We thank Referee #1 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

The paper uses aircraft tracer measurements from the POLSTRACC campaign combined with simulations with the CLaMS model to derive conclusions on the characteristics of transport in the high-latitude boreal lower stratosphere for the winter of 2015-16. The results show an increase of the mean age in the region from January to March 2016, which seems at odds with the increase in CO concentrations. The authors argue that this is due to a change in the age spectrum, which exhibits an increase in both old and young air by the end of the winter.

The article is well written and presents an interesting analysis motivated by in situ observations and nicely complemented with modeling tools. I recommend publication in ACP after the following few minor comments and technical corrections are addressed.

> **The main point that should be addressed regards the high values of CO observed during phase 1, seen in Fig. 5a at about 70 degrees and 330-340K, and in Fig. 7 (CO values above 45 ppb_v). Although these values are not the main focus of the paper, they stand out, and there are a few parts in the paper where I miss some explanation of their origin. For instance, in the description on Page 13 Lines 25-29 it is mentioned that the 'direct tropospheric impact was greater in phase 1 than in phase 2' referring to these points. What do you mean by 'direct tropospheric impact'? Is this transport across the ExTL or did the high CO values originate in the TTL?**
>
> *The expression 'direct tropospheric impact' should indicate, that the decrease of CO relative to N_2O occurs at the highest stratospheric values of N_2O (i.e. at most tropospheric influenced air masses), where 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. The values you are referring to were encountered during one specific flight (PGS 09) on 22.01.2016. The flight track crossed a filament of air with relatively high values of tropospheric trace gases and a high tropopause with a sharp PV gradient. However, since small scale processes like gravity waves, occurrence of turbulence in regions of strong wind shear at the jet or diabatic heating violate adiabatic PV conservation this may lead to a mismatch of analyzed PV fields and tracer occurrence, which could also have caused the anomalously high CO values in this case.*
>
> *Since we analyzed our dataset only for PV > 7 PVU, we expect that the overall impact of the ExTL on our analysis is small.*

Also in Section 5.2, you could look separately at the age of air spectrum for those air masses, instead of showing the results for all measurement points in phase 1 together. Does that help in interpreting the origin? Finally, some measurements in phase 1 were taken at lower latitudes (over Italy) compared to the rest of the campaign. Does that latitude difference have an impact on the CO values?

The measurements over Italy do not affect our analysis of the observed CO increase relative to N_2O. Due to technical problems, we were not able to obtain N_2O measurements during this flight, so these data points do not appear in the CO-N_2O correlation and our analysis. Furthermore, these data points would be excluded by applying the 7 PVU criterion to our data as described in the manuscript.

The number of data points with high CO between 330 K and 340 K in this region is 297, compared to 5518 data points for the whole distribution of phase 1, which just makes a fraction of 5.3%. Since the data were observed in a region of strong PV gradients as described above and the main focus of the paper is on the region above Θ = 340 K, we did not analyze these age spectra separately.

We changed Fig. 2 of the manuscript. We now distinguish between parts of the flight track below PV = 7 PVU and above. We further removed the flight over Italy since the N_2O data are missing and do not contribute to our measurements.

P1 L22: 'diabatic descent [. . .] adds to the diabatic downwelling of the Brewer-Dobson circulation'. It seems to me you are referring to the same thing twice?

We wanted to refer to the two main processes which lead to diabatic descent during the polar night over the poles, namely the absence of radiation and associated diabatic cooling and the wave driven descent. We changed the section to:

Diabatic descent in the polar stratosphere, which is strongest inside the polar vortex results as part of the Brewer-Dobson circulation (Brewer, 1949; Dobson, 1956) in mid and high latitudes as response to the breaking of planetary and gravity waves (Haynes, 1991; Plumb, 2002; Butchart, 2014) in the upper stratosphere and mesosphere.

P2 L29: tropical pipe

Sentence changed to:

The region between Θ = 380 K and the bottom of the tropical pipe around Θ = 450 K (Palazzi, 2011) is a key region for the transition between these transport regimes.

P3 L23: The McPhaden reference is not about the 2015 ENSO event. A better option could be perhaps L'Heureux et al. (2017).

References changed to Chen et al. (2016) and L'Heureux et al. (2017)

P3 L24: The impact of the 2015-16 ENSO event on the polar vortex has been analyzed by Palmeiro et al. (2017).

Manuscript changed to:

A direct influence on the polar vortex is still under debate and according to Matthias (2016) this strong El-Niño is suggested to account for a weakening of the polar vortex, while Palmeiro (2017) found a connection of this ENSO event to the strong polar vortex and the early MFW.

Only flights that were used for the analysis are shown.

Caption changed to your suggestion.

P8 L1: take → taken

Changed

P8 L20: remove 'respective'

Changed

P8 L27: remove respectively?

Changed

P9 L7-8: is this a hypothesis or do you have an argument to support this statement?

This is a hypothesis based on the assumption that a change in the lifetime of SF_6 would lead to an equal change in the absolute values of mean age for both phases of the campaign. This assumption is also supported by the fact that we do not see any indication of an influence of mesospheric air on our observations. Since the discussion in our study is based on relative changes of mean age between phase 1 and phase 2 it is unlikely that the mesospheric loss of SF_6 affects the differential analysis of the calculated mean age.

P9 L14: Are the physical altitude ranges the same for both phases?

During both phases of the campaign the flight profiles were very similar and nearly every flight reached FL450 to FL480 (pressure altitude ranges to 45000 ft and 48000 ft, respectively).

P9 L18: The mean increase of 0.29 is just below the precision of the mean age estimate from SF6. Do you still consider it a robust change?

We consider the change as significant. Even if the observed change in the estimated mean age from SF_6 is just below the precision for each individual data point, one obtains a significant change in the mean binned mixing ratios of SF_6. Note that the increase of 0.29 years is only valid for the overlapping distribution of phase 1 and phase 2 (Fig. 3c)) and also the CLaMS simulations indicate an increase of the mean age of the same

magnitude, which is consistent.

The Fig. R1 below further illustrates the change of the mean age distribution towards higher mean ages based on the SF_6 distributions for the two phases. The increase of the mean age over all observed data of the distribution is 0.79 years.

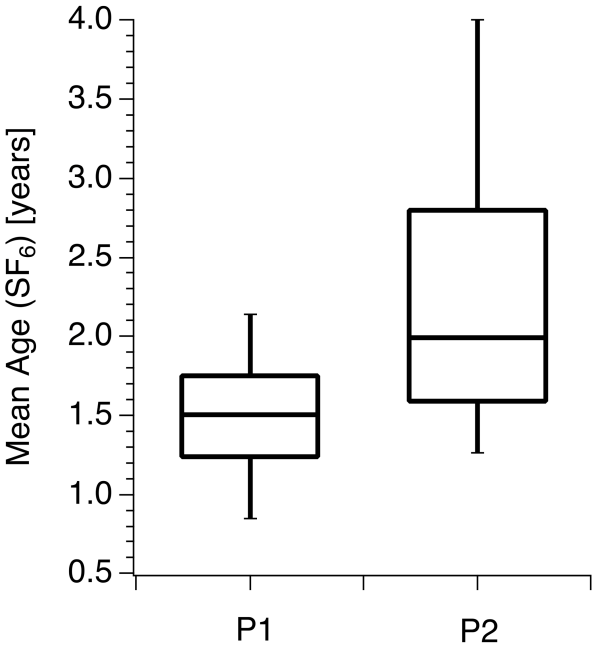

Fig. R1: Box-Whisker plot of Mean Age (SF_6) for phase 1 and phase 2.

P9 L27: "with an variability" → an interannual variability?

Manuscript changed to:

a meridional variability

P11 L2: chapter → section

Phrase „chapter" changed to "section".

These potential influences are discussed in section 6.

P12 L4: [...] general picture of enhanced downwelling of the Brewer-Dobson circulation [...]

Manuscript changed according to the suggestion

[…] which fits well in the general picture of enhanced downwelling of the Brewer-Dobson circulation in late winter/spring.

P12 L9: despite the

Changed

P13 L4-13: Could you refer to the individual panels of Fig. 6 as you describe the figure?

Changed

P13L21: anCO→aCO

Changed

P13 L25-29: This description is unclear. What you mean by 'shows higher CO relative to N2O?'. Perhaps it would make it easier to follow if you referred to the isentropic levels approximately corresponding to the N2O values when you describe Fig. 7 (it is hard for the reader to combine mentally Figs. 4, 5 and 7).

This sentence shall highlight the main result of this figure and refers to Fig.7. Higher CO values relative to N_2O are evident between N_2O = 275 ppb_V to 320 ppb_V. Compared to Fig. 6c), this is an indication that this change of the correlation can only be due to a change of the effectiveness of mixing. At this point we leave the geometric (or isentropic) coordinates since the tracer coordinate N_2O in Figure 7 serves as natural tropopause following coordinate.

If one would try to deduce the results from isentropic coordinates one could not differentiate between mixing and transport processes. Since both tracers will undergo the same transport and mixing processes these processes are accounted for in tracer tracer correlations. Relative changes of two tracers of very different lifetime like CO and N_2O therefore indicate changes of either sources or sinks or the transport efficiency.

P17 L4-5: would it be more accurate to refer to these figures as 'scatter plots' rather than 'correlations'? Also on Fig. 9 caption.

Changed to the suggestion

P17 L18: remove 'which is'

Changed

P18 L8: Green's function

Changed

P21 L1: what do you mean by 'mass balance systems of transport pathways'?

This refers to equation (4) and is changed to:

[…] the mass balance equation

The analysis of the CO-N_2O correlation and the mass balance equation as well as the model simulations consistently point towards …

P22 L27: Eventhough → Although

Changed

P24 L8-10: This sentence is unclear. Do you mean that the high fraction of young air reaches higher latitudes in 2015/16 as compared to the climatology? If so, what is the variability (e.g. standard deviation) around the climatology? Is this winter statistically different from the climatology?

The below graph R2 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.

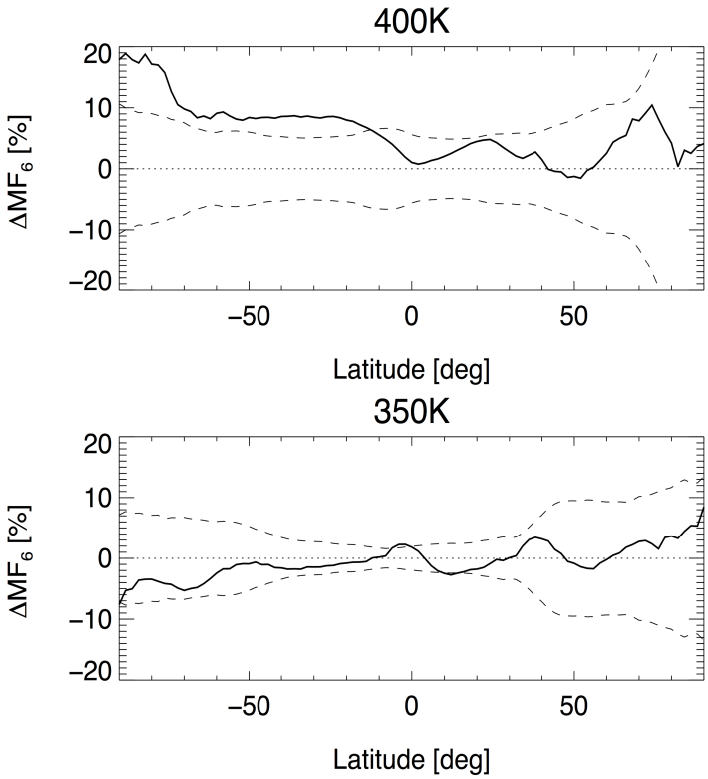

Fig. R2: Line plot of difference (thick line) between March 2016 and March of the climatology. Dashed lines denote the standard deviation.

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 fea- tures 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 (CH_4+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: $[CO]\_eq = k1/k2\ [CH_4]$ after solving $d[CO]/dt = k1[CH_4][OH] – k2[CO][OH] = 0$. 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 CO_2 photolysis and no equilibrium exists. As can be seen in Fig.7 and Fig.12 of the original manuscript a region constant CO for $N_2O < 220$ ppb_V is evident, which indicates this constant backround 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 N_2O 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 N_2O and SF_6 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 N_2O in phase 2. As indicated in Fig. 6 this is a clear indication for enhanced mixing of tropospheric air masses at for N_2O 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 N_2O occurs relatively close to the tropause at high (but stratospheric) values of N_2O (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-N_2O 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.

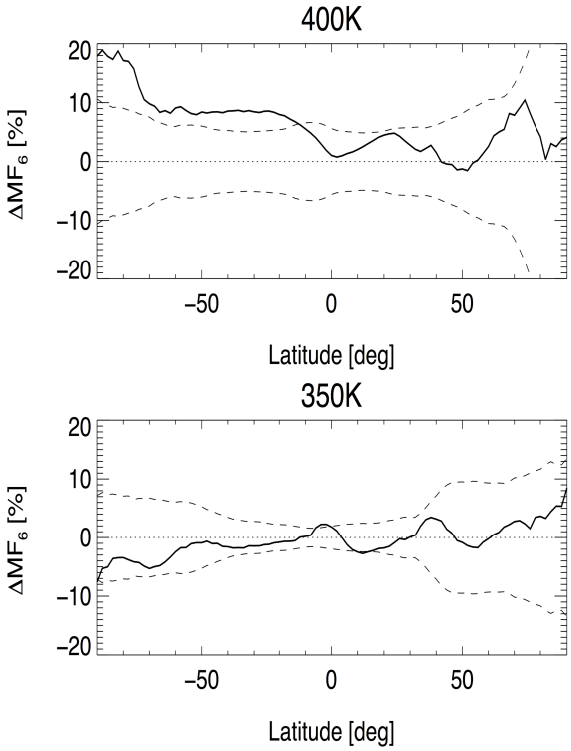

Fig. R1: Line plot of difference (thick line) between March 2016 and March of the climatology. Dashed lines denote the standard deviation.

- **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 Θ = 360 K and 50° equivalent latitude. Below Θ = 360 K more areas with decreasing values are encountered until there is no left increase at Θ = 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.

We thank Referee #3 for her/his comments and careful reading and changed the manuscript according to her/his suggestions. Our response is formatted as given below:

Referee's comments

Author's reply

Changes to the manuscript

This paper analyzes aircraft observations of N2O, SF6, and CO from a recent polar aircraft campaign to show that the Arctic lowermost stratosphere has diabatic descent between January and March (this is well known, not new), and that while the meanage increases, CO measurements indicate that younger air mixes into the vortex during winter. A trajectory model simulation of the same period is used to support their interpretation of the measurements. The main result presented is that when a region of the atmosphere gets older, it doesn't mean that younger air didn't mix in! That is, there can be simultaneous changes in the age spectrum where both ends of the distribution change. The results show how 2 tracers with different lifetimes can be used to identify simultaneous contributions from young tropospheric and older stratospheric air. This isn't exactly how the message is stated but this is the interesting result.

The analysis of the aircraft data set is fine, the results are not surprising; they are consistent with what we know about the BDC and horizontal mixing in the LMS. Because the results are consistent with expectations, please present the data and analysis more succinctly.

The paper is far too long for what it has to say and it is not clear what the motivation is for this study. There is too much rudimentary discussion that is inappropriate for a journal article; some of the CO production and loss processes discussed are not actually significant or relevant. It feels like I am reading a thesis chapter not a journal article.

There are minor scientific issues regarding assumed CO values from the TTL and the assumption that CO in the lowermost stratospheric vortex is coming from the TTL. The assumed CO values are out of date (too low) and ignore copious recent data from MLS. There is large seasonal variability in tropical CO due to biomass burning (i.e., the CO tape recorder). Revisions should be made with up to date CO and this will change the results. I am not your thesis advisor but I would like to help you produce an article that others will be interested to read. There are many suggestions below intended to improve the readability of this article. This study is worth publishing after considerable shortening. This paper has 14 figures and well over 7000 words, but there is one main result.

General science and writing suggestions

The BDC is driven by waves. Wave energy deposited in the polar region warms the stratosphere which then radiatively relaxes, resulting in diabatic descent of polar air. So please do not say (p. 1, line 22) that diabatic descent in the vortex ADDS to the BDC. It is PART of the BDC. This occurs several times in the paper.

Changed accordingly throughout the manuscript.

It is very rare that it makes sense for a paragraph to have only 1 or 2 sentences. Please review your choices of paragraph breaks. The small paragraphs here generally belong in a nearby paragraph.

Changed wherever possible

The words 'respective' and 'respectively' are misused in nearly every occurrence. Even when used correctly here they are not necessary. Please delete all uses of these words.
Regarding the use of passive voice, many science papers unfortunately use expressions like 'In Figure 3, it is evident that ...'. A paper will be more concise and interesting to read with 'Figure 3 shows ....'. I suggest eliminating passive voice wherever possible. In general, replace 'it is evident that' with 'shows'. And 'it is possible to analyse' with 'we analyzed'.

We eliminated the frequent use of passive voice and inserted the suggested expressions. Note further, that remaining grammar and language issues will be handled by production office during the copy editing process.

Line by line

Introduction. What's missing here is any motivation for this study. Why are you investigating these data sets? Why would a reader want to continue reading?

We highlighted the motivation in the first sentence and changed the order of the introduction:

Uncertainties in the description of mixing introduce large uncertainties to quantitative estimates of radiative forcing (Riese et al., 2012) which are on the order of 0.5 W m^-2 (Riese et al., 2012). Therefore it is important to quantify the contribution of the dynamical processes which act on the distribution of tracers. The arctic UTLS during winter is affected by diabatic descent from the stratosphere and quasi horizontal mixing by the shallow branch of the Brewer-Dobson circulation, which connects the tropical tropopause region with the high arctic (e.g. Rosenlof et al., 1997; Birner and Bönisch, 2011).
We present data from winter 2015/2016, which were measured during the POLSTRACC (The Polar Stratosphere in a Changing Climate) aircraft campaign between December 2015 and March 2016 in the Arctic upper troposphere and lower stratosphere (UTLS).
The focus of this work is on the role of transport and mixing between aged and potentially chemically processed air masses from the stratosphere with mid and low latitude air mass fractions with small transit times originating at the tropical lower stratosphere. By combining measurements of CO, N_2O and SF_6 we investigate the evolution of the relative contributions of transport and mixing to the UTLS composition over the course of the winter.

p. 1, line 7: "We find an increasing influence ..." Where?

We added a more precise description of the measurement region to the beginning of the abstract.

We find an increasing influence of aged stratospheric air partly from the vortex as indicated by decreasing N_2O and SF_6 values over the course of winter in the extratropical lower and lowermost stratosphere between Θ = 360-Θ = 410 K over the North Atlantic and the European Arctic.'

p. 1, lines 22: diabatic descent does not add to the diabatic downwelling of the BDC. It is a part of the BDC. Same issue on
p.2, line 7 and line 32. 'strong diabatic descent the wave-driven BDC'. Again, these are one and the same thing. And later, mixing caused by breaking planetary waves is not 'in addition to' the BDC. Breaking planetary waves are what drives the BDC.

We changed the manuscript to the suggestion. The stratospheric circulation including the descent in the polar vortices is of course driven by the wave-driven Brewer-Dobson-circulation. The usage of the word ADD should not imply that there are two distinct processes resulting in downward transport of the polar winter stratosphere.

