

We thank Eric Ray for her/his comments and careful reading and changed the manuscript according to her/his suggestions. Our response is formatted as follows:

Referee's comments

Author's reply

Changes to the manuscript

This paper uses in situ aircraft measurements and the CLaMS model to investigate the transport characteristics responsible for observed trace gas correlations in the polar lower stratosphere. This is nice work and really highlights the power of using model age spectra to better understand the causes of measured trace gas distributions in the stratosphere. The model does a reasonable job of reproducing the general features of the observed trace gases but the age spectra is what really explains why the features exist. The data and techniques are well described and the conclusion are well supported. My main comments are around the discussion of the tracer-tracer curves and the grammar, which could have used more work before submission. I suggest publication with consideration of the comments below.

Main comments:

- **Figures 3-5 and 8b:** I suggest changing the x and y axis ranges to eliminate white space and make the features more visible. You could change the Θ minimum to 290 K for instance.

Changed

- **Pg. 13:** I think this discussion of mixing and Figure 6 needs to be clearer. In Line 7 it is stated that “stratospheric CO will relax towards its stratospheric equilibrium value”. But that's not really how it works. CO has a chemical lifetime in the stratosphere so it's destroyed at a certain rate. In the absence of mixing or transport it will be completely destroyed. You should cite Minschwaner et al. (2010) here for the CO chemical lifetime discussion.

With the term 'equilibrium value' we refer to the equilibrium between CO production from methane oxidation ($\text{CH}_4 + \text{OH}$ is the rate limiting step) and the degradation of CO via OH. In the lower stratosphere there are two important reactions which determine the abundance of CO. The first one is the production of CO via methane oxidation and OH, the second one is the sink reaction of CO with OH to CO_2 , which is much faster than the production from methane. Since both reactions are driven by OH, the CO concentration in the lower stratosphere depends on the available OH and methane. In the absence of transport from the troposphere and subsequent mixing CO will be degraded rapidly, but not to zero since the CO production from methane, which has a long lifetime in the stratosphere of (190 ± 50) years (Brown et al., 2013) acts as a source as long as methane and OH are available. A steady state equilibrium is the result according to: $[\text{CO}]_{\text{eq}} = k_1/k_2 [\text{CH}_4]$ after solving $d[\text{CO}]/dt = k_1[\text{CH}_4][\text{OH}] - k_2[\text{CO}][\text{OH}] = 0$. Note that the value is independent from OH, since [OH] cancels out. Therefore in the long-term limit this leads to a CO value of 5-15 ppb_v depending on the integrated temperature history of the air masses. We used the term 'equilibrium value' to emphasize, that we

refer to a chemically driven equilibrium. As long as methane is present in the stratosphere, CO can be produced.

Looking at tracer tracer correlations, the CO-equilibrium can be seen in vertical branches of correlations, when using CO as x-axis (see e.g. Fig. 9). This vertical branch indicates vanishing CO variability and has been observed e.g. by Flocke et al. 1999 and Herman et al. 1999.

Therefore we kept the term 'CO-equilibrium value'.

The analysis of Minschwaner et al. (2010) focuses on the CO chemistry in the upper stratosphere and mesosphere with MLS measurements, where CO is enhanced from CO₂ photolysis and no equilibrium exists. As can be seen in Fig.7 and Fig.12 of the original manuscript a region constant CO for N₂O < 220 ppb_v is evident, which indicates this constant background CO_{eq}.

- In panel (d) I would recommend extending the blue curve up to the Chi_meso point since there is a background correlation curve that connects the stratospheric to the mesospheric values.

Changed. The figure now accounts for high CO values and zero N₂O in the mesosphere.

- Lines 23-24: In the discussion of Figure 7 it's not clear that it's remarkable CO is higher relative to N₂O in phase 2 compared to phase 1. The old air in the vortex that has come from high altitudes is expected to have relatively low N₂O and CO but is it expected that the correlation will remain constant, or that CO will be lower relative to N₂O? I just don't think it's well established what the correlation should be and if it is that should be justified by prior work.

Given that the fraction of descending aged air depleted in N₂O and SF₆ increases, one would expect at least not an increase in CO due to its much shorter chemical lifetime. Once CO is in steady state one would expect no sloped correlation at all (i.e. a vertical branch in Fig.7).

Lines 24-25: This sentence is too vague to understand what it is referring to.

Manuscript changed to:

It is important to note that the correlation along the mixing line which connects tropospheric values with the stratosphere shows higher CO relative to N₂O in phase 2. As indicated in Fig. 6 this is a clear indication for enhanced mixing of tropospheric air masses at for N₂O values > 273 ppb_v.

