Reply to reviewer #1 Estimating Brewer-Dobson circulation trends from changes in stratospheric water vapour and methane

We thank the referee for the review and for the helpful and detailed comments. We give a pointby-point reply below, where the reviewer comments are repeated in black. The replies to the reviewer's comments are in blue text color. The revised text is given in italics and in quotation marks, with the positions of the corrected sentences in the revised version noted in brackets.

General remarks

The analysis is well done and may help with deducing trends in the stratospheric circulation from trace gas observations, which has been notoriously difficult. The topic is appropriate for ACP, the methods are clearly described and the figures are well done. I suggest the manuscript be accepted for publication with consideration of the minor comments below.

We thank the reviewer for this positive comment. In the revised version all suggested comments have been taken into account. In particular, we improved formulations and wording throughout the text as well as the captions of Fig.1,2,6 and the discussion of Fig.7. We also improved the description of the choice of zones for the parameterised age spectrum analysis (Appendix B), related to the remarks of Reviewer #1.

Specific Comments

1. PAGE 2

Line 43: 'CO2 has a seasonal cycle...'

Thank you for the remark. It is corrected at the revised version (p2, L44). "... CO_2 has a seasonal cycle..."

2. PAGE 3

Line 55: awkward phrasing 'for other than', maybe 'compared to' instead?

It is corrected at the revised version (p3, L56). "...longer time periods compared to the canonical species SF₆ and CO₂..."

Line 77: 'Also in our consideration is meant that there are no any' is hard to understand. Perhaps rephrase to 'The only significant source of H2O considered here is CH4 oxidation (e.g. we neglect all other hydrocarbons...'

Thank you for the remark. It is corrected at the revised version (p3, L78). "The only significant source of H_2O considered in this work is CH_4 oxidation."

3. PAGE 6

Line 179: 'depths of the BDC' is unclear. Maybe 'vertical transport by the BDC'.

It is corrected in the revised version (p6, L182). "...*affected by the vertical transport of the BDC*..."

4. PAGE 7

Figure 1 caption: I would suggest moving the last two sentences of the caption up into Section 2.1 since they describe important details of the CH4 boundary conditions for the model run.

Thank you for the remark. The suggested text from the caption of Fig. 1 was moved to Section 2.1 (p4, L107).

"The boundary conditions at the surface are prescribed based on ground-based measurements in the lowest model level (below ≈ 4 km). CH₄ mixing ratios are taken from the zonally-symmetric NOAA/ESRL dataset (e.g., Masarie et al., 1991) from 1990 to 2011, and from zonally-resolved AIRS data (e.g., Xiong et al., 2008, 2013) for 2011-2017."

Why is the CH4 boundary condition changed from NOAA to AIRS after 2011?

Thank you for this question regarding clarity of the used CH₄ boundary condition.

The CH₄ boundary condition in CLaMS has been switched in 2011 to take advantage of the better sampling of AIRS data in comparison to NOAA, although accepting the apparent discontinuity. For the results of this paper, the discontinuity has only negligible effects, as also with this boundary condition the results of the CLaMS model are internally consistent. In particular, when using age spectra (not a lag time) in the FULL and C-CORR methods the change in the CH₄ boundary conditions in 2011 does not affect the results. Also for the other approximation methods CH₄ and FRF information is mainly used from the period 2002-2006, such that the impact of the change in CH₄ boundary conditions in 2011 is irrelevant.

We added the respective text to the text (p4, L110).

"The CH_4 boundary condition has been switched in 2011 to take advantage of the better sampling of AIRS data in comparison to NOAA, although accepting the apparent discontinuity. Because the results of the CLaMS model are internally consistent, the discontinuity has only negligible effects."

5. PAGE 9

Line 208: remove 'used', '...and the method of calculating FRF...'

It is corrected in the revised version (p8, L209).

"The difference between C-CORR and FULL is in the correlation between AoA-FRF and the method of calculating FRF."

6. PAGE 10

Figure 2 caption: 'lapse' is misspelled

Corrected in the revised version (p10, caption to Fig. 2). "The black line is the (lapse rate) tropopause calculated from..."

7. PAGE 15

Figure 6 caption: The text in the parenthesis after 'FULL' and 'APPROX' isn't necessary since it's partly repetitive from the Figure 5 caption and is explained in the text.

Thank you for this remark. The suggested text was removed from the caption of Fig. 6. (p15, caption to Fig. 6).

"...(*b*) *FULL*, (*c*) *APPROX*, (*d*) *C*-*CORR*..."

8. PAGE 16

Line 367: '...stems from the differences in the AoA-FRF correlations used in each method...'

Corrected in the revised version (p15, L351). "The difference between the results from C-CORR and FULL methods stems from the differences in the AoA-FRF correlations used in each method, and the explicit FRF calculation."

Line 373: '...discussed earlier in the paper...'

It is corrected at the revised version (p16, L361). "It was mentioned earlier in the paper that stratospheric H_2O is ..."

Figure 7: The constant FRF-AoA correlation approximation appears to bias the AoA trend too positive over both time periods and nearly all the stratosphere. Although you don't try to improve the APPROX method with an improved treatment of the FRF-AoA correlation it seems there might be a simple correction made for the positive trend bias. The interesting aspect is that the age trend biases are largest at the youngest ages, whereas the correlations shown in Figure 4 have no discernable differences either seasonally or latitudinally at ages younger than 3-4 years. This would be worth discussing further.

Thank you for this comment, and we agree that this is an important point.

It is stated in the text already that "...the difference between the results from C-CORR and FULL methods stems from the differences in the AoA-FRF correlations used in each method, and the explicit FRF calculation..." Hence, strictly speaking, the differences shown in Fig. 7b and Fig. 7d are caused not only by the AoA-FRF correlations but also by the FRF calculation approach. Further, it is mentioned in Table 1 regarding the $2CH_{4[entry]}(r, t) \Delta \alpha(r, t)$ description that this term and $\Delta \alpha(r, t)$ is calculated differently in the case of FULL and C-CORR (e.g. in C-CORR the FRF trend is calculated as a residual). Those differences propagate to ΔAoA as well.

Taking all of that into account, the above mentioned differences in the young air cannot be understood in a simple way. However, the large differences in the lowermost stratosphere (directly above the tropopause) should not be over-interpreted, as in this region all methods have problems (because of the time resolution of the age-spectra). Even for the FULL method the transit time resolution of the used age spectra of 1 month causes issues in the lowermost stratosphere where transit times from the tropopause are within that range. A cautionary note is included in the revised manuscript (p16, L367). "The larger differences in the lowest stratosphere directly above the tropopause should not be over-interpreted as the transit time resolution of used age spectra of 1 month is too coarse for a reliable reconstruction there."

9. PAGE 18

Line 413, Appendix B: I don't really understand the partitioning of the age into constant values in seven zones. Why not just use the actual age at each location? Is it too computationally expensive?

We fully agree that using the actual age at each location would significantly improve the calculation. However, our main goal with the improved approximation method is to provide a simple and practical solution for estimating AoA trend from observations which does not require a complete AoA dataset from a model (as it might not be always available). We reformulated the statement in the revised version (p17, L397) accordingly. *"For a simple and practical method without assuming a priori knowledge of model age of air, we propose to divide the stratosphere into seven regions, prescribing one mean value of AoA for each region..."*

Line 429: Remove second 'method' in this line.

We decided to remove the entire sentence in the revised version.