During winter the UTLS-region (Fig. 1) at high latitudes is strongly affected by the evolution of the polar vortex. Diabatic descent in the polar stratosphere, which is strongest inside the polar vortex results as part of the Brewer-Dobson circulation (Brewer1949,Dobson1956) in mid and high latitudes as response to the breaking of planetary and gravity waves (Haynes1991,Plumb2002,Butchart2014) in the upper stratosphere and mesosphere. […] These air masses are rapidly mixed quasi horizontally by breaking planetary waves with descending aged air from high latitudes as part of the shallow branch of the Brewer-Dobson circulation (Birner2011,Abalos2013). […]

p. 2, line 3, 4: 'deep stratospheric air masses' Unclear. Can you be more specific than 'deep'? Define the LMS. Yes it's in the figure, but say here the latitudes, pressures, Θs, etc.

Changed. We added definitions from literature, where available.

This downwelling leads to an increasing contribution of stratospheric air masses from the overworld (defined as the region, where isentropes are entirely located in the stratosphere, Hoskins, 1991). Over the course of winter they contribute to the composition of the lower overworld ($\Theta < 420$ K), where our measurements took place, and the lowermost stratosphere (LMS) (Rosenfield et al.,1994) (defined as the region bounded by the 380 K isentrope and the extratropical tropopause (Rosenfield1994, Holton1995))

p. 2, line 6: The first sentence of the paragraph refers to 'these' air masses. Which ones?

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 (Solomon1999).

p. 2, line 29: there is only a tropical pipe, no subtropical one.

Changed

p. 2, line 34. The reason H2O and O3 are modified by this transport is because they have large gradients. This is the underlying idea you might want to mention.

We want to refer to air masses, which have entered the stratosphere across the TTL region and the 380 K isentrope. We changed the sentence:

Above $\Theta = 380$K these air masses, which ascended through the TTL […]

and

This rapid transport modifies the abundance of particularly water vapour and ozone in this region, which have seasonally varying isentropic gradients (e.g. Plöger et al.2013) […]

p. 3, lines 1-4. These 2 sentences about radiative effects come out of nowhere. What

do they have to do with the rest of the introduction? Do you ever discuss radiative effects again in the paper? (I don't think so.) Either integrate these sentences into the context of this introduction or delete them.

We followed the suggestion and added a paragraph at the beginning of the introduction (see comment above related to the general motivation at the beginning of the introduction).

p. 3, line 5. Define what you mean by subvortex. The vortex extends below 410K, so this is unclear.

At the maximum flight ceiling of HALO, which was 15km during POLSTRACC, we couldn't reach the interior vortex, but frequently encountered large filaments of vortex air as evident from the trace gas composition. We use the term "subvortex region" for the isentropes below 420 K (approximately 120 hPa) where the lower vortex exists, but starts to break up at lower altitudes. Therefore we changed the sentence to:

In our study we focus on the transition of the tracer composition in the vortex affected UTLS region up to […]

p. 3, lines 8-13. The coldest early Arctic winter since 1948? What records do you use that go back to 1948 (no reference given). In looking at the met statistics since 1979, I see there is the occasional DAY in December 2015 that breaks a record, but that's all. This statement needs to be backed up with a reference and needs to be more specific in what way it was the coldest. Since the vortex extends below 400K, it doesn't make sense to say that the chemical influence of the vortex is seen below 400K.

As shown in Matthias et al., 2016, the winter 2015/2016 was extraordinary cold in November / December based on ERA Interim (1979-2015) and NCEP/NCAR reanalysis from 1948-2016. They state in the first sentence of their abstract: "The Arctic polar vortex in the early winter 2015/2016 was the strongest and coldest of the last 68 years."

We therefore changed the sentence accordingly:

The early Arctic winter 2015/16 (November/December) was among the coldest winters in the lower stratosphere (LS) since 1948.

p. 3, line 14 and beyond. I thought the commonly used expression was 'stratospheric final warming' (SFW), not MFW. Is there a reference for the statement about MFWs being rare earlier than mid-March?

We have used the term 'major final warming' (MFW) from Manney and Lawrence, 2016, and added the reference.

p. 3, line 22. The QBO phase impact on vortex strength is only true for the northern hemisphere (Baldwin et al., 2001).

Baldwin et al (2001) state that they found an impact for the NH polar vortex and that the NH is more sensitive to breaking of planetary waves, due to larger wave amplitudes and disrupted circulation pattern by major warmings. Therefore we changed the manuscript:

Since the QBO affects the zonal wind direction in the tropical lower stratosphere (Niwano et al., 2003) its strength and phase is crucial for stratospheric transport processes (Baldwin et al., 2001) and westerly phases are related to strong and cold polar Arctic vortex.

p. 3, lines 23-24. Suggested rewrite: "Matthias et al. [2016] argue that the strong El Nino weakened the 2016 Arctic vortex, but this is still under debate." (Same meaning using half the words. When sentences are short and to the point, the paper becomes more interesting to read.)

Changed to the suggestion.

p. 4, line 14. Spell out what PVU stands for. And what are your PV units? Yours seem to be a factor of 10 smaller than in other papers. That's not a problem as long as you define your units.

Changed.

1 PVU = 10^{-6} m^2 s^{-1} K kg^{-1}

p. 4, lines 24-29: 'Resolution of 10 seconds' and 0.1 Hz are the same thing. Infrared is one word.

Changed.

p. 4, line 30 and beyond. Reword. "... operating between wavenumbers X and Y that measures CO, N2O, and CH4. The instrument uses a multi-pass White cell with a constant pressure of 30 hPa to minimize pressure broadening of the absorption lines. All measurements were integrated for 1.5 seconds." And later "In-flight calibration is performed against compressed ambient air standards that were calibrated against primary standards before and after the campaign. The primary standards are connected to..."

The manuscript is changed to:

The TRIHOP instrument (Schiller2008) is an infrared absorption laser spectrometer with three quantum cascade lasers (QCL) operating between wavenumbers 1269 cm^{-1} and 2184 cm^{-1} that measures CO, N_2O, and CH_4. The instrument uses a multi-pass White cell with a constant pressure of 30 hPa to minimize pressure broadening of the absorption lines. All measurements were integrated for 1.5 seconds. The three species are subsequently measured during a full cycle which finally leads to a time resolution of 7 seconds due to additional latency times when the channels are switched. In-flight calibration is performed against compressed ambient air standards that were calibrated against primary standards before and after the campaign. The primary standards are tracable to the World Meteorological Organisation Global Atmosphere Watch Central Calibration Laboratory (WMO GAW CCL) scale (X2007) for greenhouse gases. During POLSTRACC it was possible to achieve a (2 sigma) precision of CO, N_2O and CH_4 of 1.15, 1.84 and 9.46 ppb_V respectively.

p. 7, lines 17-20. A better way to describe this would be in terms of the spatial resolution you have after the averaging. So if you are smoothing over a 20 minute period, then based on the aircraft speed, what spatial resolution do you end up with?

Note that the SF_6 data are not smoothed over 20 minutes. A correlation fit to the CFC-12 is used for each individual SF_6 data point with an interval length of 20 minutes. Since the slope of the SF_6-CFC-12 correlation is robust and both species are affected by atmospheric variability in

the same way, this procedure reduces the noise without smoothing atmospheric variability. Since the data output of the GhOST-MS instrument is one minute, the spatial resolution of the SF_6 measurements is 15 km; see page 4, line 26.

p. 8, lines 6-7. Suggested "A Clams simulation with full stratospheric chemistry was integrated as described by Grooss et al (2016)." Why does it matter that it is typically run for 6 months? Delete this unless you explain why this matters.

Sentence changed to the suggestion.

A CLaMS simulation with full stratospheric chemistry was integrated as described by Grooss et al. (2014). The upper boundary is set to Θ = 900 K potential temperature, […]

p. 8, line 9. 'formulation' not formation.

Changed

p. 8, line 17. Why are the gravity wave flights excluded?

Since gravity waves introduce variability and turbulence which are not covered by the PV fields of the meteorological analysis the consistency of equivalent latitude and tracer observations would be destroyed.

p. 8, lines 22-23. Equivalent latitude is not just 'linked' to PV, it is derived from PV. This paper concerns itself with diabatic descent in the vortex, but you note that PV is conserved under adiabatic conditions (true), and clearly the polar vortex is not adiabatic. How to you justify the use of PV? (The answer lies in the time scales for radiative relaxation.)

Radiative time scales are long (on the order of weeks) since cooling rates in the vortex are typically 0.5 K/day. In contrast adiabatic excursions of the tropopause by Rossby Waves occur on timescales of days, which allows to separate tropospheric and stratospheric air masses by using equivalent latitude for large scale motions. Small scale non-conservative (i.e. non isentropic) processes lead to deviations between PV and comparing tracer and PV.

p. 8, line 27. Suggest "The transport pathways create an age spectrum or transit time distribution."

We changed the sentence to your suggestion.

p. 9, lines 3-8. Which lifetime did you use for SF6 in your model?

CLaMS does not use SF_6 to calculate mean age. Therefore we added a sentence to the CLaMS section:

Mean age in CLaMS is calculated from an inert model "clock-tracer" with linear increasing mixing ratio at the surface (Hall et al., 1994). The resulting mean ages are fully consistent with mean age calculated as the first moment of the CLaMS age spectrum (Ploeger and Birner, 2016).

Another rewrite:
"Models and observations both show a high bias of up to 1 year in the polar vortex."

Changed

At what altitude in the vortex does this apply to?

It is impossible to give a unique number here, since this critically depends on the descent and mixing in the vortices and may therefore vary from winter to winter. For 2015/2016 the effect might have occurred down to Θ = 550K or 600 K (see Fig. 12)

p. 9, line 9-10. This is the only correct usage of 'respectively' I saw. Still, it's not necessary because the 'respectively' is implied.

'Respectively' removed

p. 9, lines 21-31. Way too much detail on N2O. Instead of 'distinct background value' it's really that it has a near constant value throughout the troposphere that makes stratospheric influence identifiable. I would delete everything between "The tropospheric background value of N2O ... " and " ... Ko et al (2013)." You've already said the sources are at the surface, so delete the last 2 sentences of this paragraph and add: "N2O has a weak negative gradient above the tropopause that strengthens in winter due to increased diabatic descent."

We shortened the paragraph, but kept the information on the tropospheric mean value valid for our measurements as well as the information on the annual increase and the stratospheric sink, since both are relevant for our study.

Nitrous oxide (N_2O) has a lifetime of 123 years (Ko et al., 2013) and is released at the surface with no chemical sources in the atmosphere (Dils et al., 2006). As a results N_2O has a near constant tropospheric value of 329.3 ppb_v (winter 2015/2016 according to NOAA) that makes stratospheric influence identifiable (Müller et al., 2015). The mean tropospheric increase was found to be 0.78 ppb_v (Hartmann et al.2013)
The main sink reactions of N_2O are due to photolysis in the UV-band (190 nm 220 nm) and the reaction with $O(^1D)$ which only occurs within the upper stratosphere (Ko et al., 2013). Thus, N_2O above the tropopause shows a weak negative vertical gradient which maximizes during winter and spring due to the diabatic downwelling by the Brewer-Dobson circulation. […]

Paragraph beginning p. 9, line 32. A clearer way to say this: "Figures 4a and 4b show N2O values between 276-325 ppb_v were measured during Phase 1 and values below 200 ppb_v were measured during Phase 2 above 400 K. Figure 4c shows an overall decrease in N2O in the polar LS due to diabatic descent during winter, consistent with mean age changes (Fig. 3c)."

Sentence changed according to suggestion

Section 4.1.3 CO, first paragraph. This has way too much detail. Delete the 2nd sentence. 3rd sentence: "Due to the high variability of anthropogenic surface emissions, CO mixing ratios in the northern hemisphere vary from 70-200 ppbs [prinn] and the CO lifetime is on the order of weeks." 4th sentence ok. 5th & 6th sentences: "The main sink is oxidation by OH. The CO lifetime during polar night is a few months." Don't need reaction (1).

Changed according to suggestion

The phrase 'equilibrium value' is not right – this is not about chemical equilibria. I believe you are trying to describe a minimum stratosphere CO that comes from being

far above tropospheric CO sources but well below where high mesospheric CO can influence the lower stratosphere. Please delete usage of equilibrium CO (happens a lot on p. 13). Also, 'stratospheric background value' used on the next page and elsewhere is a vague expression. Can you say what you mean by this?

With equilibrium value we refer to the equilibrium between CO production from Methane oxidation (CH_4+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 $[CO]\_eq = k1/k2\ [CH_4]$ after solving $d[CO]/dt = k1[CH_4][OH] – k2[CO][OH] = 0$. Note that the value of CO_eq is independent from OH , since [OH] cancels out. Therefore in the long-term limit CO-production partly compensates CO-destruction leading to a CO value of 5-15 ppb_V depending on the integrated temperature history of the air masses, since the reaction rates depend on k_i. We termed the resulting CO value equilibrium value to emphasize, that we refer to a chemically driven equilibrium value. 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, Herman et al. 1999 and Marcy et al., 1999.
Therefore we kept the term CO-equilibrium value.

p. 10, line 15. All you need to say is "The reaction of CH4 with Cl is an insignificant source of CO in the lower stratosphere [Flocke]."

Changed to the suggestion

p. 11, line 6. Figure 7 is mentioned before Figure 6. Reorder.

In this case both figures are relevant, we changed the reference to Figures 6 and 7.

p. 11, line 10. The CO change during winter at 340 K is not so consistent with N2O. The CO changes small and are both positive and negative in this region. The results are mixed, not clear.
The sentence refered to Fig. 5 a)and 5 b). It is correct that the differences in Fig. 10c)show changes in both directions at lower isentropes

At lower isentropes the tropospheric variability starts to affect the overall distribution. Since CO has a lifetime on the order of months part of the tropospheric CO variability affects the LMS beyond the ExTL due to the lifetime of CO. As described in the shortened N_2O paragraph, N_2O does not show the same tropospheric variability as CO and thus the N_2O change between both phases (Fig. 4c) shows a much more homogeneous distribution at low isentropes.
We changed the sentence:

Hence the overall distribution of carbon monoxide in the UTLS during the individual phases (Fig. 5ab) seems to be consistent to N_2O and mean age obtained from SF_6 measurements, despite its much shorter lifetime compared to the other species.

p. 11, line 15. Change 'rise the hypothesis' to 'suggest'

Changed

p. 11, line 17. Since there are no CO surface observations used, you can't really say anything about the strength of tropospheric source emissions.

Based on our observations, it is correct, that we can't directly account for surface emissions. We can however take our observations at low equivalent latitudes. We also included MLS data in the later discussion.
We changed the statement

[...] from the tropical lower stratosphere over the course of winter without an increase of the upper tropospheric mixing ratios, which are affected by the surface emissions.

p. 12, lines 2-5. I don't agree with the statement that the 'lowest values' are found in the 'furthest regions from the troposphere'. The difference plots (3c and 4c) show a range of differences (even a few increases), so if you were to quantify this (rather than showing color blocks) it's not obvious that you statement would be true. Can you be more quantitative, or at least modify the words here?

Exactly for this reason we want to keep the overall tracer distributions as a function of latitude and potential temperature since the lowest values are not on display in panels 3c) and 4c), which show differences. They are shown in Figure 3 b and 4 b) at Θ = 380 K. We also sharpened the text, since the statement about absolute values and changes were not clearly separated.

We found a decrease of the long lived species SF_6 and N_2O with their lowest values far above the local troposphere in late winter […]

p. 12, line 4. 'contradicting' should be replaced, perhaps with 'unexpected'.

Changed

p. 12, lines 8-16. Here – and in general – there are too many sentences telling the reader what you are going to write about (or discuss later) rather than just writing about the subject. CO's use as a tracer has already been mentioned (Fig. 5) and the insignificant source form CH4 + CO has already been noted. Delete.

We followed the suggestion and shortened the paragraph:

In the following we will discuss this hypothesis and also other potential sources for the additional CO mixing ratios. To identify mixing processes across the tropopause CO-O_3 correlations have been widely used (Fischer et al., 2000; Zahn et al., 2000; Hoor et al., 2002; Pan et al., 2004; Müller et al., 2016). Since ozone is affected by chemical processes particularly in the vortex region we use N_2O as stratospheric tracer instead of ozone.

p. 13. Line 1-2. Use active voice as much as possible, e.g., 'tropospheric data have high N2O [how high?] and are accompanied by high CO while stratospheric data have N2O < 328 ppb_v'.

Changed

p. 13, line 7. N2O is 'longer-lived' not 'chemical inert'.

We changed the phrase to:

long-lived in the lower stratosphere

p. 13, line 13. Regarding the line showing meso CO on Fig. 6d, it should point to a value of 0 N2O because there is essentially no N2O in the mesosphere.

Changed

p. 13, lines 23-28. This result is not so mysterious and it would not seem so if this were discussed in terms of the species' gradients. The changes observed during winter come from 2 processes: descent and mixing. The change in each tracer depends on the tracer's vertical and horizontal gradients as well as the balance between vertical and horizontal motions. This is a more physically meaningful way to explain the observations.

This is exactly true, that descent and mixing act on the tracer and their gradients. The statement is not in contradiction to our analysis. Differing from the classical Plumb and Ko (1994) regime using long-lived tracers the use of two tracers of very different lifetime (like CO and N_2O) introduces some additional information to help to distinguish between mixing from the tropopause region and descent of aged air (but of course does not obey slope-equilibrium any more).
Indeed, the CO gradient on N_2O isopleths is used to unmask the different contributions of increased contribution of young air during our measurements. The quantification and relation to the age spectra later allow to further quantify these contributions.

p. 13, line 31. Try instead 'The region measured during both phases is 340-380K.'
Changed

p. 13 line 34-35. Why do you assume that the increased tropospheric fraction comes from the TTL? The POLSTRACC data didn't sample air south of 40N, so how can you be so certain of its origin? Isentropic mixing across the LMS tropopause (320-360K range) between the midlatitude UT and the polar LS would have the same effect, right? Without observations or a trajectory analysis, there isn't convincing evidence presented that the TTL is the source.

We checked this and added a plot which shows that the trajectories lead to the tropical tropopause region, particularly for the data above PV = 7 PVU. We used different tropopause definitions and identified the tropics and subtropics above Θ = 370 K as source regions for the trajectories.

