Reply to the reviewer #3
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. 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 paper is very well organised and written. The topic discussed here – quantifying the uncertainties in mean age of air (AoA) trend estimation from stratospheric water vapour and methane observations (for different assumptions) – is of high relevance... This work advertise the use of a practical method for obtaining more reliable AoA trends from H2O and CH4 observations. This is valuable... However, some open questions remain and some points should be clarified/changed/added. The paper should be submitted after addressing the comments below.

We thank the reviewer for this very encouraging comment and the suggestions for improvement. In the revised version, all suggested comments have been taken into account.

General comments

I highly appreciate the great effort and the way how the authors build up their argumentation, why it is reasonable and valuable using water vapour and methane observations to derive AoA trends... As outlined in my recommendation, it is still an open and pressing question to diagnose BDC changes and I totally agree in general with the conclusions of this paper. There are two general comments from my side.

1. Total hydrogen in the stratosphere is defined here as the sum of H_2O_{entry} + 2·CH_4. This definition is often used, but it might make sense to include also hydrogen (H_2) in this budget or at least shortly discuss the role of hydrogen for the stratospheric water vapour trend, especially in respect of a future hydrogen economy. Disregarding stratospheric moistening by increasing tropospheric hydrogen could lead to a misinterpretation of BDC trends in the future using only the conservation of total hydrogen as defined here.

   Thank you for this important comment.

There is already a discussion about H_2O sources in the introduction of the paper, and the limitations are mentioned with respect to CH_4 oxidation as the only considered source of stratospheric H_2O (p3, L76).

However, following the reviewer's suggestions, we expanded the discussion on the causes for H_2O changes in Sect. 2.3, see our main additions below. For further details, please refer to the difference .pdf file between the submitted and updated version of the manuscript.

- (p.6, L174) *"Changes in stratospheric H_2O are determined by the stratospheric H_2O entry mixing ratio through troposphere–stratosphere exchange (Fueglistaler and Haynes, 2005), and by chemical sources, mainly oxidation of CH_4 and molecular hydrogen (H_2) in the middle and high stratosphere (Dessler et al., 1994; Harries, 2015). H_2O in the troposphere is continuously supplied from the Earth's surface. CH_4 is largely emitted at the Earth's surface because of anaerobic reactions, and H_2 is originated from biomass burning and other natural sources; CH_4 and H_2 are transported from the troposphere into the stratosphere. Based on satellite and balloon observations, the sum of the principal components of the hydrogen budget (H_2O, $2\times CH_4$ and H_2) is constant with altitude over most of the stratosphere (e.g., Dessler et al., 1994)."*
- (p.6, L182) *"... assuming that H_2 production from CH_4 oxidation is balanced by H_2 oxidation, namely that the sum of H_2O and two times CH_4 mixing ratios is approximately constant..."*
- (p.7, L189) *"Note that the usage of the simple parameterization (see Eq. 4) for the ratio between oxidized CH_4 and produced H_2O has its limitations, e.g., a ratio of 2 overestimates the production of H_2O in the lower stratosphere and somewhat underestimates it in the upper stratosphere (Frank et al., 2018). It is questionable whether this parameterisation parameterisation can be used for future climate projections, when the BDC is expected to accelerate (e.g., Austin and Li, 2006; Li et al., 2008; Garcia and Randel, 2008), and, as a result, the transport of H_2 molecules becomes an important factor for the vertical profile of the H_2O in the stratosphere. We also note that an increase in tropospheric H_2 might gain importance in a future hydrogen economy (e.g., Vogel et al., 2012)."*

2. It is possible to deduce stratospheric circulation trends from FRF trends, but these FRF trends are generally a consequence of changing transit times (age spectra) and circulation patterns (pathways or path spectra). This means that unambiguous deduction of AoA trends from FRF trends is only valid under the assumption that only transit times (and not circulation patterns) change or that the chemical decay of a tracer is path-independent or that the changes in the circulation patterns compensate each other (for a specific non-inert tracer).
   The path-dependency of FRF trends is to my opinion no problem in this paper. The reasons are, that you implicitly account for it by using AoA-FRF correlations for all methods (non-stationary or stationary) and that your conclusions are not affected: 1.) AoA trends

from methane and water vapour are significantly affected by the assumed approximations and 2.) Using an idealised age spectra to calculate H_2O_{entry} and CH_{4entry} improves the APPROX method in respect of a more reliable AoA trend estimation.
However, it should be clarified that the reconstructed quantities ΔH2O and ΔAoA, as they are derived here, includes also possible changes in transport pathways and that disentangling the effect of changing transit times and transit pathways on the BDC is still an unsolved issue for diagnosing BDC changes from observations.

Thank you for this comment. This point was already partially addressed in the introduction of the previous paper version, moreover, some clarifications have been added there (p.3, L81).
"The strength of the chemical source of H_2O depends on transit path of air since entering the stratosphere and transit time, and is thus related to AoA, which in turn is a measure for only the transit time but not the transit pathways dependency. The full complexity of these processes is very challenging to represent in the analysis of stratospheric H_2O, in particular, it remains an issue to disentangle the effects of changing transit time and changing transport pathways when diagnosing trends of the BDC from observations."

3. It might be worth to think about changing the title of this very sound paper to a question... For my feeling, posing a question would better fit to the storyline of this paper and the answer, that the improved method to obtain more reliable AoA trends derived from water vapour and methane observation is currently one of the few promising ways to estimate BDC changes from observations, would strengthen the conclusion of this paper.

