Review of the manuscript

Hemispheric asymmetry in recent stratospheric age of air changes

submitted to ACP by K. Dubé et al.

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General Comments

This study is a timely and useful contribution on the long-term changes in the BDC and their hemispheric asymmetry, a topic which is currently much discussed in the middle atmosphere community.

I found the text very well written and pleasant to read, except for the description of the derivation methods (see MC1 and SC1 below). I also feel that the submitted manuscript misses an important opportunity, as the discussion of ACE N_2O -derived AoA trends (section 3.2) does not explicitly compare the results with those presented in the modelling study which is the steppingstone for this paper (i.e. Ploeger and Garny, 2022): see MC2 below.

Since they do not imply any new model experiment or additional dataset, all suggested revisions are minor. Once they are addressed, I wholeheartedly recommend publication – preferably in *Atmos. Chem. Phys.*

Major comments

MC1. The derivations of trends in AoA interhemispheric differences (section 3.1) and of latitudedependent AoA derived from ACE-FTS retrievals of N₂O (section 3.2) both rely on the preliminary derivation of the linear regression coefficients (the "slope") between model-derived AoA and modelderived relative anomalies of N₂O (also done with CH₄ and CFC-12 in section 3.1). The derivation of these slopes is thus the key point of the methodology, and all studies aiming to derive AoA from observations of long-lived gases are expected to rely on similar methods. Hence it is important to describe this step in a clear and complete manner, which is currently not the case.

Rather than a cursory description aimed at explaining Figure 3, this should be expanded into one or two paragraphs and an additional figure to illustrate the derivation prior to showing its outcome. Focusing on the N₂O case, I would expect such a figure to show a scatter plot with N₂O relative anomalies on the X-axis and modelled AoA anomalies on the Y-axis, as well as the least-squares fit by a linear function (or better, three fits: one with the derived slope and two to show its $\pm 2\sigma$ uncertainty). Or maybe the axes of such a plot should rather be the *SH-NH differences* of the N₂O relative anomalies and AoA anomalies?

The methodology description in section 3.1 should also clearly state when the SH-NH differences (of N_2O and AoA) are computed: does this happen before or after the derivation of *relative* anomalies of $N_2O/CH_4/CFC12$ vmr?

The relative anomaly is itself defined (line 161) as "*the anomaly divided by the* [its] *overall mean*". I understand that these overall means have one single value for each species, pressure level and reanalysis, either for each hemisphere separately or directly for the interhemispheric differences. If that is correct, consider adding a figure in the Appendix to show the vertical profiles of these overall means. If this is not correct, please clarify.

MC2. This study is built on prior CLaMS simulations of AoA and long-lived tracers using four different reanalyses (Ploeger and Garny, 2022). This earlier paper showed latitude-pressure distributions of AoA trends (Fig.1a-1d in P&G2022), confirming that such CTM results are very dependent on their input reanalysis (Chabrillat et al., 2018). Hence, even though CTMs use as input observation-derived reanalyses of stratospheric dynamics, they cannot be used to unambiguously evaluate the ability of pure models (i.e. GCCM) to simulate the past evolution of the BDC. As explained in the introduction of the present manuscript, this is the motivation to derive AoA trends by combining CTM results with satellite-based observations of long-lived tracers: here ACE-FTS observations of N_2O are scaled by model-derived ratios ("slopes") representing the linear relationship between AoA and the tracer, as a function of latitude and pressure level, and this scaling is appropriately applied prior to the derivation of the trends.

The final figure shows very well that such "hybrid" derivations of AoA trends do not depend much on the input reanalysis used by the CTM, *thanks to* the injection of observations of long-lived tracers. Yet the final figures and their discussion (lines 198-233) fail entirely to mention the role played by observations. While this role is explained in prior sections and repeated in the conclusion, I still think that the discussion could be much improved by highlighting this point. Specific comments SC13-SC15 suggest some steps in that direction, especially SC15 to expand the last figure with CTM results.

Specific Comments

Original text is copied in italics, suggestions for corrections are typed in bold.

SC1. The words "*ACE-FTS gases*" are used throughout the text, with "gases" as a shorthand for "long-lived gas-phase tracers". This is not good wording because these "gases" are material substances, while you are manipulating numerical quantities which describe the variations in the abundances of these gases. To stick with rigorous vocabulary for remote-sensing measurements: the actual ACE-FTS "observations" are spectra of solar light partially absorbed by the atmosphere, while the volume mixing ratio (vmr) profiles which are delivered by the ACE-FTS team are "retrievals" from these observations. The text will become much clearer if you replace all relevant instances of "gases" by the quantities actually retrieved from ACE-FTS observations or derived from these retrievals, i.e. "ACE-FTS vmr" (volume mixing ratios) for the natively retrieved quantities or "ACE-FTS (relative) vmr anomalies" for the quantities which you derive from them. You will only need to define "vmr" once in section 2 or in the introduction of section 3.

SC2. The introduction is very well written. It only lacks an (often overlooked) reference to Garcia et al. (2011) which explains fundamental difficulties and limitations in deriving significant AoA trends from observations of trace gas species.

