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 air-borne 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 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

- <u>Abstract</u>. 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.
- 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.

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). 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).

• <u>Conclusions</u>. 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. 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 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).
- 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.
- 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?
- 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.
- 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.
- <u>ENSO</u>. I have the feeling that the discussion about the impact of the ENSO is out of scope in this study for two reasons:
 - 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.
 - 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.

<u>Results</u>. 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...".

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.
- P1L25 "... N2O atmospheric growth rate anomaly ..." could the authors specify the months when this anomaly occurs?
- 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).
- P2L41 I suggest adding the reference to Tian et al., (2020) together with Canadell et al., (2021).
- P2L62 the QBO here should be briefly explained/introduced. See also my major comment above.
- P2L65 I suggest removing the acronym STE as it is barely mentioned in the rest of the manuscript.
- 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.
- 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.
- 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 re-phrasing the paragraph like: "Section 2 describes the data and methods used in this work, Section 3"

- P3L84 The ENSO is already defined, no need to repeat the meaning of the acronym.
- P3L90 The GEOSCCM acronym is already defined.
- P3L93-95 Could the authors provide more details on the implementation/meaning of these four N2O tracers?
- 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.
- P3L97-98 "... 2000-2019 ..." could the authors mention the spin-up time for this run?
- 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.
- P4L107 I suggest changing "(NOAA/HATS) (Thompson et al., 2004)" to "(NOAA/HATS, Thompson et al., 2004)".
- P4108 "(NOAA/CCGG)" is it possible to provide a reference here?
- P4L109 "... 5 baseline sites." I suggest mentioning the location of the sites.
- P4L115 I suggest replacing "near" with "approach".
- P4L118 Is it possible to provide a reference instead of (or together with) the link?
- P5L137 What is "Kriging"? I suggest explaining it or re-phrasing.
- P5L137 I suggest changing "(Hammerling et al., 2012)" with "Hammerling et al., (2012)".
- P5L138 I suggest replacing "QCLS" (not defined yet) with "airborne".
- P5L143 Could the authors provide a reference for the HIPPO project?
- P5L151-152 "(Note:)" I suggest removing this sentence as the Atom-1 deployment is not mentioned further in the manuscript.
- 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.
- 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.
- 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...."
- 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.
- 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) ...".
- P7L194 Could the authors provide the meaning of the MERRA-2 acronym?
- P7L195 "... stratospheric downwelling" I suggest adding "... stratospheric downwelling due to the BDC (e.g., Holton, 2004)."
- 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.
- 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.
- P7L208 Is it possible to have a reference together with the link?
- P7L215 The ENSO acronym was already defined before, there's no need to define it again.

- 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.
- 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).
- P8L236 Figure 1 needs to be introduced. See my major comment above.
- 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)")?
- P8L240 "the surface minimum ... hemispheres" please rephrase with something like "the minimum in the lower troposphere does not reach the surface until ..."
- P8L240 please introduce Figure 2.
- P9, Figure 1 Since the tropopause is mentioned in the text, it would be nice to show the tropopause level in Figure 1.
- 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.
- P9, Figure 1, caption "GEOSCCM N2O anomalies" please specify the units of the N2O anomalies.
- 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?
- P9, Figure 1, caption please insert something like "From left to right: January, February, March, April (top row); May, ... (middle row); September, ... (bottom row)".
- 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.
- 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.
- P9, Figure 2 Also here, please provide the units on the right of the colorbar.
- 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.
- P9, Figure 2 caption please replace "N2O anomalies vs month" with something like "N2O anomalies as a function of month and altitude".
- 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)".
- P9, Figure 2 caption please specify the meaning of the columns. Something like: "From left to right: 70°, 60° and 50°".
- 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.
- P10L252 please introduce Figure 3.
- P10L252 I suggest replacing "stratospheric loss" with "N2O-depleted air".
- P10L254 I suggest adding "for all sites" after "N2Ost minimum".
- P10L255 I suggest replacing "Southern Hemisphere" with "SH".

- P10L256 I suggest replacing "northern latitudes" with "latitudes in the NH".
- 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).
- 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.
- 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?),".
- P10, Figure 3 caption "The black heavy ... observed N2O" should come before "the red line ...".
- 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".
- P11L264 please introduce Figure 4.
- 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.
- P11L266 please replace "is felt in" with something like "reaches".
- P11L268 again, please introduce Fig. 5 here.
- 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.
- P12L285 please introduce Figure 6.
- P12L289 I suggest removing "April".
- 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 ...".
- 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.
- 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?
- P13, Figure 6 caption could the authors explain the meaning of "transects"?
- 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)."
- 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.
- P13L301 "(PLST)" PLST is already defined no need to define it again here.
- P13L303 "seasonal minimum" I suggest adding the reference to the specific panel of the Figure (Fig. 7a).
- P13L304 I suggest removing the IDs of the stations (CGO, PSA, SPO).
- P13L304 I suggest changing "(Figure 7)" to something like "(Fig. 7b,c)".

- P13L308 I suggest changing "(Figure 7)" to something like "(Fig. 7e,f)".
- P13L308 "In support of" before this sentence please introduce Figure 8.
- 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)".
- P14L313 I suggest changing "(strong Brewer Dobson circulation)" with "(strong BDC)".
- 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.
- P14, Figure 7 I suggest removing the bottom left panel (GEOS ΔN2O 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.
- P15, Figure 7 caption "The correlation between ... its seasonal minimum" I suggest moving this sentence to the text.
- P15L339 please introduce Figure 9.
- 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.
- P16L353 please remove "of the weak".
- P16L353 please replace "correlations" with "correlation".
- P17L364 please add "(Fig. 9b)" after "NOAA".
- P17L365 please replace "(Figure 9b,d)" with "(Fig. 9d)".
- 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.
- 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".
- P18L382 please re-phrase "big picture" with something like "broad".
- P19L387 please replace "Brewer Dobson circulation" with "BDC".
- 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.
- P19L394 please replace "N2O-depleted" with "N2O-poor".
- P19L396 "a large seasonal amplitude in the polar lower stratosphere" could the authors specify the amplitude of what?
- P19L405 I suggest replacing "upwells" with something like "is transported".
- P19L410 I suggest replacing "(~32 km) (Strahan et al., 2021)" with "(~32 km, Strahan et al., 2021)".

- P20L423 I suggest replacing "in the July-September period" with "in July-September".
- P20L430 I suggest removing the quotes from surf zone.
- P20L433 I suggest removing "relatively".
- P20L436 also here, I suggest removing "relatively".
- P20L436 I suggest replacing "is felt at" with "reaches the".
- P20L437 I suggest replacing "permits" with "results in".
- 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)".
- P21L461 "mixing of air" I suggest adding a reference here. For example, Shepherd, 2007.
- P21L466 I suggest removing the link because the text between parentheses is not a citation.
- P21L474 I suggest removing "stratosphere-troposphere exchange" as it was barely mentioned before in the manuscript. I suggest replacing it with something like "cross-tropopause transport".
- P22L496 I suggest replacing "heightened" with "increased".
- P23L516 I suggest re-phrasing "testament to the strength" with something like "demonstration of 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.
- P23L527 "summer to early-autumn period" could the authors specify the months here?
- P23L530 I suggest replacing "delivered" with "transported".
- P23L532-533 "Stratosphere-troposphere exchange". As one of my comments on this before, I suggest replacing "Stratosphere-troposphere exchange" with something like "Cross-tropopause transport".

References

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