. **Done** 

• P21L474 – I suggest removing "stratosphere-troposphere exchange" as it was barely mentioned before in the manuscript. I suggest replacing it with something like "crosstropopause transport".

#### Done

• P22L496 - I suggest replacing "heightened" with "increased".

#### Done

• P23L516 – I suggest re-phrasing "testament to the strength" with something like "demonstration of the strength".

#### Rewritten as "demonstrated the strength"

• P23L522 – "Conclusions". As it is, the Conclusions section reads more like a summary than a Conclusion. Because of that, I suggest renaming this section as "Summary and Conclusions". In addition, the Conclusions section needs to re-emphasize what is new in this study and address more points of conclusion and possible future work. More on this in my major comment above.

We have added some foreward looking lines to the conclusions, renamed as the Summary and Conclusions, "To further refine our understanding of variability in tropospheric N<sub>2</sub>O, long-term monitoring at surface and aircraft-based sites is essential and would be complemented by more global airborne surveys extending into the lower stratosphere. The latter provide new insights into stratospheric influences on tropospheric N<sub>2</sub>O and advance our ability to interpret and quantify surface N<sub>2</sub>O sources."

• P23L527 – "summer to early-autumn period" could the authors specify the months here? Replaced with, "late summer to early autumn (August-September in the NH, April-May in the SH)."

• P23L530 – I suggest replacing "delivered" with "transported".

#### Done

• P23L532-533 – "Stratosphere-troposphere exchange". As one of my comments on this before, I suggest replacing "Stratosphere-troposphere exchange" with something like "Crosstropopause transport".

#### Done

#### References

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#### Reviewer 3 Anonymous

This study investigates the seasonal and interannual variability of N2O at the surface driven by transport from the stratosphere. This is a follow up of previous work by the first author on this topic with the addition of model output and aircraft data. The strength of the study is the use of aircraft data to fill in the seasonal transport picture from the tropopause region to the surface. Analysis of this type is important to more fully understand the causes of the variability of surface N2O and subsequently the variability of emission sources. But the manuscript needs to be revised to be more readable, primarily by focusing the discussion on the main points. Some specific comments are included below.

#### Specific comments

Section 2.1: You mention that the 'temperature and QBO are both internally generated by the GEOS GCM'. I assume that means this is a free-running simulation? That is, not forced by a reanalysis meteorology? It would be helpful to clarify this point. The model run is referenced to the Liang et al., 2022 study which likely clarifies the details of the run but it would be nice to have just a bit more information here.

# Added, "GEOSCCM temperature and QBO do not necessarily correspond to observations since both are internally generated by the GEOS GCM, which is free-running rather than forced by a reanalysis meteorololgy."

Section 3: Some of the figures could be consolidated. For instance, Figure 7b and c as well as 7e and f are so similar that it doesn't seem necessary to show them both. **We have reduced Figure 7 (now Figure 9) to 4 panels.** 

In Figure 8 it's hard to see a clear overall difference between these aircraft data sets. In some regions the ORCAS data seems lower. Maybe a difference plot between the two years would be easier to interpret and more concise.

The discussion has been expanded in Section 4.0 to acknowledge that, "QCLS data are measured across a narrow longitude band of the flight track for any given latitude on a limited number of days, while the NOAA empirical background is shown as a monthly mean, zonally averaged across most of the western hemisphere (170°-50°W).

Consequently, QCLS data are more likely to display synoptic-scale variability, such as the apparent surface source plumes over the Southern Ocean seen in Figure 10." (We have retained the original figure rather than a difference plot because it is easier to visualize than a difference plot, especially given the different spatial domains of ORCAS and ATom.)

There are also some figures that could use more discussion such as Figure 5. The contrast between the positive summer anomalies in the lower troposphere and the observed negative anomalies seems significant. Where is the positive anomaly in the model coming from? In general, only cursory mention of the differences between modeled and observed features are made rather than any insight into why the model differs.

The second paragraph in the discussion now addresses this, "the seasonality of

# surface N<sub>2</sub>O emissions may not be well represented in the GEOSCCM simulation, e.g., summer soil emissions may be overestimated, leading to unrealistic surface maxima in July (*Liang et al.*, 2022)."

Section 4: This section is too lengthy and repetitive. Since there aren't really any figures that focus on the stratosphere, aside from averaged metrics or the lowermost stratosphere, the discussion here asks a lot of the reader to follow the descriptions of stratospheric processes and is essentially a summary of previous work anyway. Some of the discussion in the section could be incorporated into Section 3 where the figures are discussed. But at a minimum this section should be considerably shortened to focus on the main findings, which as the title suggests is the NH vs. SH differences.

#### The discussion of the QBO and Brewer-Dobson circulation has been greatly shortened and summarized. The bulk of the original discussion was moved to a section in the Supplementary Materials for those interested in reading the details.

Line 520: The mention of a multivariate correlation that can explain more of the N2O variance almost seems like a throwaway here since it's just before the conclusions and there is no previous mention of it. Yet this seems like a promising result and worth more exploration or more discussion if the analysis has already been performed for the observations and model output.

These sentences were deleted, since we agree that there is not enough room to do the topic justice, especially as other reviewers are recommending that the discussion of ENSO be removed entirely.