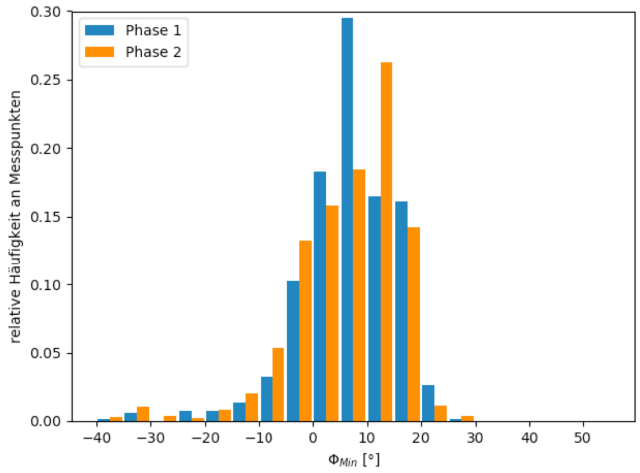

Fig. R1 (a): Probability density function against minimum trajectory latitude

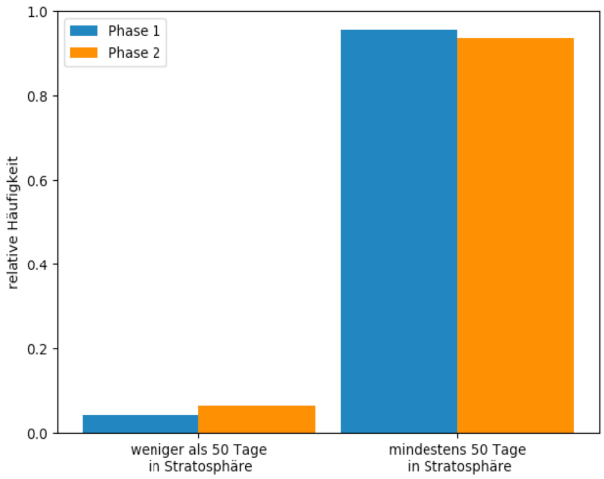

Fig. R1 (b): Probability density function for phase 1 (blue) and phase 2(orange). Left: less than 50 days in the stratosphere, right: at least 50 days in the stratosphere

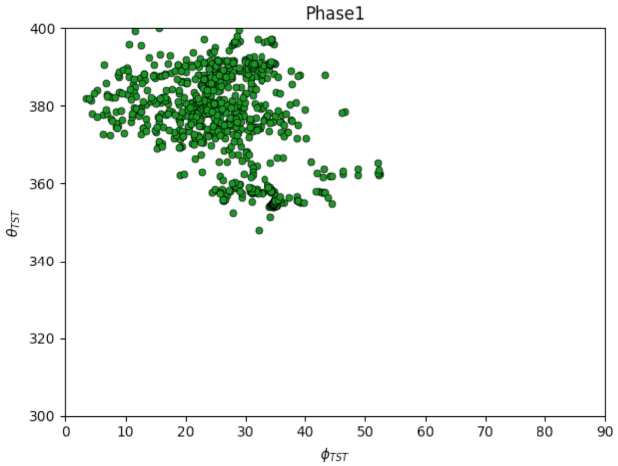

Fig. R1 (c): Potential temperature of TST against latitude of TST for phase 1

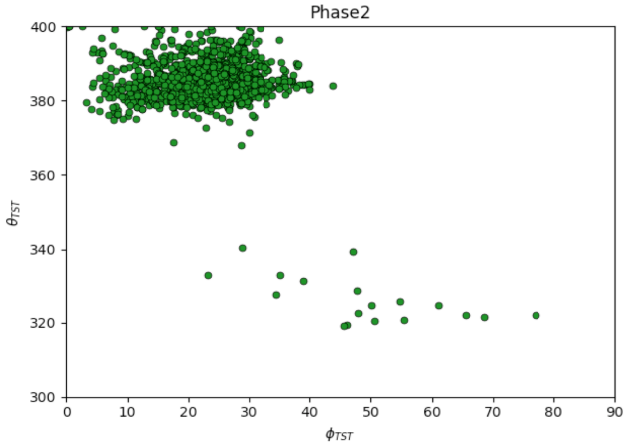

Fig. R1 (d): Potential temperature of TST against latitude of TST for phase 2.

The trajectory analysis indicates that the fraction of air masses from the TTL increases for phase 2. This is particularly true for those air masses originating above Θ = 370 K.
1. Panel (a): Relative distribution of minimum latitude of backward trajectories, which undergo a TST within the last 50 days. The contribution of trajectories originating from the TTL region is larger in phase 2 compared to phase 1.
2. Panel (b): The relative amount of trajectories which stayed less than 50 days in the stratosphere is in phase 2 greater than in phase 1.
3. Panel (c) and (d): Potential temperature and location of TST points of trajectories indicate that most trajectories in phase 2 have TST in the region of the TTL and the tropical tropopause, whereas the TST location in phase 1 extends to lower isentropes.

p. 16. Equations 2, 3, and 4 use mixing rations and fractions. I don't see any use of mass here. Why then is this described as mass balance?

The air mass density cancels out for the final equation 4 since the ratio of mixing ratios is calculated.

p. 16, line 15. Why are you citing 10-20 year old studies for the value of CO at the tropical tropopause? You could be using something far more precise and current by looking at MLS CO measurements (2004-present, as low as ~ 300 hPa). CO at the tropical tropopause has considerable seasonal variation due to seasonal biomass burning influence. MLS shows CO above 100 ppb_v in the TTL much higher than what is used in your study – see Huang et al. [ACP, 2016]. This assumed TTL CO value impacts your results. This section needs to be revised based on recent measurements of TTL

Since we use in-situ data we want to compare our measurements to in-situ measurements of other data sets, regions and years despite the disadvantage of the small data coverage during an aircraft based campaign. We appreciate the availability of satellite observations which provide a global view. For quantitative comparisons of aircraft observations with satellite data averaging kernels have to be applied, which is difficult in our case since most of the data were measured along horizontal flight tracks. In addition we are detecting rather small changes of CO on the order of 3 ppb_v.
As stated in Huang et al. (ACP 2016), the estimated single measurement precision for CO is 19 ppb_v and the systematic uncertainty is on the order of +/- 30 % for CO (MLS V4.2) based on Livesey et al., (2015).
Zonal and seasonal means as shown in Huang et al., (2016) however provide the possibility to compare our data qualitatively to the observations by MLS.
As can be seen in Huang et al., (2016) Fig. 12 and 13 CO climatologies show maximum values of 110 ppb_v at 215 hPa and 80 ppb_v at 100 hPa with higher peak values in certain years or regions. To our knowledge there is no airborne data set, which shows such high mean CO values in the tropics besides individual plume encounters.
Therefore we performed our calculations for CO values of 60, 70 and 80 ppb_v according to the climatological regional profiles (Fig.14 in Huang et al., (2016)) at 100 hPa. The estimates of our calculated fractions are changing by 2.2% and are shown below. Since the denominator of our equation (3) depends linearly on the difference between CO (tropical) and the observed stratospheric background an increase of tropical CO from 60 ppb_v to 80 ppb_v reduces our fraction by 32 %.
We accounted for this by indicating the range for different estimates of the CO-tropical and modified the text.

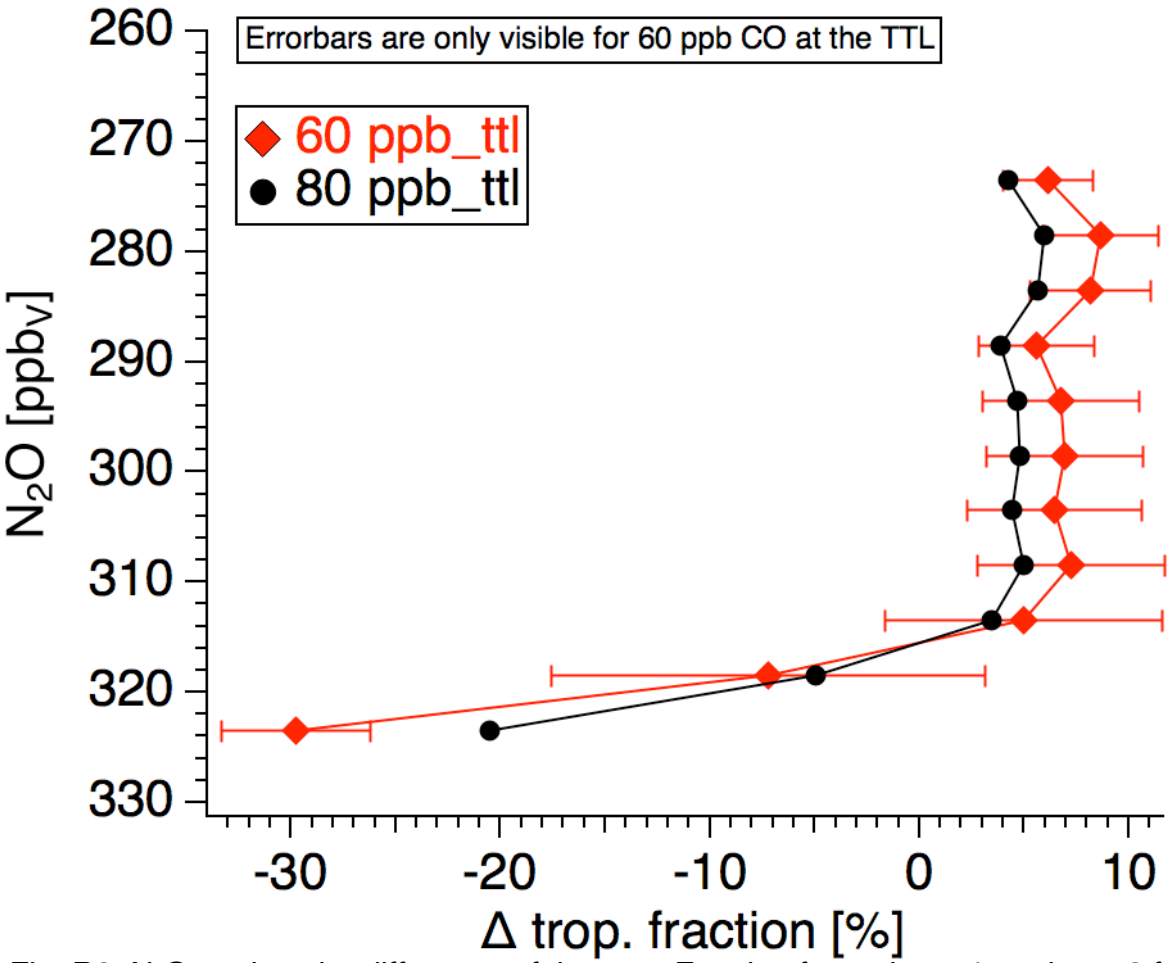

Fig. R2: N_2O against the difference of the trop. Fraction from phase 1 to phase 2 for different tropical CO entry values in the TTL

The CO increase over the course of winter corresponds to an increase by f_trop of (6.8 +/- 3.7)% between 313 ppb_V and 273 ppb_V N_2O by assuming 60 ppb_V of CO at the tropical tropopause as provided by in-situ aircraft data from Herman (1999) and Marcy et al. (2007). Using CO_trop = 80 ppb_V as indicated by MLS at 100 hPa one obtains 32% lower values for f_trop, which is still a significant increase of tropospheric air masses

p. 16, lines 28-29. Should be 'longer transit times' and 'mixed with young tropospheric air with shorter transit times ... '

Changed to the suggestion

p. 17, line 17. Try 'Figure 10b shows there is ....' 0.3% is a very small change. What is the uncertainty on this number? Is 0.3% statistically different from zero?

Changed to the suggestion.

We thank the reviewer for this point indicating a typo. The relevant unit in Fig. 10 is '% per month' and was not given in the text. The correct phrase now gives '…0.3 % per month…' as indicated by the vertical axis label in Fig.10. Note, that these 0.3 % per month are absolute values. The age spectra in Fig. 10 a) indicate the maximum difference of 0.3 % per month at e.g. 3.5 years transit time (panel (b)). The absolute value in panel (b) for phase 1 is 0.41 % per month and for phase 2 is 0.71 % per month. Therefore the relative increase in the age spectra

is 76 % which is significant. The relative increase of the young fraction (less than six month) is 19 %.

p. 18, line 2. Try '... the observed CO increase, indicating increased mixing with air..'

Changed to the suggestion

p. 18, line 7. This is really the lowermost not the lower stratosphere you are talking about.

Changed to:

[…] the overall ageing in the lower and lowermost stratosphere over […]

p. 18, lines 11-12. Try 'To further investigate the relationship of young versus aged air, we calculate the accumulated fraction ...'

Changed to the suggestion

p. 18. The paragraph beginning on line 15 should be part of the previous one.

Changed

p. 21, line 1-3. You could make a good point here that mean age is an incomplete or oversimplified way to characterize air. The age spectrum shows there changes in the contributions from older and younger age masses that determine the change in mean age.

We added a sentence to the manuscript:

Therefore our results demonstrate, that the mean age is an incomplete descriptor when referring to chemical properties of air masses involving different chemical life times of species. Since the mean age is just a single number it might be insensitive to changes of the processes and time scales contributing to the mean, but affecting chemical properties and impact of the air parcel. Therefore it is important to account for the full spectral shape when referring to chemical properties of an air mass rather than only the mean age.

p. 21, lines 5-10. You can cite Rinsland 1999 and Rosenfield 1994 that mesospheric CO just doesn't go this low into the stratosphere, and then you are done with this issue. The CH4+CL sources of CO has already been labeled insignificant. There is no need to talk about these insignificant sources again. This means that all of Section 6 (Discussion) before about p. 23 can be deleted.

We shortened the discussion. We kept the individual points in the manuscript, since the observed change of CO relative to N_2O is small and we have to exclude potential sources of CO. Further the reaction CH_4+Cl is in principle able to produce the respective amount of CO according to CLaMS. The additional CO is however reacting via OH leading to a zero net increase. We only showed the net effect in the manuscript to keep it short.

Carbon monoxide is produced in the mesosphere due to the photo-dissociation of carbon dioxide. Therefore the composition of mesospheric air masses is clearly distinct from air mass composition of the stratosphere. Rinsland et al. (e.g. 1999) found increased CO mixing ratios up to 90 ppb_V at altitudes around 25 km or Θ = 630K - 670 K and Engel et al. (2006b) found CO values of 600 ppb_V at an altitude of 32 km. Both studies show very low N_2O mixing ratios (< 50

ppb$_v$). Although the authors found layers of mesospheric air descending down to 22 km, this is not evident for the Arctic winter 2015/16 and lowest N_2O mixing ratios are found to be in the order of 200 ppb$_v$.
This is reflected in the MLS observations that determine the CLaMS upper boundary at Θ = 900 K potential temperature (Fig 12.). The simulation indicates the expected downward transport of mesospheric influenced air, but down to Θ = 600 K at the end of March 2016 in agreement with our observations which minimize at the highest flight levels and equivalent latitudes.
Furthermore, an additional influence of descended mesospheric air into the lower stratosphere would lead to mixing lines very strongly differing from the observed relationship (see Fig. 6), which is not observed in agreement with the CLaMS N_2O-CO scatter plot (Fig. 9).
In general, another important source of carbon monoxide in the atmosphere is the reaction of methane with reactive chlorine, which is not significant in the lower stratosphere (Flocke et al., 1999). However, air masses enriched in reactive chlorine could have been transported downwards, providing potentially reactants for the chemical production of CO. Therefore, we simulated the CO yield from the reactions of CH_4 with chlorine, OH and O(^{1}D) using CLaMS simulations in the box model mode. A large number of air parcel backward trajectories starting on 15 March from locations within the vortex core (equivalent latitude > 65 °N; potential temperature between Θ = 350 K and Θ = 500 K) ending on 15 January and chemical composition changes were calculated using the CLaMS chemistry module running forward in time for a subset of the trajectories with equivalent latitudes > 50 °N on 15 January (21480 trajectories).
Figure 13 shows the statistical evaluation of the net CO change due to chemistry over the period as function of potential temperature on 15 March. The blue line represents the statistical mean and the dashed lines the 1-sigma standard deviation. The mean overall change is even negative over the entire profile, which is due to the oxidation of the produced CO by the reaction with OH. Therefore we conclude that the observed increase of CO in phase 2 is not due to the additional chemical source reaction.
Additionally the age spectrum calculations of the CLaMS model provide mass fractions of air masses regarding their stratospheric residence time. As is evident from Fig. 9 there is a significant increase of air masses younger than six months at typical mean ages for lower stratospheric air masses and mesospheric influence on the basis of our analysis is highly unlikely.

p. 23, 10. Here's a suggestion for Fig. 14. Since you say that the SH results should not be compared to the NH, then don't show them. Figure 14 would be more readable if you just showed the regions of interest. Try making a 4-panel figure (Jan and March), 0-90N or 30-90N only.

We changed the Figure according to suggestion

p. 25, line 5. What are the significant figures here? Is 0.29 really different from 0.27, or are they both 0.3?

The increase by 0.29 years of SF_6 is deduced from the measured SF_6 distribution in Fig. 3c) and the increase by 0.27 years is calculated from CLaMS mean ages along the flight track in Fig. 10 a) which is now changed in the manuscript. (see also reply to reviewer 1):
Note that the increase of 0.29 years is only valid for the overlapping distribution of phase 1 and phase 2 and also the CLaMS simulations indicate an increase of the mean age of the same magnitude, which is consistent.

The Figure below further illustrates the change of the mean age distribution towards higher mean ages based on the SF_6 distributions for the two phases. The increase of the mean age over all observed data of the distribution is 0.79 years.

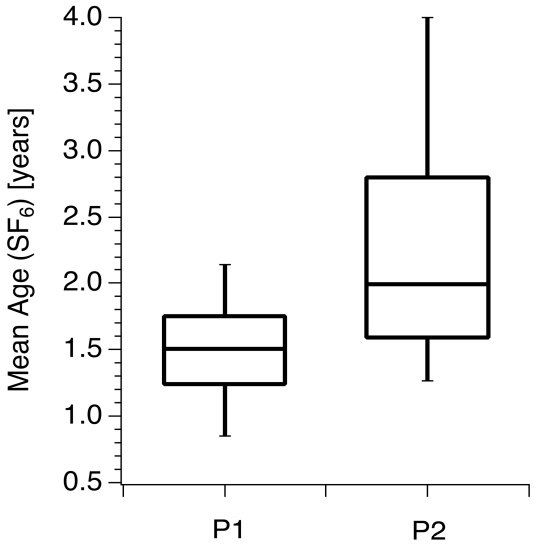

Fig. R3: Box-Whisker plot of Mean Age (SF_6) for phase 1 and phase 2

p. 25, line 18. The conclusions should be about the lowermost, not lower stratosphere as all the results are below 380K

We changed our manuscript accordingly.

References:

M. P. Baldwin, L. J. Gray, T. J. Dunkerton, K. Hamilton, P. H. Haynes, W. J. Randel, J. R. Holton, M. J. Alexander, I. Hirota, T. Horinouchi, D. B. A. Jones, J. S. Kinnersley, C. Marquardt, K. Sato, and M. Takahashi, "The quasi-biennial oscillation," Rev. Geophys., vol. 39, no. 2, pp. 179–229, May 2001.

Mixing and ageing in the polar lower stratosphere in winter 2015/2016

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Abstract. We present data from winter 2015/2016, which were measured during the POLSTRACC (The Polar Stratosphere in a Changing Climate) aircraft campaign between December 2015 and March ~~2016.~~ 2016 in the Arctic upper troposphere and lower stratosphere (UTLS). The focus of this work is on the role of transport and mixing between aged and potentially chemically processed air masses from the stratosphere with mid and low latitude air mass fractions with small transit times originating at the tropical lower stratosphere. By combining measurements of CO, N_2O and SF_6 we estimate the evolution of the relative contributions of transport and mixing to the UTLS composition over the course of the winter.

We find an increasing influence of aged stratospheric air partly from the vortex as indicated by decreasing N_2O and SF_6 values over the course of winter in the extratropical lower and lowermost stratosphere between $\Theta = 360$ K and $\Theta = 410$ K over the North Atlantic and the European Arctic. Surprisingly we also found a mean increase of CO by (3.00 ± 1.64) ppb_V from January to March relative to N_2O in the lower stratosphere. We show that this increase of CO is consistent with an increased mixing of tropospheric air as part of the fast transport mechanism in the lower stratosphere surf zone. The analysed air masses were partly affected by air masses which originated at the tropical tropopause and were quasi-horizontally mixed into higher latitudes.

This increase of the tropospheric air fraction partly compensates for ageing of the UTLS due to the diabatic descent of air masses from the vortex by horizontally mixed, tropospheric influenced air masses. This is consistent with simulated age spectra from the Chemical Lagrangian Model of the Stratosphere (CLaMS), which show a respective fractional increase of tropospheric air with short transit times lower than six months and a simultaneous increase of aged air from ~~deep~~ upper stratospheric and vortex regions with transit times larger than two years.

