

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 point-by-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).

"...CO₂ 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 H₂O considered in this work is CH₄ 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 CH₄ 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 H₂O 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 $2\text{CH}_4_{[\text{entry}]}(r, t)$ $\Delta\alpha(r, t)$ 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.

Reply to reviewer #2

Estimating Brewer-Dobson circulation trends from changes in stratospheric water vapour and methane

We thank the referee for the detailed review and for the helpful comments and suggestions. We give a point-by-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

This is an interesting and well written paper, but overly detailed. I recommend that the authors take some time to significantly reduce the content to the salient points, briefly summarizing the experiments and driving toward the main conclusions (which are a little nebulous). Concensing and consolidating the paper would improve the focus and make it more accessible to the reader.

We thank the reviewer for the encouraging comments and the advice for improvement. The suggestions have been taken into account in the revised version. As the text had already been consolidated amongst all authors in many iterations, a further shortening indeed proved to be a challenge. However, as we agree that a better focus would make the paper more accessible to readers, we have further worked on the text throughout the entire paper, improved formulations and wording and tried to shorten and focus – taking into account the reviewers comments.

Specific Comments to the Summary

1. “The basic idea, as I understand it, is that the authors want to use measurements of water vapor and methane to determine trends in the BDC. . . .No actual observations (except boundary forcing of methane and water) are used in this paper.”

Thank you for the comment.

The goal of the paper is to present a proof-of-concept study within an idealized model environment. We aim to give a simple and practical advice for obtaining more reliable AoA trends estimation from the observed H₂O and CH₄, in comparison to broadly used approaches. Therefore, we test several methods within the “model world” and state the conclusions on the possible improvements to the standard approximation. We agree that this idea was not clearly stated at the beginning of the paper. We rephrased the abstract in the revised version of the paper (p1, L8).

“In this work, we explore how mean age of air trends can be estimated from the combination of stratospheric H₂O and CH₄ data, by carrying out a proof-of-concept within the model environment of the Chemical Lagrangian Model of the Stratosphere (CLaMS).

In particular, we assess the methodological uncertainties related to the two commonly-used approximations of (i) instantaneous stratospheric entry mixing ratio propagation, and (ii) constant correlation between mean age and the fractional release factor of CH₄. Performing various sensitivity studies with CLaMS, we test different methods of the mean age of air trend estimation, and we aim to provide a simple and practical advice on the adjustment of the used approximations for obtaining more reliable mean age of air trend from the measurements of H₂O and CH₄.”

2. The authors assess various methods of using model water vapor and methane to determine changes in the BDC, or basically AoA trends and associated errors. I liked the evaluations they produce and an analysis of various errors (Fig. 5), but I think there is WAY too much detail, and the paper could use more of a reminder of the goals in the results section. For example, near the end perhaps you should show only 3 cases – True, Full and Improve Approx. Discussion of the other cases can be put in an Appendix since the average reader will give up while wading through this material. I think about 30% of this paper could be deleted with no loss of information content.

We agree that the paper includes many technical details. We have made many internal iterations, but since the subject is a comparison of methods where there is no generally accepted “standard method”, one cannot assume the reader will be able to understand what exactly is done without detailed instructions. So, it is important to provide enough details such that the results are reproducible by others. We took into account the above suggestions in the revised version and, overall, we reduced the text of the manuscript (please, see the difference .pdf file between the submitted and updated version of the manuscript).

3. It was interesting that if you assume a simple age spectrum (Eq. 7) rather than try and reconstruct it, the methodology might work (Fig. 8) pretty well. I look forward to the authors applying this technique to real data, and I wonder how observational uncertainty will impact the results given the size of the existing errors.

We agree that the application of the improved approximation method to real measurements seems promising. The present manuscript should be seen as a first step into that direction by assessing the potential of the methods within the “model world”. The application of the methods laid out in this paper to real observational data will be the focus of the future work.

4. As an aside, the authors mentioned a number of times that their analysis won’t work in the polar regions, yet they show these regions in the figures which is distracting. Perhaps cutting the figures at $\pm 50^\circ$ might be reasonable.

Thank you for this comment. We certainly discussed and considered this comment. Although the reconstruction methods does not work at SH polar regions (because of dehydration by sedimentation of ice crystals), we think it is important to keep these regions in the plots. On the one hand this makes the problem evident to all readers directly at first glance. Also, cutting figures only at the SH would lead to a non-centered equator,

what could potentially confuse readers, as there is little dehydration in the polar Arctic stratosphere in winter in the NH.