- Lines 26-28: What does the "direct tropospheric impact" mean? This sentence should also be clarified.

The expression 'direct tropospheric impact' should indicate, that the increase of CO relative to N₂O occurs relatively close to the tropopause at high (but stratospheric) values of N₂O (i.e. at most tropospheric influenced air masses). Here, air parcels which have been recently transported into the lowermost stratosphere have in general the shortest stratospheric residence time. It is not possible to derive from Figure 7 the information whether transport out of the TTL region occurred in this specific case or from the ExTL.

Therefore we changed the sentence to:

Therefore we can conclude that regarding the CO-N₂O correlation the tropospheric impact on short timescales through the ExTL was greater in phase 1 than in phase 2, [...]

- **Figure 14:** I'd suggest making these plots NH only to see the features and differences in the region of interest more clearly. It would also be interesting to see line plots at 350K and 400K for example of mass fraction vs. latitude for climatology and 2016.

We changed figure 14 to a 4x4 plot of January and March for the northern hemisphere only.

The below graph R1 shows line plots of the relative difference of air masses with transit times smaller than six months (MF06) from March 2016 to March of the climatology (thick line) at 350 K and 400 K.

At latitudes northwards 60° there are up to 10% more MF06 air masses as compared to the climatology, which also supports our hypothesis.

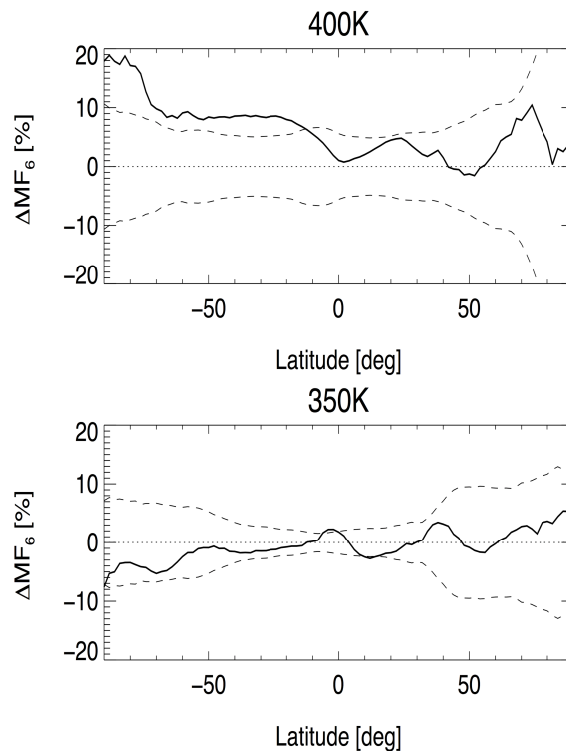


Fig. R1: Line plot of difference (thick line) between March 2016 and March of the climatology. Dashed lines denote the standard deviation.

- **Grammar comments:**
- **Pg. 2, line 6:** “these air masses”, what air masses are you referring to? Be more specific.

Manuscript changed to:

Air masses descending from the upper stratosphere and mesosphere chemically differ from the composition of the LMS, since they are potentially affected by ozone depleting catalytic cycles [...]

- **Pg. 2, line 23: comma needed after “vortex”**

Changed

- **Pg. 2, line 24: comma needed after “result”**

Changed

- **Pg. 2, line 25: “. . .establishes a relatively tropospheric. . .”**

Changed to the suggestion

- **Pg. 3, line 9: “. . .conditions existed due to. . .”**

Changed to the suggestion

- **Pg. 3, line 11: replace “was” with “were”**

Changed

- **Pg. 3, line 13: replace “to” with “on”**

Changed

- **Pg. 3, line 15: comma after second “warming”**

Changed

- **Pg. 3, lines 19-21: be consistent with use of either “eastward” and “westward” or “easterly” and “westerly”**

Changed to westerly and easterly

- **Pg. 3, line 24: “. . .El Nino could have accounted for a. . .”**

Changed to the suggestion

- **Pg. 3, line 28: comma after “TTL”**

Changed

- **Pg. 4, line 12: remove “the”**

Changed

- **Pg. 4, line 13: remove “the aim of”**

Changed

- **Pg. 4, line 16: remove “about”**

Changed

- **Pg. 4, line 18: replace “of” with “that measured”**

Changed to the suggestion

- **Pg. 4, line 22: add “and” between N₂O and CO**

Changed

- **Pg. 8, line 1: change “take” to “taken”**

- *Changed*

- **Pg. 8, line 2: change to “Green’s”**

Changed

- **Pg. 8, line 4: “. . .allows the calculation of time. . .”**

Changed

- **Pg. 8, line 9: change “formation” to “formulation”**

Changed

- **Pg. 8, line 10: “box model”**

Changed

- **Pg. 8, line 21: remove “respective”**

Changed

- **Pg. 8, line 27: change “constitute” to “contribute” and remove “, respectively”**