We thus conclude that the lowermost stratosphere in winter 2015/16 was affected by aged air from the ~~deep~~ upper stratosphere and vortex region. These air masses were significantly affected by increased mixing from the lower latitudes, which led to a simultaneous increase of the fraction of young air in the Arctic lowermost stratosphere ~~over the course of winter.~~ by 6 % from January to March 2016.

~~The upper troposphere~~

1 Introduction

Uncertainties in the description of mixing introduce large uncertainties to quantitative estimates of radiative forcing which are on the order of 0.5 W m^{-2} (Riese et al., 2012). Therefore it is important to quantify the contribution of the dynamical processes which act on the distribution of tracers. The Arctic UTLS during winter is affected by diabatic descent from the stratosphere and quasi horizontal mixing by the shallow branch of the Brewer-Dobson circulation, which connects the tropical tropopause region with the high Arctic (e.g. Rosenlof et al., 1997; Birner and Bönisch, 2011). We present data from winter 2015/2016, which were measured during the POLSTRACC (The Polar Stratosphere in a Changing Climate) aircraft campaign between December 2015 and March 2016 in the Arctic upper troposphere and lower stratosphere (UTLS) (see Fig. 1)~~region~~.

The focus of this work is on the role of transport and mixing between aged and potentially chemically processed air masses from the stratosphere with mid and low latitude air mass fractions with small transit times originating at the tropical lower stratosphere. By combining measurements of CO, N_2O and SF_6 we investigate the evolution of the relative contributions of transport and mixing to the UTLS composition over the course of the winter. During winter the UTLS region (Fig. 1) at high latitudes ~~during winter~~ is strongly affected by the evolution of the polar vortex. Diabatic descent in the polar stratosphere, which is ~~most pronounced~~ strongest inside the polar vortex ~~adds to the diabatic downwelling~~ results as part of the Brewer-Dobson circulation (Brewer, 1949; Dobson, 1956) in mid and high latitudes as response to the breaking of planetary and gravity waves (Haynes et al., 1991; Plumb, 2002; Butchart, 2014) in the upper stratosphere and mesosphere~~, respectively~~. This downwelling leads to an increasing contribution of ~~deep~~ stratospheric air masses ~~in~~ from the overworld (defined as the region, where isentropes are entirely located in the stratosphere (Hoskins, 1991)). Over the ~~UTLS over the~~ course of winter ~~where~~ they contribute to the composition of the lower overworld ($\Theta < 420$ K), where our measurements took place, and the lowermost stratosphere (LMS) ~~(Rosenfield et al., 1994; Holton et al., 1995)~~(Rosenfield et al., 1994) (defined as the region bounded by the 380 K isentrope and the extratropical tropopause (Holton et al., 1995)).

~~Chemically these air masses~~ 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 (Solomon, 1999). Since the air inside the polar vortex is largely isolated and exhibits a strong diabatic descent due to radiative cooling and the wave-driven Brewer-Dobson circulation this leads to an increased fraction of air masses with a high mean age of air in the UTLS of high latitudes (e.g. Engel et al., 2002; Ploeger et al., 2015).

The mean age of air is defined as the first moment of the transit time distribution (or the so-called age spectrum) (Hall and Plumb, 1994; Waugh et al., 1997). Mean age can be determined from the observation of long-lived tracers, which ideally have no sources or sinks in the stratosphere and of which the temporal evolution of the mixing ratio at the tropical tropopause is well known (Waugh et al., 1997). Notably, the mean age is a bad descriptor for the full age spectrum, which is highly skewed (e.g. Hall and Plumb, 1994) and sometimes even multimodal (Andrews et al., 1999; Boenisch et al., 2009). For the estimate of the potential chemical impact of species particularly with lifetimes on the order of weeks to only a few months the mean age is insufficient and the full spectrum is needed (Schoeberl et al., 2000), which is however only available under very idealized conditions (Schoeberl et al., 2005; Ehhalt et al., 2007).

Observations of SF_6, N_2O and CO_2 from the ER-2 aircraft show that the mean age at northern high latitudes at an altitude of 20 km is on the order of 4-6 years (Andrews et al., 2001; Engel et al., 2002). Satellite observations of SF_6 confirm this and show further a strong interannual variability of the mean age in northern high latitudes (Stiller et al., 2008, 2012; Haenel et al., 2015). The observations also indicate a potential transport of mesospheric air to lower altitudes (Engel et al., 2006b; Ray et al., 2017), which however strongly depends on the strength and persistence of the Arctic polar vortex during the individual winters. In addition to diabatic descent inside and outside the polar vortex quasi-isentropic mixing from lower latitudes leads to a contribution of relatively young air to the UTLS. As a result a seasonal cycle of the chemical composition of the UTLS up to $\Theta = 430$ K establishes ~~with a more~~ a relatively tropospheric character during northern summer / autumn and a more stratospheric characteristic in late winter/spring (Hegglin and Shepherd, 2007). The chemical composition and age structure of the extratropical UTLS (ExUTLS) are affected by the competing diabatic downwelling of aged air and rapid quasi isentropic mixing down to the tropopause (Hoor et al., 2005; Engel et al., 2006a; Boenisch et al., 2009; Garny et al., 2014). The region between $\Theta = 380$ K and the bottom of the ~~subtropical~~ tropical pipe around $\Theta = 450$ K (Palazzi et al., 2011) is a key region for the transition between these transport regimes. The $\Theta = 380$ K isentrope coincides with the tropical tropopause and is therefore directly affected by diabatic vertical transport of tropospheric air through the tropical transition layer (TTL) (Fueglistaler et al., 2009) into the stratosphere. ~~These air masses~~Above $\Theta = 380K$ these air masses, which ascended through the TTL are rapidly mixed quasi horizontally by breaking planetary waves with air from high latitudes in addition to the shallow branch of the Brewer-Dobson circulation (Birner and Bönisch, 2011; Abalos et al., 2013). This rapid transport modifies the abundance of particularly water vapour and ozone in this region~~(Rosenlof et al., 1997; Randel et al., 2006; Hegglin and Shepherd, 2007; Pan et al., 2007) .To estimate the radiative effects of these species with large gradients at the tropopause, the details of mixing are essential (Forster and Shine, 1997, 2002; Riese et al., 2012). Uncertainties arising only from uncertainties in mixing may lead to significant uncertainties of the radiative forcing, which are on the order of 0.5 W m^{-2} (Riese et al., 2012)~~, which have seasonally varying isentropic gradients (Rosenlof et al., 1997; Randel et al., 2006; Hegglin and Shepherd, 2007; Pan et al., 2007; Ploeger et al., 2013) .

In our study we focus on the transition of the tracer composition in the ~~subvortex~~ vortex affected UTLS region up to $\Theta = 410$K during winter 2015/2016. We will quantify the effects of quasi-isentropic mixing from the tropics and diabatic downwelling and its effect on the chemical composition as well as the evolution of the age spectrum and the mean age in this region.

2 Meteorological conditions during winter 2015/16

The early Arctic winter 2015/16 ~~was the coldest winter~~ (November/December) was among the coldest winters in the lower stratosphere (LS) since 1948. These extreme cold conditions ~~could establish~~ existed due to a strong and cold Arctic polar vortex which developed in November 2015 due to very low planetary wave activity in the stratosphere (Matthias et al., 2016). From late December 2015 to early February 2016 the temperatures at $\Theta = 490$ K decreased below 189 K. Therefore strong dehydration and denitrification ~~was~~ were seen in low H_2O and HNO_3 volume mixing ratios, which finally led to a strong chlorine activation in early winter. Using MLS data the chemical influence of the vortex could be observed ~~to~~ on isentropes below

$\Theta = 400$ K (Manney and Lawrence, 2016).

The major final warming (MFW) (Manney and Lawrence, 2016) occurred on 5 March 2016 which led to a split of the vortex one week later. This early final warming was unusual, as only five other MFWs since 1958 appeared before middle of March. Due to this early warming air masses in the polar lower stratosphere were mixed with non-vortex air and prevented chemical ozone depletion reaching record low values during winter 2015/16 (Manney and Lawrence, 2016, and references therein).

The winter 2015/16 was characterised by an unprecedented anomaly of the quasi biannual oscillation (QBO) with a ~~westward~~ westerly jet formed within the ~~eastward~~ easterly phase in the lower stratosphere (Newman et al., 2016; Osprey et al., 2016). Since the QBO affects the zonal wind direction in the tropical lower stratosphere (Niwano et al., 2003) its strength and phase is crucial for stratospheric transport processes (Baldwin et al., 2001) and westerly phases are related to a strong and cold polar ~~vortices~~ Arctic vortex (Holton and Tan, 1980).

Further the winter 2015/16 was also affected by a strong warm phase of the El-Niño Southern Oscillation (ENSO) ~~(McPhaden et al., 2015) . A direct influence on the polar vortex is still under debate, but according to Matthias et al. (2016) this~~ (Chen et al., 2016; L'Heureux et al., ~~. Matthias et al. (2016) argue that the~~ strong El-Niño ~~is suggested to account for a weakening of the polar vortex~~weakened the 2016 Arctic vortex, while Palmeiro et al. (2017) found a connection of this ENSO event to the strong polar vortex and the easterly MFW.

3 Project overview and measurements

This work will address the evolution of composition, age structure and the influence of transport and mixing of air masses in the lower stratosphere. The composition of air masses inside the LS, which is affected by diabatic descent of upper stratospheric air masses, irreversibly mixed with younger air from the TTL, is analysed by combining measurements of in-situ data with model calculations of the Chemical Lagrangian Model of the Stratosphere (CLaMS) (McKenna et al., 2002; Grooß et al., 2014; Ploeger et al., 2015).

3.1 The POLSTRACC campaign 2015/16

The data presented in this study were obtained during the POLSTRACC (Polar Stratosphere in a Changing Climate) mission, which was part of the combined PGS (POLSTRACC/GW-LCYCLE/SALSA) framework. The main objectives of the POLSTRACC mission were the investigation of structure, composition and dynamics of the Arctic LMS and processes involving chemical ozone depletion and polar stratospheric clouds in the Arctic winter UTLS. In total 17 scientific flights were performed from December 2015 until end of March 2016 on board the new German research aircraft HALO (High Altitude Long Range) from Oberpfaffenhofen, Germany (48.05 °N, 11.16 °E) and Kiruna, Sweden (67.49 °N, 20.19 °E) covering the region from 25 °N to 87 °N and 24 °E to 80 °W (Fig. 2). Typical flight altitudes ranged from 10 km asl[1] to 14.5 km asl corresponding to potential temperatures in the stratosphere from $\Theta = 320$ K up to $\Theta = 410$ K. The total flight time was about 157 hours, of which 19 hours were spent in December 2015, 62 hours were spent from January to February and 76 from February to

[1] above sea level

March, respectively. For this study we focus on Arctic measurements starting from Kiruna, which took place during two campaign phases, representing flights from 12. January 2016 to 02. February 2016 (phase 1) and from 26. February 2016 to ~~the~~ 18. March 2016 (phase 2). For ~~the aim of~~ this work we use approximately 50 hours of measurements of those flights which were conducted to probe air masses ~~predominantly~~ above the extratropical transition region (ExTL) and underneath the polar vortex above PV = 7 PVU(1 PVU = 10^{-6} m^2 s^{-1} K kg^{-1}).

The research aircraft HALO is a modified business jet type Gulfstream G-550. It has a maximum range of 12500 km with a maximum altitude of 15.5 km and can carry ~~about~~ up to 3 tons of scientific payload. The payload was a combination of different remote sensing (e.g. WALES lidar (Wirth et al., 2009; Fix et al., 2016), Väisälä RD 49 dropsondes and GLORIA limb sounder (Friedl-Vallon et al., 2014; Kaufmann et al., 2015)) and in-situ instruments ~~of~~ that measured trace gases with different lifetimes, sources and sinks.

3.2 In-situ trace gas measurements

In this study we analyse measurements of N_2O ~~,~~ and CO, which were measured with the TRIHOP instrument (Müller et al., 2015) and SF_6 by the GhOST-MS instrument (Sala et al., 2014). For our analysis the data are synchronised to a common time resolution of ~~10 seconds or~~ 0.1 Hz~~respectively~~, corresponding to a horizontal resolution of 2.5 km at typical HALO flight speeds. GhOST data is available with a resolution of 60 seconds at an integration time of one second which leads to a horizontal resolution of 15 km.

3.2.1 The TRIHOP instrument

The TRIHOP instrument (Schiller et al., 2008) is an ~~infra red~~ infrared absorption laser spectrometer with three quantum cascade lasers (QCL) operating between wavenumbers 1269 cm^{-1} and 2184 cm^{-1} ~~. It was set up to measure~~ that measures CO, N_2O and CH_4~~during the POLSTRACC campaign. To quantify mixing ratios in the order of ppb$_v$ the~~ . The instrument uses a ~~multi pass White-cell which is pressure regulated at a~~ multi-pass White cell with a constant pressure of 30 ~~mbar~~ hPa to minimize pressure broadening of the absorption lines. The ~~measurements were performed~~ three species are subsequently measured with an integration time of 1.5 ~~seconds per species . The three species are subsequently measured~~ s per species during a full cycle which finally leads to a time resolution of 7 seconds due to additional latency times when the channels are switched. ~~The instrument is regularly calibrated in-flight against compressed standards of ambient air which are in turn calibrated prior~~ In flight calibration is performed against compressed ambient air standards that were calibrated against primary standards before and after the campaign~~against primary standards , connected~~ . The primary standards are traceable to the World Meteorological Organisation Global Atmosphere Watch Central Calibration Laboratory (WMO GAW CCL) scale (X2007) for greenhouse gases. During POLSTRACC it was possible to achieve a (2σ) precision of CO, N_2O and CH_4 of 1.15, 1.84 and 9.46 ppb$_v$ respectively.

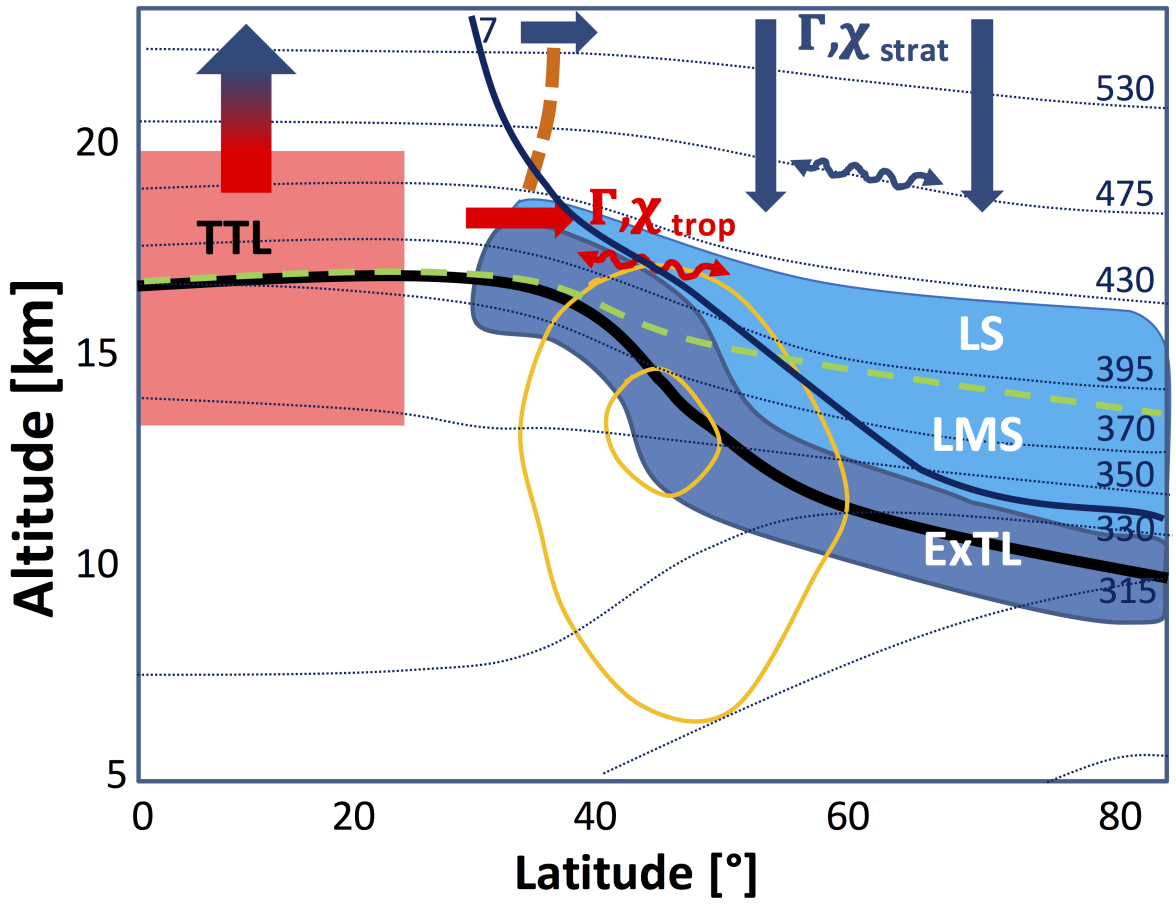

Figure 1. Cross section of the northern hemispheric UTLS (Upper Troposphere / Lower Stratosphere) region, adapted from Riese et al. (2014) and Müller et al. (2016). The thermal tropopause is denoted by the thick black solid line. The measurement region is depicted as blue box subdivided into the extratropical tropopause layer (ExTL), the lowermost stratosphere (LMS) and the lower stratosphere (LS). LMS and LS are separated by the 380 K isentrope (green dashed line). Transport pathways of air masses are denoted by coloured, thick arrows from the tropical tropopause layer (TTL) (red) and the polar Arctic upper stratosphere (blue) with respective mean age Γ and trace gas volume mixing ratio χ. Quasi horizontal mixing is represented by wavy double side arrows, indicating no net mass transport of air masses. Dotted lines are isentropes in K, the solid dark blue line indicates the 7 PVU contour, which is used to separate the regime of the ExTL from the LMS and LS (for details see text). Thin orange contour lines depict the zonal view of the jet stream.