SC3. Line 29: "The BDC is the mechanism for long-range mass transport within the stratosphere".

SC4. Lines 42-43: reanalyses are *not* weather forecast models. They are datasets with high vertical and horizontal resolution providing, every few hours over several decades, physically consistent snapshots of atmospheric wind fields and temperature (and many other parameters not used here). These datasets are generated by NWP assimilation systems which combine weather model forecasts with many sources of homogenized observations. This is a good place to cite the S-RIP, either by citing its introductory paper (Fujiwara et al., 2017), the corresponding ACP/ESSD issue, or the whole S-RIP report.

SC5. Line 97: please provide some details about the initial states of these four simulations. When do they start? Do they all use the same initial conditions?

SC6. Lines 15-118: are these NOAA/GML in-situ observations are collected at the surface rather or the free troposphere? Please clarify.

SC7. Line 122: "...and interpolating the CLaMS output to the representative ACE-FTS profile locations and times at 30 km". Please clarify, e.g. "...and interpolating the CLaMS output to a location and time representative of the whole ACE-FTS profiles, using their locations and times at 30 km".

SC8. Figure 1: it is difficult to distinguish the ACE-FTS lines from the MERRA-2 lines (and probably not readable by color-blind readers but I do not know how to fix this). Please consider plotting ACE-FTS in black or grey, which is a common choice for observational datasets.

SC9. Lines 134-135: "there is a are clear differences between the datasets when considering changes over the *full* shared 17-year period.".

SC10. Lines 135-139: the year 2004 is not a very good exemple, as SH-NH AoA (fig. 1F) still differs for this initial year (e.g. anomaly of -1 year with MERRA-2 versus ~0.7 year with ERA-Int). Maybe use instead 2005 or 2006?

SC11. Line 150 and legend of Fig. 2: "...*ACE-FTS CFC-12 observations are only available up to ~8* hPa." It should still be possible to show the results for model-only correlations above 8 hPa (i.e. in panel C) ?

SC12. Line 188: as written, the sentence makes no sense. It is not possible to subtract "emission trends" from "ACE-FTS N2O observations" as these quantities do not have the same dimensions (strictly speaking, emission trends are mass/surface/time/time). Since you describe the actual procedure in the next paragraph, you could simply replace this sentence by a more general one introducing the next paragraph, e.g. "We give here a simple procedure to remove the effect of emission trends on trends of stratospheric N_2O . *This cannot*..."

SC13. The legends of Figs 5 and 6 should be clarified as follows:

Figure 5/6. AoA anomaly/trends derived from ACE-FTS retrievals of N_2O . Results are shown for using CLaMS driven with four different reanalyses. [...]

SC14. Line 217: "*The positive AoA trend in the NH is consistent with reanalysis results from Monge-Sanz et al.* (2022)". The cited reference is the chapter 5 of S-RIP report, which compiles many different papers discussing BDC trends using reanalyses. What specific result/figure are you referring? Please add a citation to the corresponding paper and develop this part of the discussion. **SC15.** I strongly suggest expanding Figure 6 by inserting a first row showing the corresponding "pure" CLaMS results, i.e. a repetition of Figs 1a-1d in P&G2022. The AoA trends should of course be recomputed using the same 2004-2017 period and MLR processing as used here, and the contour plots should keep the same, simpler design as in Fig. 6 (no potential temperature nor zonal wind contours; WMO tropopause very useful; hatching quite necessary). This would allow an easy comparison between earlier CLaMS results and the current "hybrid" results and could allow a quite fruitful discussion.

SC16. Line 263: "In general, global chemistry-climate models predict that..."

Typos, wording etc.

I did not find any: congratulations!

Additional bibliographical references

- Fujiwara, M., Wright, J. S., Manney, G. L., Gray, L. J., Anstey, J., Birner, T., Davis, S., Gerber, E. P., Harvey, V. L., Hegglin, M. I., Homeyer, C. R., Knox, J. A., Krüger, K., Lambert, A., Long, C. S., Martineau, P., Molod, A., Monge-Sanz, B. M., Santee, M. L., Tegtmeier, S., Chabrillat, S., Tan, D. G. H., Jackson, D. R., Polavarapu, S., Compo, G. P., Dragani, R., Ebisuzaki, W., Harada, Y., Kobayashi, C., McCarty, W., Onogi, K., Pawson, S., Simmons, A., Wargan, K., Whitaker, J. S., and Zou, C.-Z.: Introduction to the SPARC Reanalysis Intercomparison Project (S-RIP) and overview of the reanalysis systems, Atmos. Chem. Phys., 17, 1417–1452, <u>https://doi.org/10.5194/acp-17-1417-2017</u>, 2017.
- Garcia, R. R., Randel, W. J., and Kinnison, D. E.: On the Determination of Age of Air Trends from Atmospheric Trace Species, J. Atmos. Sci., 68, 139–154, <u>https://doi.org/10.1175/2010JAS3527.1</u>, 2011.