Changed

- **Pg. 8, lines 30-31: “. . .mean age from long-lived tracer measurements, the tracer must have a. . .”**

Changed to the suggestion

- **Pg. 9, line 26: change “the last” to “recent”**

Changed to the suggestion

- **Pg. 9, line 27: remove “an”**

Changed

- **Pg. 11, line 9: change “to” to “with”**

Changed

- **Pg. 11, line 15: not all of the CO decreases below 360 K.**

Manuscript changed to:

Note that the main increase is observed above $\Theta = 360$ K and 50° equivalent latitude. Below $\Theta = 360$ K more areas with decreasing values are encountered until there is no left increase at $\Theta = 340$ K.

- **Pg. 11, line 16: change “rise” to “make”**

Changed to the suggestion

- **Pg. 11, line 17: add “the” before “winter”**

Changed to the suggestion

- **Pg. 12, line 5: “. . .with air from the tropical lower stratosphere.”**

Changed to the suggestion

- **Pg. 12, line 9: change “of” to “the” and “as” to “of”, “. . .this increase originated. . .”**

Changed to the suggestion, but left the expression as the mesosphere, because the mesosphere is an example of one of the potential sources for stratospheric CO.

- **Pg. 12, line 10: “. . .TTL, into the extratropical lower stratosphere.”**

Changed to the suggestion

- **Pg. 12, line 11: add comma after “tropopause”**

Changed

- **Pg. 12, line 13: “. . .as a stratospheric. . .”, “used here as a tropospheric. . .”**

Changed

- **Pg. 12, line 17: “effects”**

Changed

- **Pg. 13, line 5: remove “actual”**

Changed

- **Pg. 13, line 6: “. . .correlation is established. . .”**

Changed to the suggestion

- **Pg. 13, line 26: remove “to” before “the”**

Changed

- **Pg. 13, line 32: add a comma after “before”**

Changed

- **Pg. 13, line 35: “. . .is the main source. . .”**

Changed

- **Pg. 16, line 18: change “by” to “in”**

Changed

- **Pg. 16, line 19: does the (3.7) refer to the uncertainty?**

Yes, changed to

6.8 +- 3.7 %

- **Pg. 16, line 30-31: “. . .information on the. . .”**

Changed

- **Pg. 17, line 12: solid lines, not dotted lines**

Changed

- **Pg. 18, line 4: add “the” after “as”, change “by” to “in”**

Changed

- **Pg. 22, line 5: “. . .average profiles throughout. . .”**

Changed to the suggestion

- **Pg. 22, line 6: change “from” to “of”**

Changed

- **Pg. 22, line 14: remove “to”, add comma after “(Fig. 5)”**

Changed

- **Pg. 22, line 27: “even though”**

Changed

- **Pg. 22, line 28: change “potentially” to “potential”**

Changed

- **Pg. 22, line 30: add comma after “Therefore”**

Changed

- **Pg. 22, line 32: “box model”**

Changed

- **Pg. 22, line 34: “calculated”**

Changed

- **Pg. 23, line 1: add “a” after “as”**

Changed

- **Pg. 25, line 3: “decreased”**

Changed

- **Pg. 25, line 4: “denoted”**

Changed

- **Pg. 25, line 17: “observed”**

Changed

References:

Brown, A. T., C. M. Volk, M. R. Schoeberl, C. D. Boone, und P. F. Bernath (2013). Stratospheric lifetimes of CFC-12, CCl₄, CH₄, CH₃Cl and N₂O from measurements made by the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS). *Atmos. Chem. Phys.* 13(14), 6921–6950, doi:10.5194/acp-13-6921-2013.

K. Minschwaner, G. L. Manney, N. J. Livesey, H. C. Pumphrey, H. M. Pickett, L. Froidevaux, A.

Lambert, M. J. Schwartz, P. F. Bernath, K. A. Walker, and C. D. Boone, "The photochemistry of carbon monoxide in the stratosphere and mesosphere evaluated from observations by the Microwave Limb Sounder on the Aura satellite," *J. Geophys. Res. Atmos.*, vol. 115, no. 13, pp. 1–9, 2010.