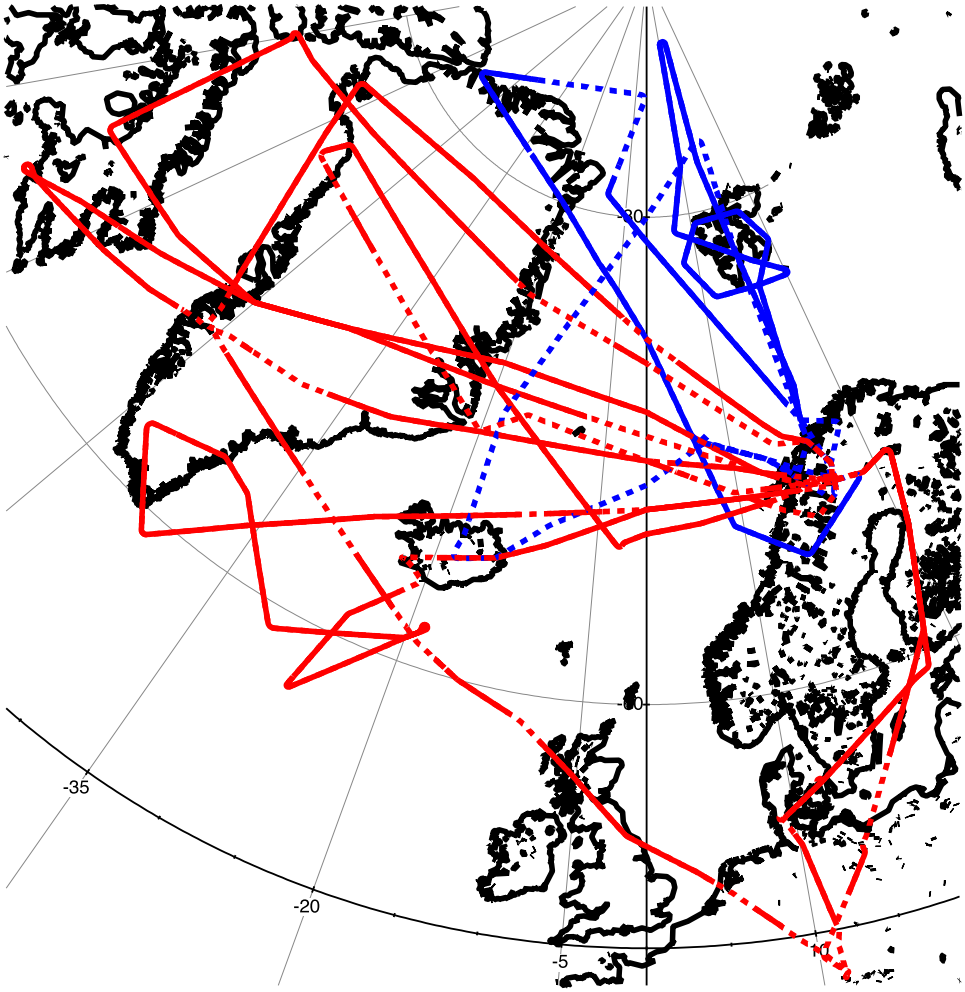

Figure 2. Flight tracks during the POLSTRACC campaign. Blue colours indicate flights during phase 1 (12. January-02. February), red colours indicate flights during phase 2 (26. February-18. March). ~~Only flights are shown, which~~ Flights that were used for the analysis are shown as solid lines. Dotted lines denote flight legs with PV < 7 PVU. For details see Sect. 3.1.

3.2.2 GhOST-MS in-situ measurements

The GHOST-MS instrument is a two channel gas chromatograph for airborne measurements of trace gases. One channel uses a mass spectrometer (Agilent MSD 5975) for the detection of atmospheric trace gases at a time resolution of four minutes. This channel uses negative ion chemical ionization as described in Sala et al. (2014) to measure brominated hydrocarbons. The other channel measures SF_6 and CFC-12 using an ECD (Electron capture detector) with a time resolution of one minute. For the POLSTRACC campaign the precision for SF_6 was 0.6% and the precision for CFC-12 was 0.2%.

Mean age of air is inferred from SF_6 measurements (Engel et al., 2009). Due to its much higher atmospheric mixing ratio, the precision of CFC-12 measurements is better than that of SF_6 measurements. Prior to calculating mean age, the SF_6 time series has therefore been smoothed using the CFC-12, by applying a local (ten minutes of data before and after the time of measurement) fit between CFC-12 and SF_6. This procedure removes parts of the instrumental scatter but retains the local information, keeps particularly the atmospheric variability (unlike averaging) and does not introduce any offset to the mean age values. Mean age derived in this way has an overall precision of better than 0.3 years and an estimated accuracy of 0.6 years, as explained in Engel et al. (2006a). Both SF_6 and CFC-12 are reported on the SIO-2005 scale.

3.3 The Chemical Lagrangian Model of the Stratosphere (CLaMS)

The analysis of trace gas measurements is complemented by simulations with the Chemical Lagrangian Model of the Stratosphere CLaMS (McKenna et al., 2002; Konopka et al., 2004). CLaMS is a Lagrangian chemistry transport model, based on forward trajectory calculations and a parameterization of small-scale atmospheric mixing which depends on the deformation rate of the large-scale flow. The model simulation is driven with meteorological data (e.g., horizontal wind fields) from European Center of Medium Range Weather Forecasts (ECMWF) ERA-Interim reanalysis (Dee et al., 2011) and covers the period 1979-2017. The model uses an isentropic vertical coordinate throughout the stratosphere and the vertical velocity is deduced from the reanalysis total diabatic heating rate. Further details about the model set-up and the included chemical reactions (relevant species here are CO and N_2O) are given in Pommrich et al. (2014). This long-term CLaMS simulation has been shown to reliably represent transport processes in the lower stratosphere for the relevant trace gas species CO and N_2O (Pommrich et al., 2014) as well as for mean age of air (Ploeger et al., 2015).

Recently, a method to calculate the age of air spectrum has been implemented in CLaMS (Ploeger and Birner, 2016), which will be used in the following analysis. The age spectrum is the transit time distribution of air masses for transport from a control surface (usually ~~take~~ taken as the tropical tropopause or the Earth's surface) to a given location in the stratosphere (e.g. Hall and Plumb, 1994; Waugh, 2002) and can be related to the ~~Greens~~ Green's function of the transport equation. The calculation method in CLaMS is based on inert tracer pulses, with different tracers released every other month at the surface in the tropics. This method allows ~~calculating~~ the calculation of time dependent age spectra for the non-stationary atmospheric flow at any location and time in the model domain (see Ploeger and Birner (2016) for further details). Mean age in CLaMS is calculated from an inert model "clock-tracer" with linear increasing mixing ratio at the surface (Hall and Plumb, 1994). The resulting mean ages are fully consistent with mean age calculated as the first moment of the CLaMS age spectrum (Ploeger and Birner, 2016).

~~For the aim of this work also a~~ A CLaMS simulation with full stratospheric chemistry was ~~performed by CLaMS with the setup~~ integrated as described by Grooß et al. (2014). ~~This setup is typically used for periods up to six months.~~ The upper boundary is set to Θ =900 K potential temperature, where tracers like O_3, N_2O and CO are constrained by MLS satellite observations. Due to its Lagrangian ~~formation~~formulation, a box-trajectory model setup is also possible in which the identical chemistry scheme is used along single air mass trajectories. This setup is also used here to diagnose chemical pathways and chemical conversion rates. This ~~boxmodel~~ box model setup is also used here to estimate CO production and loss rates.

4 Results

As shown in Hoor et al. (2010) rapid and frequent mixing with tropospheric air mainly affects the region of PV < 7 PVU. To exclude mixing with air masses of recent tropospheric origin or from the exTL (extratropical tropopause layer) we only selected data above this level of potential vorticity. Therefore the composition of analysed data is mainly affected by isentropically, irreversibly mixed air mass signatures originating out of the tropics and diabatically descended air masses from the upper stratosphere in the polar region. In this analysis we further excluded flights, which were dedicated to the observation of gravity waves.

4.1 Tracer distributions and mean age

Figures 3, 4 and 5 show tracer distributions ~~respective~~ as a function of equivalent latitude and potential temperature θ (Strahan et al., 1999; Hoor et al., 2004; Hegglin et al., 2006). Equivalent latitude is directly linked to the potential vorticity, which is conserved under adiabatic processes (Holton, 2004). Therefore, these coordinates are suitable to account for reversible adiabatic tracer transport.

4.1.1 Age of air

An air parcel in the stratosphere is a mixture of fractions of air with different histories, transport pathways and individual transit times. The ~~several transport pathways constitute to~~ transport pathways create an age spectrum or transit time distribution~~, respectively~~. The age spectrum can be obtained by calculation of the Green's function of the tracer continuity equation for a conserved and passive species (Hall and Plumb, 1994).

The mean age is defined as the first moment of the transit time distribution. To determine the mean age from ~~measurements,~~ long-lived ~~tracers can be used, which~~ tracer measurements, the tracer must have a well known source distribution at the tropical tropopause and a well defined vertical gradient in the stratosphere (Hall and Waugh, 1997). Since SF_6 is a long-lived inert trace gas with a well known increase of its mean surface mixing ratio, it is ~~a commonly used species~~ commonly used for calculations of mean age (Boenisch et al., 2009). The sink of SF_6 is in the mesosphere, where it is destroyed by shortwave UV radiation. The lifetime of SF_6 is assumed to be 3200 years, but recent studies indicate a significantly shorter lifetime of about 850 years (Ray et al., 2017). This implies that mean age derived from SF_6 may be too old. ~~Especially for polar vortex air, this has been~~

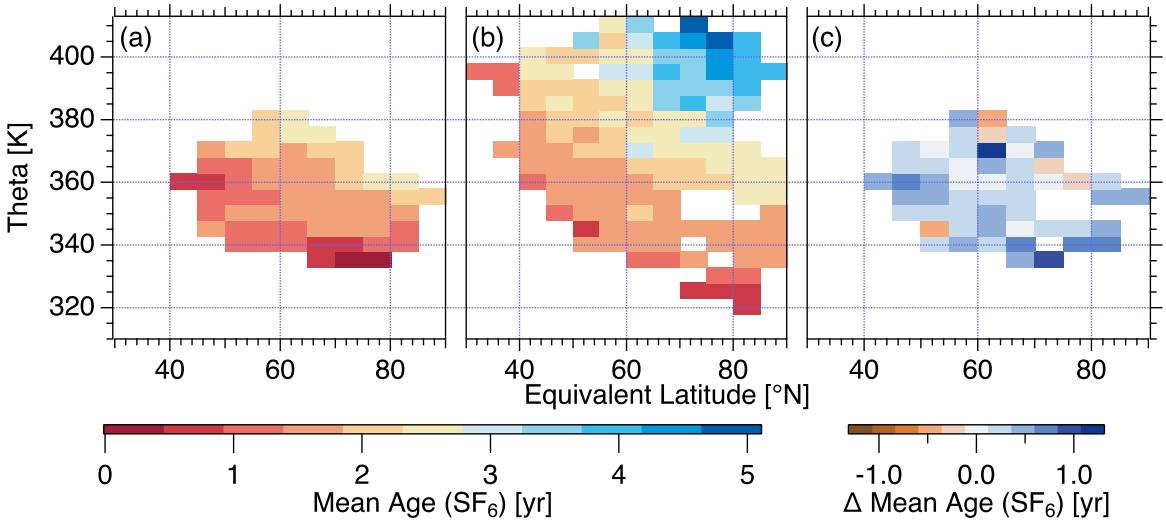

Figure 3. Distributions of mean age from SF_6 measurements in potential temperature - equivalent latitude coordinates for PV > 7 PVU. Panel (a) shows data for phase 1, (b) for phase 2 and panel (c) shows their absolute difference (phase 2-phase 1). The colour code represents the mean age. Blue colours in panel (c) indicate an increase of mean age in the subvortex region from January to March. Only bins with more than ten data points are shown.

~~modelled and observed to cause~~ Models and observations both show a high bias of up to one year in the polar vortex (Ray et al., 2017) or even more in mesospheric air (Engel et al., 2006b). ~~While this may cause a significant offset in mean age for polar vortex air, it is estimated that relative changes in mean age as discussed in this paper can be reliably derived from SF_6 observations~~Since we focus on the lower stratosphere far below the mesospheric loss region for SF6 and we further focus on age changes our data are not affected by this fact.

Figure 3 a) and b) show the distribution of mean age calculated from SF_6 measurements for phase 1 (January) and phase 2 (February / March)~~, respectively. As is evident on panel~~ . Panel (a) shows that the LS during phase 1 ~~the LS~~ is dominated by air masses of mean ages between 0.5 years to less than three years at maximum. The oldest air masses with mean ages older than two years were encountered at largest distances from the tropopause and potential temperatures ranging from $\Theta = 360$ K to $\Theta = 380$ K. In contrast, during phase 2 (panel (b)) in general much older air masses up to five years were found at potential temperatures of $\Theta = 410$ K. These higher potential temperatures at flight altitude are the result of the diabatic descent over the course of winter and indicate an increasing influence of air masses originating deeper in the stratosphere or from the Arctic polar vortex. To directly compare the temporal evolution of the age of air in the lower stratosphere panel (c) shows the difference of age of air between both phases ~~. It can be seen~~ and shows that the bulk of air inside the LS is getting older between $\Theta = 330$ K and $\Theta = 380$ K. The mean increase is 0.29 years, indicating diabatic downwelling due to the evolution of the polar vortex and thus an increased mean age in late winter.

4.1.2 Nitrous oxide

Nitrous oxide (N_2O) ~~is a very stable molecule with a~~ has a lifetime of 123 years (Ko et al., 2013) ~~. Its sources are~~ and is released at the surface ~~due to natural and anthropogenic emissions with a very small seasonal variability~~ with no chemical sources in the atmosphere (Dils et al., 2006). As a result ~~of the well-mixed troposphere and the absence of tropospheric sinks (Ciais et al., 2013)~~ N_2O has a ~~distinct background value in the troposphere, so mixing ratios below this value can be identified as stratospheric influenced (Müller et al., 2015). The tropospheric background value of N_2O between November 2015 and March 2016 in the northern hemisphere was~~ near constant tropospheric value of 329.3 ppb_V ~~, measured by the NOAA Global Monitoring Division (NOAA). Its annual increase was found to be~~ (winter 2015/2016 according to NOAA) that makes stratospheric influence identifiable (Müller et al., 2015). The mean annual tropospheric increase is currently approximately 0.78 ppb_V ~~in the last years (Hartmann et al., 2013)with an variability of 3-5 ppb_V (Kort et al., 2011).~~ per year (Hartmann et al., 2013) .

The main sink reactions of N_2O are due to photolysis in the UV-band (190 ~~nm~~ $\leq \lambda \leq$ ~~220~~ 220 nm) and the reaction with $O(^1D)$ which only occurs within the upper stratosphere (Ko et al., 2013). ~~Since there are no sources of~~ Thus, N_2O ~~in the stratosphere its profile~~ above the tropopause ~~changes and~~ shows a weak negative vertical gradient ~~. During~~ which maximizes during winter and spring ~~a stronger negative vertical gradient establishes because of the enhanced diabatic downwelling due to~~ due to the diabatic downwelling by the Brewer-Dobson circulation.

~~Figure 4 shows the tracer distribution of N_2O. During phase 1 (panel(a)~~ Figures 4 a) ~~values between~~ and b) show N_2O values between 276 and 325 ppb_V ~~and 276 ppb_V N_2O were obtained. Consistent with the distribution of mean age lowest values of N_2O less than~~ measured during phase 1 and values below 200 ppb_V ~~were measured by the end of February above $\Theta = 400$ K(panel (b)). Corresponding to Figure 3 panel (~~measured during phase 2 above Θ = 400 K. Figure 4 c) shows ~~the difference between phase 2 and phase 1 and it is evident that there is a general decrease of~~ an overall decrease in N_2O ~~observed in the whole LS~~in the polar lower stratosphere due to diabatic descent during winter, consistent with ~~the measurements of mean age , indicating an enhancement of the diabatic downwelling over the course of winter.~~ mean age changes (Fig. 3 c)).

4.1.3 Carbon monoxide

Carbon monoxide (CO) is released to the atmosphere mainly through incomplete combustion processes and methane oxidation as the only significant in-situ source. ~~It has therefore a large variability in the troposphere which is also affected by anthropogenic emissions.~~ Due to the high variability of ~~surface emissions CO has variable~~ anthropogenic surface emissions, CO mixing ratios in the ~~range of~~ nothern hemisphere vary from 70 ppb_V to 200 ppb_V (Prinn et al., 2000) ~~in the northern hemispheric troposphere with lifetimes~~ and the CO lifetime is on the order of weeks. In the lower stratosphere the main source of CO is the methane oxidation with the OH radical. The main sink ~~reaction is the oxidation with the OHradical where CO gets oxidized into CO_2, which leads to a longer lifetime in the order of several months under dark vortex conditions:~~

$$CO + OH \longrightarrow CO_2 + H$$

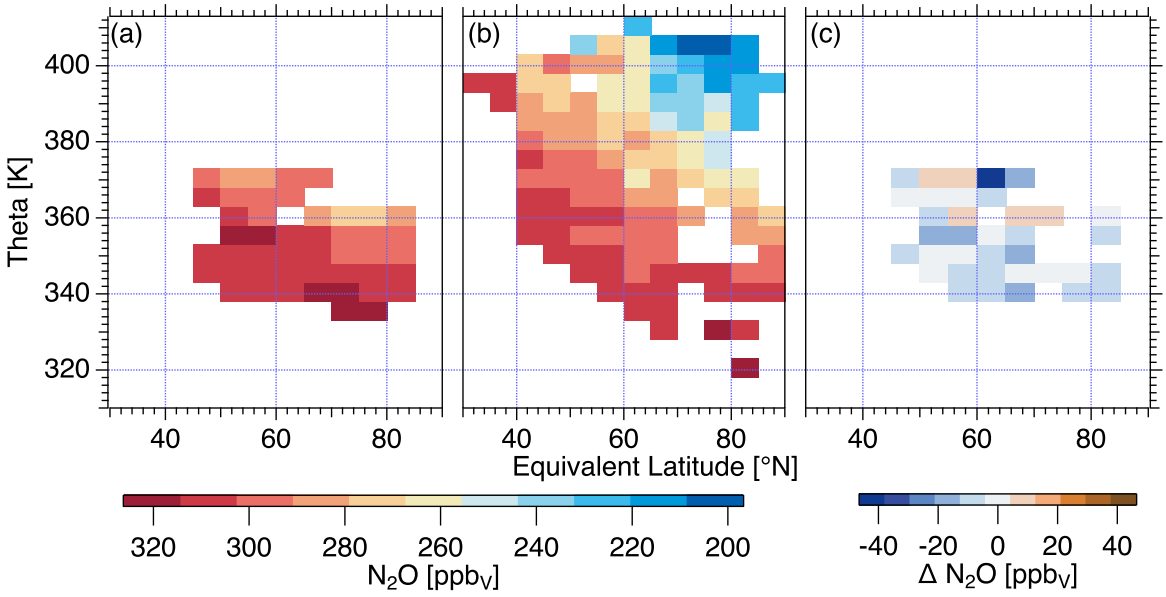

Figure 4. As Fig. 3 but for N_2O. Negative (blue) values in panel (c) indicate an overall decrease of N_2O in the measurement region in accordance with increasing mean age (Fig. 3c).

is oxidation by OH. The CO lifetime during polar night is a few months. In the stratosphere CO is controlled by production from methane oxidation and CO degradation. In the absence of transport from the troposphere this leads to an equilibrium between production and destruction of CO. We found an equilibrium value of 10-15 ppb_V in winter 2015/2016, depending on the integrated temperature history of the respective air mass in agreement with previous studies (Müller et al., 2016; Herman et al., 1999).

~~A potential additional source of CO in the Arctic winter stratosphere is the~~ The reaction of CH_4 with ~~reactive chlorine (Cl_x) which is nearly insignificant for~~ Cl is an insignificant source of CO in the lower stratosphere (Flocke et al., 1999). Transport from the mesosphere, where CO is produced from the photolysis of CO_2, also provides a potential source of CO via strong diabatic descent during winter under persistent polar vortices (Engel et al., 2006a). These potential influences are discussed in chapter 6.

Figure 5 shows the distribution of CO. During phase 2 (panel (b)) the lowest mixing ratios of 15 ppb_V were found at potential temperatures between $\Theta = 380$ K and $\Theta = 410$ K and equivalent latitudes > 60 °N. As can be seen by the vertical branch of the CO-N_2O correlation (Fig. 6 and 7), this value is the stratospheric equilibrium during late winter. Phase 1 (panel (a)) values ranged between 60 ppb_V and 17 ppb_V, hence the stratospheric background value was not measured in January 2016. A strong tropospheric influence is evident below $\Theta = 340$ K with CO values up to 57 ppb_V at phase 1 and 47 ppb_V at phase 2. Hence the overall distribution of carbon monoxide in the UTLS during the individual phases (Fig. 4 a) and b))seems to be consistent ~~to~~ with N_2O and mean age obtained from SF_6 measurements, despite its much shorter lifetime compared to the other species.

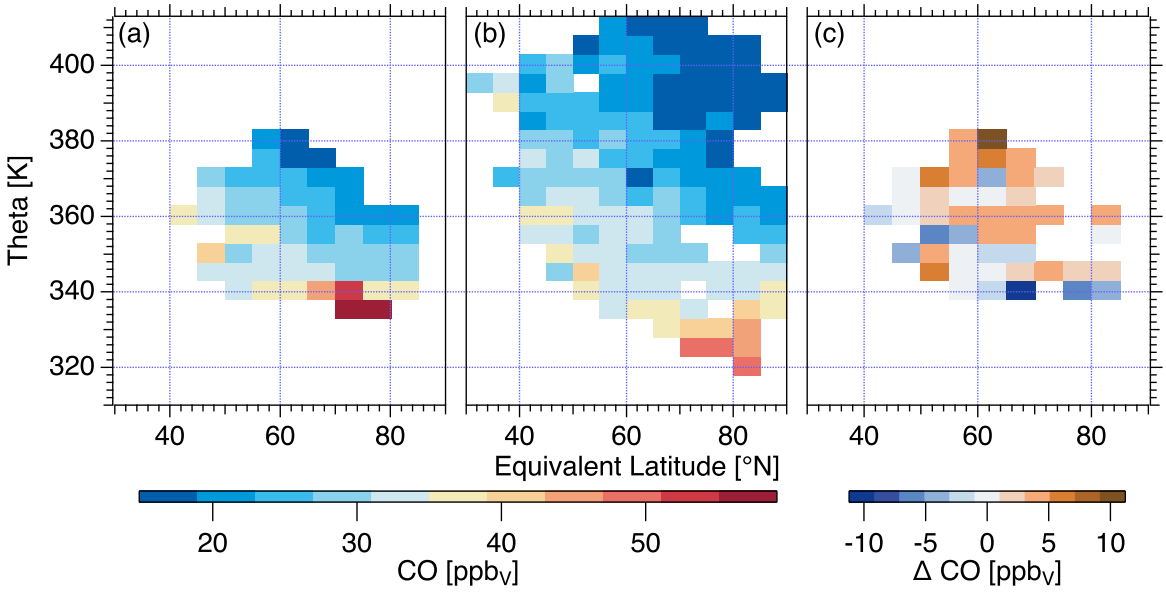

Figure 5. As Fig. 3 and Fig. 4 but for CO. Note the positive difference of CO in panel (c) indicating an increase of CO in the measurement region.

However, when comparing the differences of the respective phases (panel c), we see a different behaviour compared to N_2O and SF_6. We encountered an increase of carbon monoxide mixing ratios over the course of winter, which is at first glance in contradiction to the distributions of mean age and N_2O. While the distributions of long-lived tracers SF_6 and N_2O indicate an ageing of air masses, the increase of short-lived CO indicates a source of CO either from the troposphere or the stratosphere. Note that the increase is observed above $\Theta = 360$ K ~~, whereas below this level a decrease occurs.~~ and 50 °N equivalent latitude. Below $\Theta = 360$ K decreasing values are encountered. We will analyse the potential sources of CO in the following and ~~rise the hypothesis~~ suggest that CO increases due to an enhancement of mixing of tropospheric air from the tropical lower stratosphere over the course of ~~winter without a direct strengthening of tropospheric source emissions and mixing ratiosfrom below~~the winter without an increase of the upper tropospheric mixing ratios, which are affected by the surface emissions.

5 Analysis

We found a decrease of the long lived species SF_6 and N_2O with their lowest values ~~in the furthest regions from the~~ far above the local troposphere in late winter, which fits well in the general picture of enhanced downwelling of the Brewer-Dobson circulation ~~and the enhanced downwelling~~ in late winter/spring. The ~~contradicting~~unexpected, simultaneous increase of the short lived CO over the course of winter could indicate a strengthening of tropospheric transport by enhanced mixing with ~~fraction of air with low transit times into the~~ air from the tropical lower stratosphere.

5.1 ~~Identification of mixing on the basis of tracer-tracer correlations~~

In the following we will discuss this hypothesis and also other potential sources for the additional CO mixing ratios. ~~We will show that, despite of different potential source regions as the mesosphere or chemical in-situ production, this increase is originating from an enhanced isentropic mixing out of the TTL, interacting with the diabatic descent in the polar stratosphere.~~

5.1 Identification of mixing on the basis of tracer-tracer correlations

To identify mixing processes across the tropopause CO-O_3 correlations have been widely used (Fischer et al., 2000; Zahn et al., 2000; Hoor et al., 2002; Pan et al., 2004; Müller et al., 2016). Since ozone is affected by chemical processes particularly in the vortex region we use N_2O as a stratospheric tracer instead of ozone. Carbon monoxide, ~~here used as~~ used here as a tropospheric tracer also has sources in the mesosphere and via chlorine chemistry in the stratosphere. In the LS the influence of chlorine is small, compared to the reaction with the hydroxyl radical, therefore we investigated the influence of chlorine chemistry regarding methane which leads to the formation of CO. This influence will be discussed in detail later.

To analyse the ~~effect~~ effects of transport and mixing on the evolution of the UTLS composition we used the N_2O-CO relation as shown in Fig. 6. Tropospheric data ~~are represented by~~ have high N_2O values (> 328 ppb_V) and are accompanied by high CO values~~. Thus, stratospheric data appear for~~ , while stratospheric data have $N_2O < 328$ ppb_V. Due to the tropospheric background value of N_2O and the stratospheric equilibrium of CO, the troposphere can be identified as horizontal (high amount of N_2O, variable CO) branch and the stratosphere, free of tropospheric influence can be identified as vertical branch (low amount of CO, variable N_2O) of the correlation. Without any recent mixing, the tracer-tracer correlation of N_2O and CO would form an L-shape structure (Fischer et al., 2000). In the presence of ~~actual~~ rapid mixing a straight mixing line between two end members of the correlation ~~establishes~~ is established (panel a) (Hoor et al., 2002; Müller et al., 2016). As stratospheric CO will relax towards its stratospheric equilibrium value while N_2O is ~~chemical inert~~long-lived in the lower stratosphere, the initial linear correlation will become curved with time in case of inefficient mixing when the chemical lifetime is shorter than the time scale of mixing (panel b). Depending on the strength of mixing relative to the chemical CO sink the curvature will change and is less pronounced as the mixing gets more efficient (panel c). It is important to note that the change of CO relative to a given N_2O value can only be explained by a change of the ratio between mixing and chemical time scales. Mixing alone acts on both tracers N_2O and CO. Therefore a change of the shape of the curve is a direct result of the increased mixing relative to the chemical timescale, which is less efficient when mixing becomes stronger. Panel (d) of Fig. 6 shows additionally the correlation under mesospheric influenced conditions. In this case the correlation would rise to higher CO mixing ratios and lower N_2O mixing ratios, since N_2O gets destroyed and CO produced in the mesosphere.

Figure 7 shows the N_2O-CO correlation for POLSTRACC separated for phase 1 and phase 2, respectively binned in intervals of 5 ppb_V N_2O. It is evident that mixing of tropospheric and stratospheric air masses occurs in both phases. During phase 1 (blue curve) CO ranges between 20 ppb_V and 60 ppb_V at N_2O values between 323 ppb_V and 270 ppb_V. Notably the red curve (phase 2) shows a steeper gradient with CO values between 43 ppb_V and 15 ppb_V at N_2O values between 323 ppb_V and 180 ppb_V. There are higher CO mixing ratios for N_2O values lower than 310 ppb_V in the later phase 2 of the measurements. Additionally

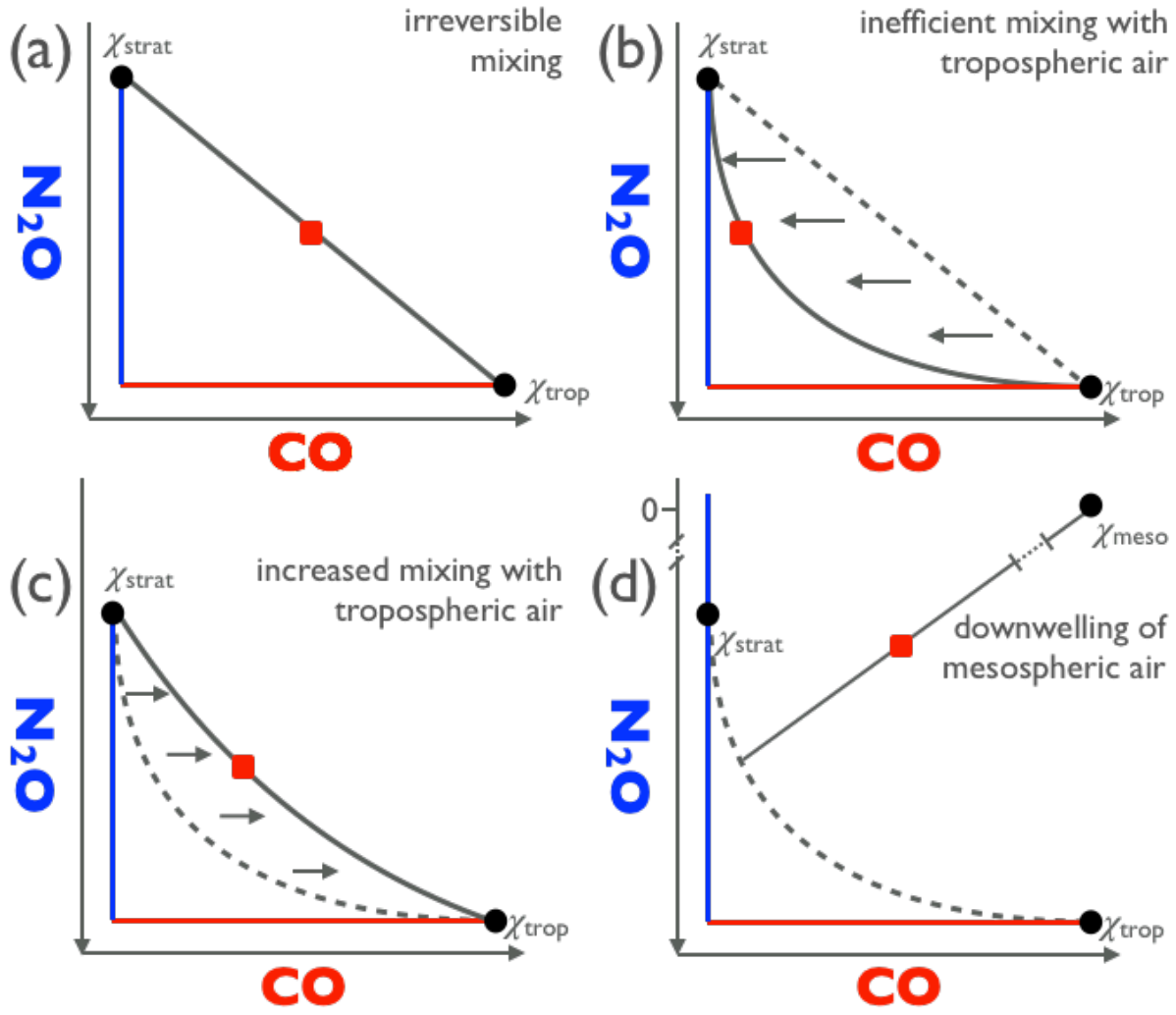

Figure 6. Sketch of tracer-tracer correlations with different lifetimes. Note that the N_2O axis is reversed. Panel (a) shows the L-shape structure with an air parcel (red box) on a straight mixing line for fast mixing timescales. The horizontal red line represents the tropospheric N_2O background, the blue vertical line the stratospheric CO equilibrium. Panel (b) shows the resulting curve in case of inefficient mixing compared to the chemical lifetime. Panel (c) shows the change of curvature, depending on the strength of mixing and panel (d) shows the influence of the mesosphere on the correlation.

the red curve tends towards ~~an~~ a CO equilibrium value of 15.67 ppb_V for N_2O values in the range of 220 ppb_V to 180 ppb_V. Most importantly, there is an increase of CO on N_2O isopleths between 313 ppb_V and 273 ppb_V N_2O over the course of winter. This is a remarkable result since we expect that due to the ageing of air inside the lower stratosphere in winter, the CO mixing ratio decreases with time. It is important to note that the correlation along the mixing line which connects tropospheric values with the stratosphere shows higher CO relative to N_2O in phase 2. As indicated in Fig. 6 this is a clear indication for enhanced mixing of tropospheric air masses for N_2O values > 273 ppb_V. Furthermore phase 1 shows higher CO values relative to N_2O compared to phase 2 for N_2O values larger than 313 ppb_V. Therefore we can conclude that regarding ~~to~~ the CO-N_2O correlation the ~~direct tropospheric impact~~ tropospheric impact on short timescales through the ExTL was greater in phase 1 than in phase 2, indicating enhanced mixing with tropospheric influenced air originating in the TTL region during phase 2.

A potential mesospheric impact is highly unlikely due to the fact that during phase 2 the N_2O-CO correlation tends towards the equilibrium value in the region of lower N_2O values. This influence will be discussed later in detail.

~~As shown before the analysed measurement region, which is covered in both phases , lies between potential temperatures of $\Theta = 340$ K and $\Theta = 380$ K . Additionally, the measurement data is filtered for potential vorticity values larger than PV = 7 PVU.~~

During both phases the UTLS between $\Theta = 340$ K and $\Theta = 380$ K was covered by our measurements. Therefore we assume the TTL (Fueglistaler et al., 2009) region (Fig. 1), where most of the tropospheric air masses are transported into the stratosphere (Schoeberl et al., 2006, and references therein) ~~as~~ is the main source for the enhanced CO values (Fig. 5 panel (c)). Further on, rapid eddy mixing of air from the TTL leads to an increase of tropospheric tracer signatures in the Arctic region (Rosenlof et al., 1997).

To quantify the increasing influence from tropospheric air masses in the lower stratosphere, we applied a simple mass balance approach to quantify the composition of the lower stratosphere. Therefore, we assume an air parcel in the lower stratosphere may consist of either upper stratospheric or tropospheric origin (Fig. 1). This mass balance system is solved to get the amount of tropospheric fraction f_{trop} of the measured air.

For a mixing ratio χ on a specific isentrope θ we assume

$$\chi(\theta) = f_{\text{trop}} \cdot \chi_{\text{trop}} + f_{\text{strat}} \cdot \chi_{\text{strat}} \tag{1}$$

and

$$f_{\text{trop}} + f_{\text{strat}} = 1 \tag{2}$$

which leads to the tropospheric fraction f_{trop} based on CO measurements

$$f_{\text{trop}} = \frac{\chi_{\text{CO,m}} - \chi_{\text{CO,strat}}}{\chi_{\text{CO,trop}} - \chi_{\text{CO,strat}}} \tag{3}$$

with $\chi_{\text{CO,m}}$ the measured CO mixing ratio, $\chi_{\text{CO,strat}}$ the stratospheric CO background which was set to 15.7 ppb_V as mean of the vertical branch of the CO-N_2O correlation and $\chi_{\text{CO,trop}}$ the tropospheric CO entry value in the TTL.

~~Earlier studies~~ In-situ measurements have shown that CO mixing ratios above the tropical tropopause are at levels between 50

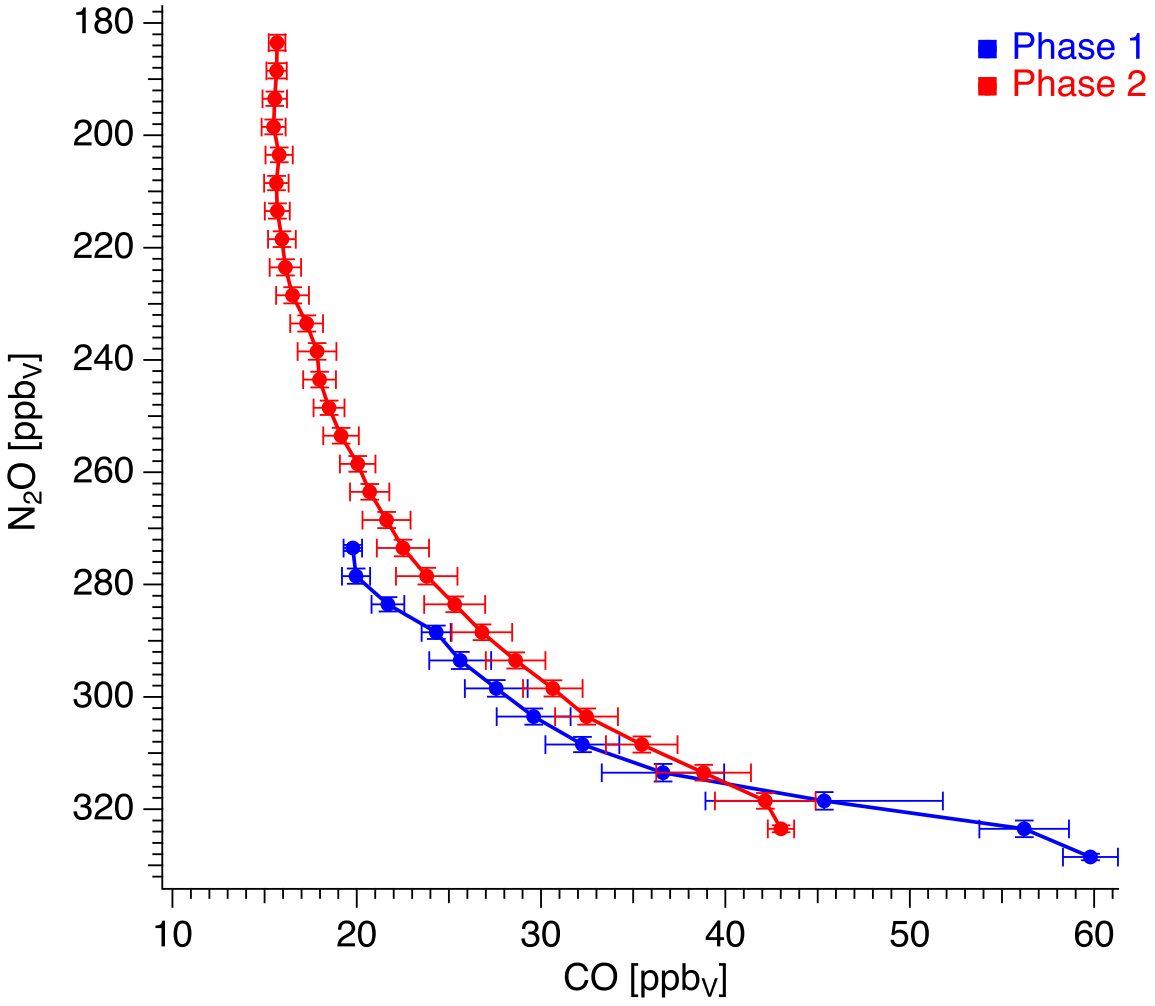

Figure 7. N_2O-CO correlation for POLSTRACC flights with PV > 7 PVU. The blue curve represents phase 1, the red curve represents phase 2. Data are binned in steps of 5 ppb_V N_2O. The variability in each bin is given by the vertical and horizontal lines, respectively.

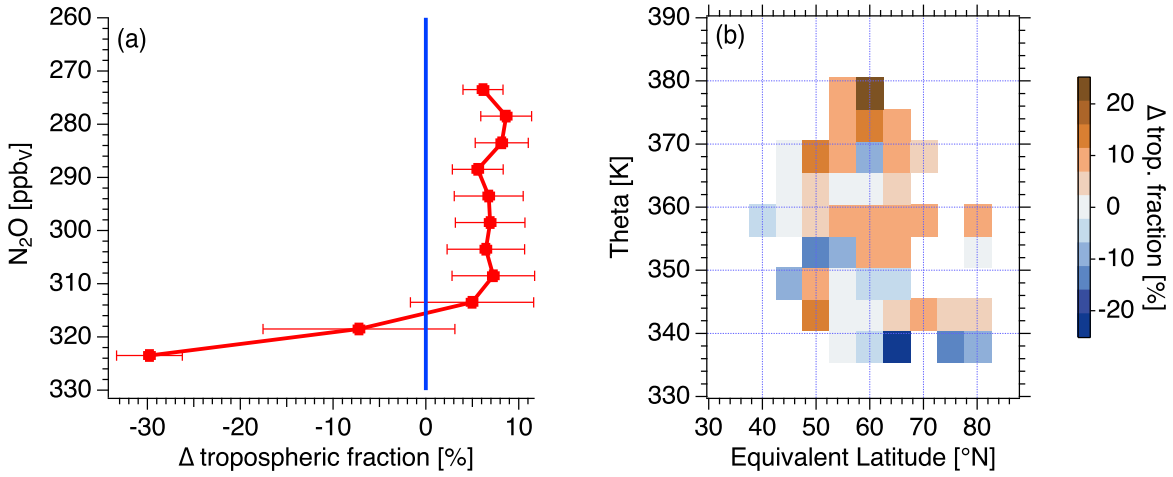

Figure 8. Panel (a): Tropospheric CO fraction from the mass balance equation as a function of N_2O showing the difference between phase 2 and phase 1. Panel (b): The same as (a) but as distribution against equivalent latitude. Red colours indicate an increase of the tropospheric CO fraction.

and 60 ppb_V (Herman et al., 1999; Marcy et al., 2007).

~~The~~ Figure 8 shows the difference of the calculated tropospheric fraction f_{trop} between phase 2 and phase 1 ~~is shown in Fig. 8~~ as a function of N_2O, which acts as a quasi vertical coordinate. The CO increase over the course of winter corresponds to an increase by f_{trop} of ~~6.8(3.7)%~~ $(6.8 \pm 3.7)\%$ between 313 ppb_V and 273 ppb_V N_2O ~~.~~by assuming 60 ppb_V of CO at the tropical tropopause as provided by in-situ aircraft data from Herman et al. (1999); Marcy et al. (2007). Using CO_{trop} = 80 ppb_V as indicated by MLS at 100 hPa one obtains 32 % lower values for f_{trop}, which is still a significant increase of tropospheric air masses. Note that additionally the tropospheric fraction decreases towards more tropospheric N_2O values from phase 1 to phase 2. This is a clear evidence that an increase of the CO mixing ratio at the tropopause is not the cause for the observed lower stratospheric CO increase. This would be consistent with an increase of the fraction of young air of tropospheric origin and more efficient mixing as indicated in Fig. 6.

Panel (b) shows the distribution against equivalent latitude. Note that the observed increase is most prominent above $\Theta = 360$ K. This is a clear indication that mixing at $\Theta < 360$ K is suppressed due to the strong subtropical jet, which acts as a barrier for mixing (Haynes and Shuckburgh, 2000) and would be consistent with enhanced mixing out of the TTL region.

5.2 Age spectra analysis

For further analysis of the relationship between diabatically descended, aged air with ~~high~~ longer transit times and potentially mixed with young tropospheric air with ~~low~~ shorter transit times we use age spectrum calculations of the CLaMS (Chemical

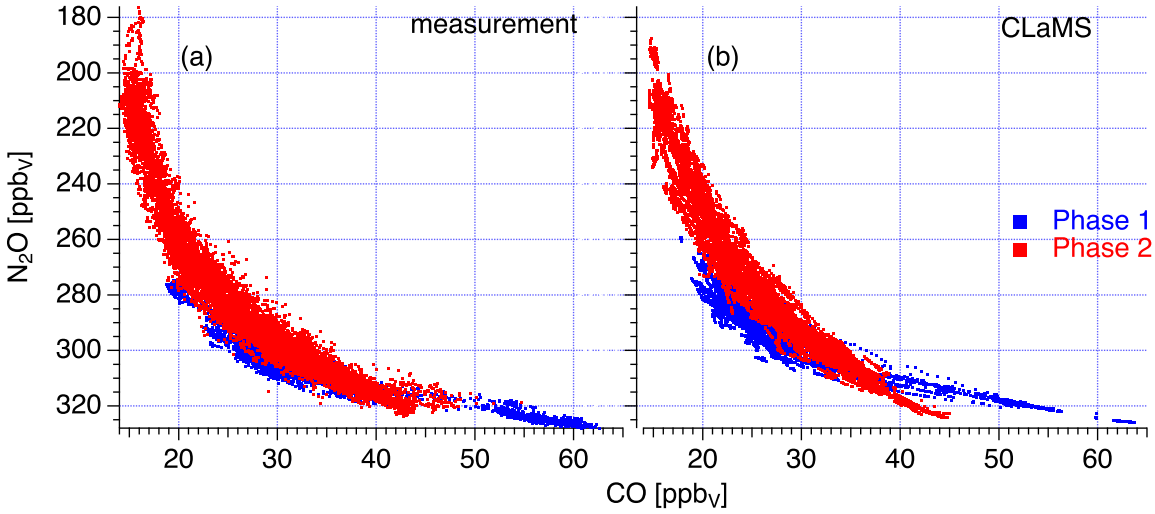

Figure 9. N_2O-CO ~~correlation~~ scatter plot measured by the TRIHOP instrument (a) and CLaMS model output (b). Phase 1 coloured in blue, phase 2 coloured in red. The model output is available along the flight track with a time resolution of ten seconds.

Lagrangian Model of the Stratosphere) (McKenna et al., 2002; Ploeger et al., 2015; Ploeger and Birner, 2016) model, which gives ~~informations of~~ information on the full transit time distribution. Notably we have the age spectral information for each individual data point along the flight track and therefore can directly compare our measurements with the spectrum.

To test if the model is able to reproduce the observations of tracers we compared CO and N_2O from CLaMS with the measurements (Fig. 9). Model output is available along the flight track with a time resolution of ten seconds. Figure 9 shows the N_2O-CO ~~tracer correlation~~ scatter plot for each data point. Panel (a) shows the correlation measured with the TRIHOP instrument, panel (b) shows the correlation calculated out of the CLaMS model. As is evident CLaMS correctly represents the increase of CO relative to N_2O from phase 1 to phase 2. Also the separate branches of the two phases are reproduced and the crossing of the correlation at 40 ppb_V CO and 310 ppb_V N_2O is consistently simulated.

This remarkable agreement between model and observations further motivates the usage of CLaMS for age analysis of our measurements.

As mentioned before, CLaMS is able to calculate the full transit time distribution of analysed air masses for each individual data point along the flight track. Figure 10 shows the averaged age spectra of the CLaMS model for the respective phase (panel (a)) and ~~the difference of them~~ their difference (panel (b)). Vertical ~~dotted~~ solid lines represent the mean age of the respective phase (blue and red) calculated by the CLaMS model, the dashed vertical lines ~~separates~~ separate young air masses with a mean age lower than 0.5 years and old air masses with mean age larger than 2 years. Since we have the full transit time distribution of each data point, we can compare this relation between the different parts of the age spectrum. An increase of the tropospheric

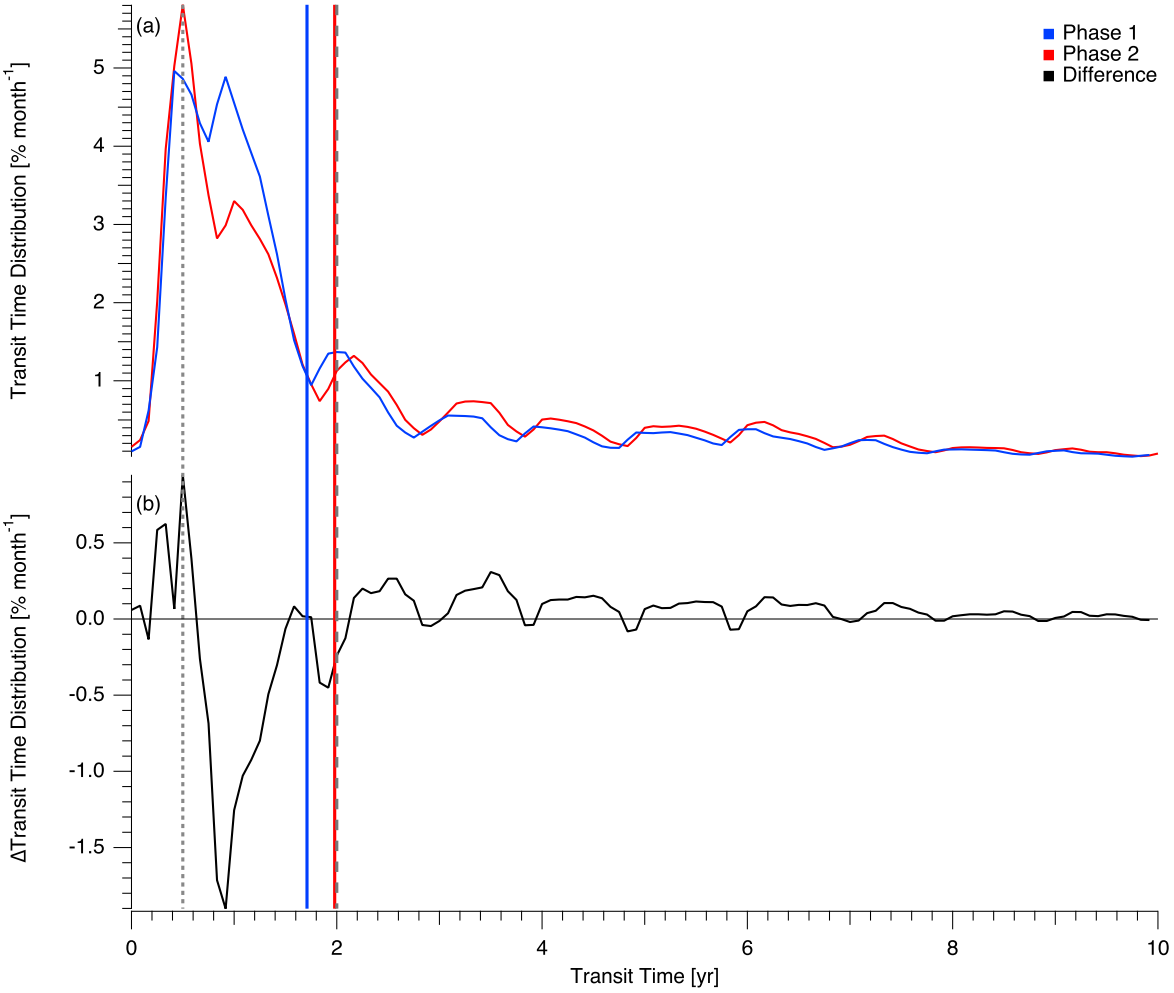

Figure 10. Panel (a): Averaged age spectra simulated by CLaMS for phase 1 (blue) and phase 2 (red). These spectra represent the mean of the individual age spectra available for each data point along the flight track. The mean age is indicated by the respective coloured vertical lines (phase 1: 1.71 years, phase 2: 1.98 years). The difference between both spectra is given in panel (b) showing an enhancement of both, young and old air masses from phase 1 to phase 2. Vertical dashed lines indicate the transit times of six months and 24 months, respectively (see next figures). The bin size of a data point is one month.

fraction would be linked to an increase of the part of the age spectrum with low transit times as indicated by the observed increase of CO relative to N_2O.

~~From Fig. 10 panel (b) it is evident that there is an~~ Figure 10 shows an absolute increase of air masses older than two years up to 0.3% ~~. In the range of~~ per month. For air masses younger than six months, there is also an increase of the age spectrum between phase 2 and phase 1 evident~~which is~~, with maximum values up to 0.9% per month, larger than the increase of the old

air masses. The relative change of air masses younger than six months is 19.5 % and for air masses older than two years 76.4 %. The increase of the young fraction is in agreement with the observed ~~increase of CO . It is therefore an indication for an~~ CO increase, indicating increased mixing with air from the TTL at the end of winter.

Since the mean age is calculated as the first moment of the distribution its value is most sensitive to changes ~~by~~ in the old tail of the distribution (Hall and Waugh, 1997). Therefore the mean age rises by 0.27 years from 1.71 years to 1.98 years as a result of the increase of the age spectrum distribution for air masses older than two years. This matches the mean age increase of SF_6 and indicates, ~~combined~~ in agreement with the decrease of N_2O, the overall ageing in the lower and lowermost stratosphere over the course of winter. Since the integral over the ~~Greens~~ Green's function is normalised to one, the increases of air masses older than two years and younger than six months must result in a relative decrease in between. Therefore air masses with mean ages between 0.5 years and 2 years are more enhanced in phase 1 than in phase 2, which is evident by the change of the transit time distribution up to -1.9% per month.

To further investigate the relationship of young versus aged air~~we used the spectral information for each individual data point in the following way. We~~ , we calculated the accumulated fraction of air masses with transit times lower than six months and older than two years ~~, respectively,~~ for each data point. Figure 11 shows the binned fraction of air masses with transit times lower than six months versus the modelled mean age.

The comparison of the ~~correlation~~ scatter plot for different times (phase 1 and phase 2) shows that for a given mean age a significant increase of the young tropospheric contribution is evident. Thus, according to the model and in agreement with the observed increase of CO, the late winter LS is more affected by tropospheric young air. Therefore our results demonstrate, that the mean age is an incomplete descriptor when referring to chemical properties of air masses involving different chemical life times of species. Since the mean age is just a single number it might be insensitive to changes of the processes and time scales contributing to the mean, which however affect the chemical properties of the air parcel by e.g. enhanced mixing of short-lived species. Therefore it is important to account for the full spectral shape when referring to chemical properties of an air mass rather than only the mean age.

During winter 2015/16 CO mixing ratios in the LS increased from January to March while long-lived trace gases denote an ageing of the LS. The analysis of CO-N_2O correlations, ~~mass balance systems of~~ the mass balance equation of irreversible mixing and transport transport pathways in the LS and model simulations points towards an increased influence of tropospheric air masses from the tropical lower stratosphere. Additional potential sources of CO in the LS are discussed in the following.

6 Discussion

Since there are different sources for CO at different locations in the atmosphere an increase of carbon monoxide mixing ratios can be due to (i) an increase of isentropic mixing out of the TTL, (ii) an increase of the tropospheric source strength, (iii) a potential influence of the mesosphere and (iv) a change of chemical reaction cycles due to higher amounts of reactive chlorine in the stratosphere. As already discussed the increase of enhanced tropospheric source emissions (ii) is highly unlikely (see Fig. 7). Since our analysis points to an increase of isentropic mixing out of the TTL (i), the possible influence of points (iii)

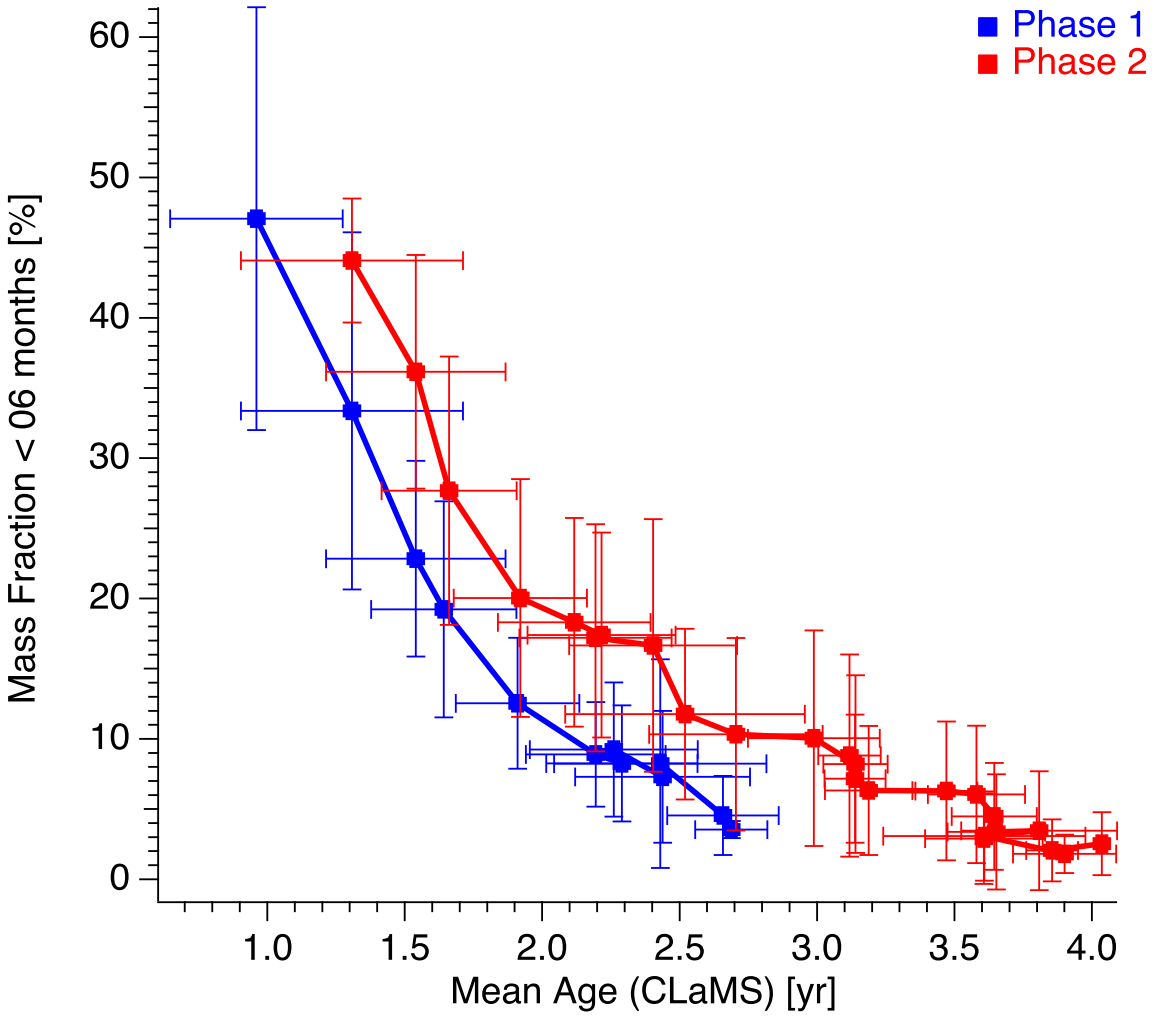

Figure 11. Mean age versus air fractions with transit times < 6 months from the age spectra simulated by CLaMS for phase 1 (blue) and phase 2 (red). Each data point is binned in steps of 5 ppb_V N_2O. The variability in each bin is given by the vertical and horizontal lines, respectively.

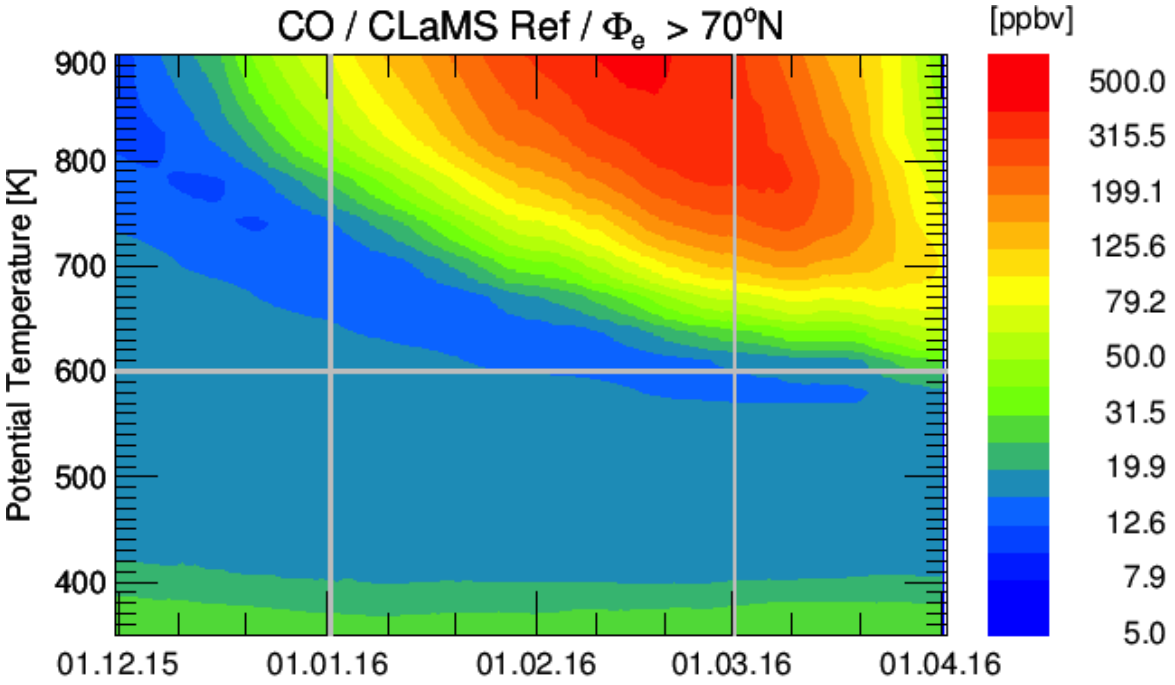

Figure 12. Temporal evolution of zonal mean CO for equivalent latitudes > 70 °N simulated by CLaMS for winter 2015/2016.

and (iv) have to be further discussed.

Carbon monoxide is produced in the mesosphere due to the photo-dissociation of carbon dioxide~~through high energetic sunlight~~. Therefore the composition of mesospheric air masses is clearly distinct from air mass composition of the stratosphere. Rinsland et al. (e.g. 1999) found increased CO mixing ratios up to 90 ppb_V at altitudes around 25 km or $\Theta = 630 - 670$ K and Engel et al. (2006b) found CO values of 600 ppb_V at an altitude of 32 km. Both studies show very low N_2O mixing ratios (< 50 ppb_V). Although the authors found ~~a layer~~ layers of mesospheric air descending down to 22 km, this is not evident for the Arctic winter 2015/16 and lowest N_2O mixing ratios are found to be in the order of 200 ppb_V. ~~CO mixing ratios simulated by CLaMS in the chemistry setup are shown in Fig. ?? as vortex core average profile along the time throughout the winter and spring. Potential downward transport from mesospheric air is largest in the polar vortex core due to diabatic descent of air masses. With this, air with enriched CO mixing ratios is transported downward into the stratosphere over the course of winter.~~ This is reflected in the MLS observations that determine the CLaMS upper boundary at $\Theta = 900$ K potential temperature ~~.~~ (Fig. 12). The simulation indicates the expected downward transport ~~but sees no~~ of mesospheric influenced air~~masses in the LS.It is evident that CO mixing ratios increase at $\Theta = 900$ K from December 2015 to end of February 2016 which descended from $\Theta = 900$ K~~ , but down to $\Theta = 600$ K at the end of March ~~2016. Therefore these enhanced CO mixing ratios do not affect~~

our measurement region below about Θ = 410 K. For potential temperatures below Θ = 420 K a slight increase in the CO mixing ratio with time is simulated, which does not originate in the stratosphere or mesosphere in 2016 in agreement with our observations(see Fig. 7). Regarding to the composition of CO (Fig. 5) it is evident that the measured CO mixing ratios decrease with altitude and the lowest values are found . CO values minimize at the highest regions flight levels and equivalent latitudes.This indicates that the increase of measured CO mixing ratios has no mesospheric origin, because the enhanced CO mixing ratios are only transported down to Θ = 600 K potential temperature in the CLaMS model.

Furthermore, an additional influence of descended mesospheric air into the lower stratosphere would not only impact CO but also N_2O. Due to the chemical differences between the stratospheric and the mesospheric composition, mixing of mesospheric air, enriched in CO and depleted in N_2O, would lead to mixing lines very strongly differing from the observed relationship (see Fig. 6). Importantly, , which is not observed in agreement with the CLaMS N_2O-CO correlation scatter plot (Fig. 9)almost perfectly mirrors the observations, further indicating no mesospheric influence on the simulated correlation.Additionally the age spectrum calculations of the CLaMS model provide mass fractions of air masses regarding their stratospheric residence time. As is evident from Fig. 11 there is a significant increase of air masses younger than six months at typical mean ages for lower stratospheric air masses and mesospheric influence on the basis of our analysis is highly unlikely.

Net change of CO from January to March for air masses in the Arctic vortex (equivalent latitude > 65 °N) due to chemical reactions in the stratosphere and mesosphere calculated by CLaMS. The blue line represents the statistical mean, the dashed lines the 1-σ standard deviation.

In general, another important source of carbon monoxide in the atmosphere is the reaction of methane with reactive chlorine, which is not significant in the lower stratosphere (Flocke et al., 1999). Eventhough the influence of the methane reaction with Cl on CO is lowHowever, air masses enriched in reactive chlorine can be could have been transported downwards, providing potentially potential reactants for the chemical production of CO. It may not be the case in this specific winter because of unprecedented low temperatures and resulting higher chlorine activation. Thereforethis aspect was also investigated in more detail. For this aim Therefore, we simulated the CO yield from the reactions of CH_4 with chlorine, OH and $O(^1D)$. To investigate the chemical sources and sinks of CO, (^{1}D) using CLaMS simulations in the boxmodel modewere performedbox model mode. A large number of single air parcel backward trajectories were calculated starting on 15 March from locations within the vortex core (equivalent latitude > 65 °°N; potential temperature between Θ = 350 K and Θ = 500 K)ending Θ = 350 K and Θ = 500 K). Trajectories end on 15 January and chemical composition changes were calculates using the CLaMS chemistry module running forward in time for a subset of the trajectories with equivalent latitudes > greater 50 °°N on 15 January (21480 trajectories). Figure 13 shows the statistical evaluation of the net CO change due to chemistry over the period as function of potential temperature on 15 March. The blue line represents the statistical mean and the dashed lines the 1-σ standard deviation. The mean overall change is even negative over the entire profile, which is due to the oxidation of the produced CO by the reaction with OH. Therefore we conclude that the observed increase of CO in phase 2 is not due to the additional chemical source reaction. The significant increase of air masses younger than six months (Fig.11) also indicates a strong contribution of young rather than mesospheric air.

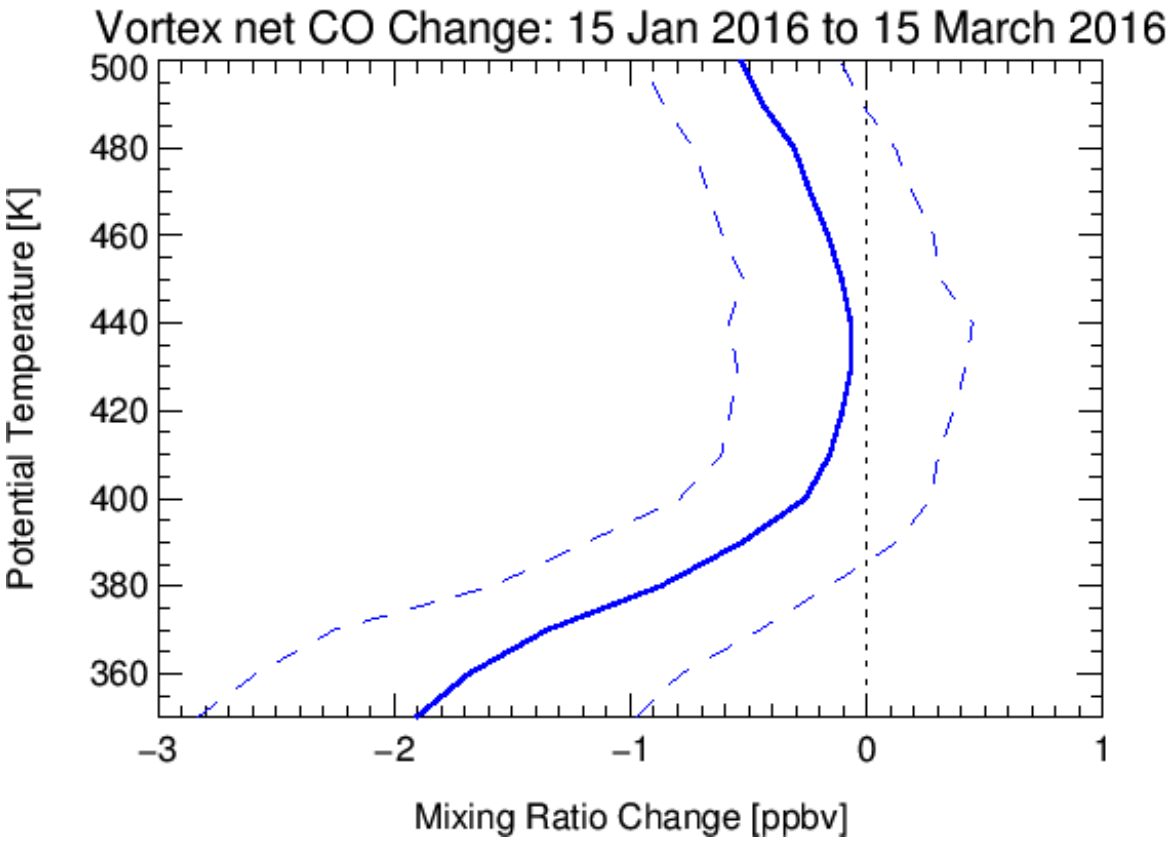

Figure 13. Net change of CO from January to March for air masses in the Arctic vortex (equivalent latitude > 65 °N) due to chemical reactions in the stratosphere and mesosphere calculated by CLaMS. The blue line represents the statistical mean, the dashed lines the 1-σ standard deviation.

Figure 13 shows the statistical evaluation of the net CO change due to chemistry over the period as function of potential temperature on 15 March. The blue line represents the statistical mean and the dashed lines the 1-σ standard deviation. As is evident the mean overall change is even negative over the entire profile, which is due to the oxidation of the produced CO by the reaction with OH. Therefore we conclude that the observed increase of CO in phase 2 is not due to the additional chemical source reaction.

To investigate if transport and increased mixing of air mass fractions with transit times smaller than six months in winter 2015/16 was special compared to other years we analysed the climatology of these fractions from 2004 to 2016 and compared it to the calculated fractions in winter 2016, both from the CLaMS model (Fig. 14). The colour code represents the fractions of air masses with transit times smaller than six months, the contour lines represent the mean age and the thick black line indicates the WMO tropopause.~~Note, that mixing of these air masses is significantly stronger depicted in the southern hemispheric polar~~

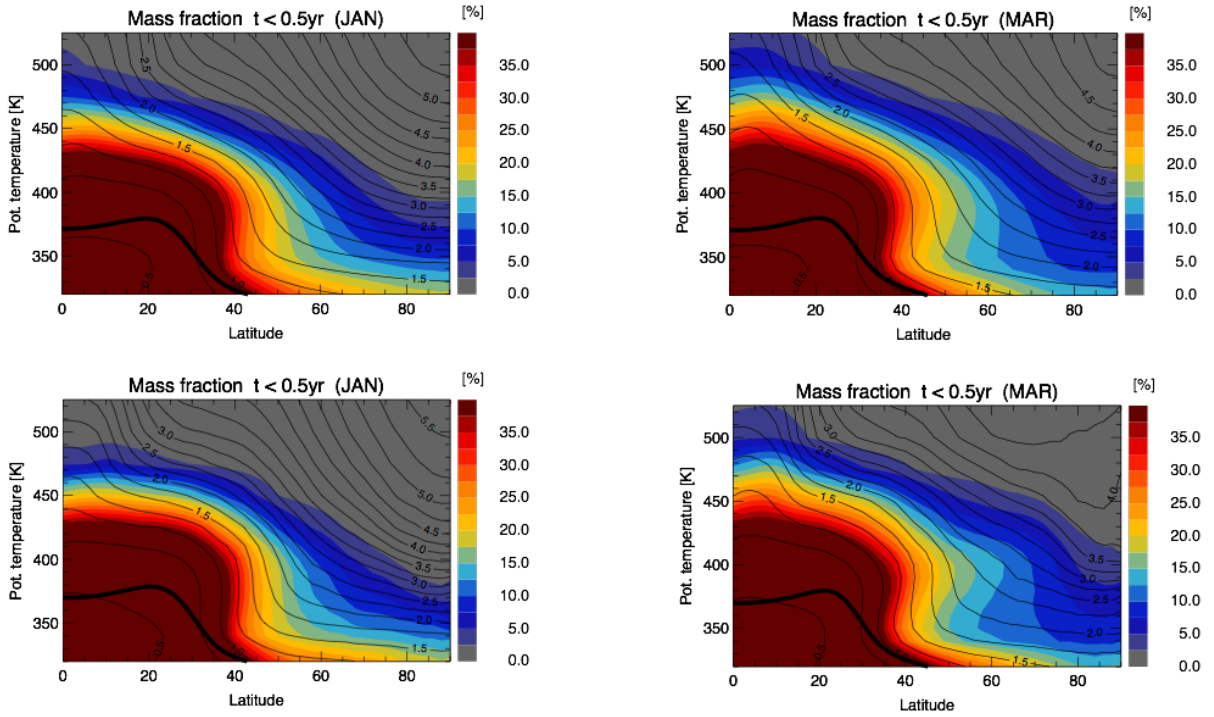

Figure 14. Zonal mean of air mass fractions (colour code) with transit times < 0.5 years for ~~JFM~~ January (left) and March (right) from 2004 to 2016 climatology (upper row) and for 2016 only (lower row) against potential temperature. The contour lines show mean age in years and the thick black line the WMO tropopause.

~~region. This must not directly be compared to the northern hemisphere, since this time span represents summer months in the southern hemisphere, where mixing is larger compared to the winter.~~

~~From the climatology it is evident~~ The climatology shows that the largest fraction of air masses with transit times smaller than six months exceeding 73% is found between ~~30 °S~~ the equator and 30 °N up to $\Theta = 430$ K. In January this strong signal has a sharp gradient at $\Theta = 450$ K. These air fractions are transported from January to March to the poles~~so that between~~ . As a result northward of 70 °N ~~and 90 °N~~ the fraction of air masses with transit times smaller than six months increases by 5% ~~at $\Theta = 380$ K~~ between $\Theta = 360$ K and $\Theta = 420$ K. From January 2016 to March 2016 this transport is even stronger than in the climatology, as ~~is evident from the convex structure of the distribution gradient to the north pole. From~~ shown by the different horizontal gradients in Fig. 14~~it is also evident, that the~~ . The mean age in March compared to January at $\Theta = 400$ K shows a ~~simultaneous~~ higher value in both, the climatology as well as the winter 2015/2016, whereas the structure of the mean age contours show a more horizontal meridional gradient in the winter 2016 compared to the climatology.

Finally, these findings ~~show an enhancement of young fraction of~~ highlight the role of mixing of young air in the lower stratosphere of the polar Arctic region with an underlying increase of mean age of air indicating downward transported air

masses of older air fractions. ~~Both the~~ The enhanced transport of young air ~~and the increased downwelling are~~ is evident from the climatology and turn out to be particularly strong in the winter 2016.

7 Summary

We present tracer measurements of CO and N_2O measured during the POLSTRACC campaign in winter 2015/16 on board the German HALO research aircraft. The winter 2015/16 was characterised by an extreme cold and stable polar vortex which broke up due to an MFW on 5. March 2016. In combination with measurements of SF_6 and model simulations by the CLaMS model it was possible to analyse the contributions of diabatic transport and isentropic mixing in the ~~region of the upper troposphere / lower stratosphere~~UTLS region. The mixing ratios of the long-lived trace gases N_2O and SF_6 ~~decrease~~ decreased over the course of winter and therefore ~~denote~~ denoted an overall ageing due to subsiding air masses in the Arctic polar lower stratosphere. The calculated mean age ~~shows for SF_6~~ based on measured SF_6 shows an ageing of 0.29 years (see Fig. 3 (c)) and for CLaMS 0.27 years (see Fig. 10 (a)), respectively. Remarkably, the short-lived species CO increases at the same time. Since mixing can be identified by tracer-tracer correlations we used CO-N_2O correlations to quantify the relation between transport and chemistry. Our analysis shows an increase by 3.7 ppb_V CO relative to N_2O, which can be linked to an increase by 6.8 % of mixed air masses out of the TTL region. The comparison with the CLaMS model shows a very good agreement between measurements and model calculations. The CO-N_2O correlation is well reproduced by the model. Analysis of the averaged age spectrum for the respective phase shows that there is a simultaneous increase of fractions of air with transit times larger than two years and fractions of air with transit times smaller than six months. Since the mean age itself is most sensitive to changes on the old tail of the age spectrum, the ageing of air masses in the LS over the course of winter can be explained by the increase of old air masses, characterised by low N_2O and SF_6 measurements. Increased mixing of young air masses adds to this and leads to an increased fraction of the younger part of the age spectrum, consistent with the observed increase of CO. It is evident that this enhancement is due to stronger mixing processes out of the TTL region, where fresh tropospheric air is mixed into the polar lower stratosphere. Other potential sources of CO like mesospheric air and chemical reaction of CH_4 with chlorine are unlikely to have caused the ~~observerd~~ observed increase of CO.

Therefore we conclude that the Arctic lower stratosphere in March was strongly affected by mixing with young tropospheric air, which partly compensates for the overall ageing. These aged air masses are isentropically mixed with younger air masses out of the TTL region. The observations are in-line with the climatology of mixing from ~~2005-2015~~ 2004-2015 on the basis of Era-interim by the CLaMS model and highlight the importance of horizontal mixing from the tropics for the Arctic winter UTLS.

Author contributions. Jens Krause carried out the measurements and analysed the data with the help of Peter Hoor. Felix Plöger and Jens-Uwe Grooß did the model simulations with the CLaMS model. Andreas Engel, Harald Bönisch and Timo Keber provided the measurement data of SF_6 and mean age. Peter Hoor, Andreas Engel, Felix Plöger and Jens-Uwe Grooß provided helpful discussions and comments. Jens

Krause and Peter Hoor wrote the manuscript. Hermann Oelhaf, Björn-Martin Sinnhuber and Wolfgang Woiwode coordinated the POLSTRACC project.

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