   Thank you for this suggestion. We formulated the title of the paper as a question. The current title is *"How can Brewer-Dobson circulation trends be estimated from changes in stratospheric water vapour and methane?"*

Specific comments

- L.70-73: *"They (Hegglin et al., 2014) showed that... are related to an accelerating shallow branch ... and to a deceleration of the deep branch of the BDC..."*
  For completeness this sentence should be extended by the following:
  "... as suggested by Engel et al. (2009) and shown by Bönisch et al. (2011) for the same period."

  Thank you for the remark. The suggested text was added in the revised version (p3, L70).
  "Bönisch et al. (2011) and Hegglin et al. (2014) showed that a decrease in the H_2O mixing ratios in the lower stratosphere, below about 10 hPa, and an increase in the H_2O mixing ratios above this level from the mid 1980s to 2010 are related to an accelerating shallow branch of the BDC (decreasing AoA below about 10 hPa) and to a decelerating deep branch of the BDC (increasing AoA above), as originally suggested by Engel et al. (2009)."

- L.79-81: *"The strength of the chemical source of H2O..."*
  It is true that methane (and hydrogen) oxidation is related to AoA, but AoA is only a measure for the transit time but not the transit pathway dependency (this is of particular interest, if you're a looking for changes in the BDC patterns). This differentiation should be added for clarity.

  Thank you for the specific comment. We have added the proposed statement to the text (p3, L81).
  "The strength of the chemical source of H_2O depends on the transit time and the transit path of air since entering the stratosphere and, thus, is related to AoA, which in turn is a measure for only the transit time but not the transit pathways dependency."

- L.92-93: *"Precisely, the source region covers the potential temperature layer from 10 K below to 10 K above the WMO (lapse rate) tropopause."*
  Is this criteria sufficient in the Subtropical Jet (STJ) regions with strong distortion of the tropopause and even double tropopauses? If not, does it matter for this study?

  We agree that the choice of the source region causes some uncertainty in the analysis as it does not exactly match the region defining the H_2O and CH_4 entry. We added the related sentence at the end of Sect. 2.2 (p6, L163).
  "As a remark, this specific choice of the source region causes the uncertainty in our analyses as it does not exactly correspond to the region defining the H_2O and CH_4 entry mixing ratios; the tropically controlled transition region bounds between approximately 380 K and 450 K (Rosenlof et al., 1997; Li et al., 2012). However, this mismatch impacts the results only close to the tropopause, so the reconstruction of H_2O and CH_4 by the modelled age spectrum ensures the reliability of the method in most of the stratosphere."

- L168-170: *"Trends in AoA... by using the conservation property of total hydrogen in the stratosphere, namely that the sum of H2O and two times CH4 mixing ratios..."*
  How about hydrogen (see general comments)?

  Thank you for the comment. We have added the statement about hydrogen in the revised version (p6, L182).
  "Trends in AoA can be calculated from trends in stratospheric H_2O mixing ratios by using the conservation property of total hydrogen in the stratosphere and assuming that H_2 production from CH_4 oxidation is balanced by H_2 oxidation, namely that the sum of H_2O and two times CH_4 mixing ratios is approximately constant..."

- L.185-186: *"The FRF is strongly affected by the vertical transport of the BDC. Hence, information on circulation trends (in particular on AoA) can be deduced from trends in FRF (Hegglin et al., 2014)."*
  This is generally only valid for AoA trends under the assumption that only transit times and not circulation patterns change or that the chemical decay of a tracer is path-independent or if the changes in the circulation patterns compensate each other (see general comment 2.)).

We inserted the required clarification to the text (p7, L207).
"It should be noted that the change in FRF is due to transit times (age spectra) and circulation pathways (path spectra) changes, but AoA is a measure for only transit times and not the transit pathways dependency."

- L.189, EQ (6): It might be good to point out here, that the first two terms in the equation dependent only on changes in transit times and the third term (including $\Delta\alpha$ or ΔFRF) depends also on changing transit pathways (see general comment 2.)).

  Thank you for the remark, we added the mentioned clarification to the text (p7, L214).
  "Note that $\Delta \mathrm{H_2O}_{[\mathrm{entry}]}(r,\ t)$ and $\Delta \mathrm{CH_4}_{[\mathrm{entry}]}(r,\ t)$ depend only on changes in transit times, while $\Delta\alpha(r,\ t)$ generally also depends on the changes in transport pathways."

- L.196: *"... and can be converted to an AoA trend."*
  See again general comment 2.).

  We inserted an additional sentence to the revised version of the paper (p8, L221).
  "Besides, the dependency of FRF changes on circulation pathways is implicitly taken into account by the AoA-FRF correlation functions in all used methods (Sect. 2.4).

- L232-234: *"The location of entry to the stratosphere is approximated as the 390-400K layer between 30°S-30°N, which is located just above the cold point tropopause."*
  Could this have an impact on your results, because the distance in potential temperature between the 390-400K level and the thermal (or dynamical) tropopause could be large, especially in the winter hemisphere at the edge of the defined entry region (>25°) (see also comment for L.92-93 above)?

  Thank you for this question. We added the explanation to the revised version of the paper (p10, L261).
  "The small difference between the age spectrum source region (tropopause ± 10 K) and the trace gases entry region has only a negligible impact on our results from above $\approx$420 K due to the small difference in the transit time between the two regions."

- L.259-261: *"Outside of the Southern high-latitude regions, the overall differences shown in Fig. 3c are small ..."*
  I would add here: *"Outside of the Southern high-latitude regions and in the proximity of the extratropical tropopause (>30°N/S), the overall differences..."*
  The reason for this is that direct in-mixing into the LMS not via the defined entry layer occurs and that especially water vapour mixing ratios at the extratropical tropopause are rather different from the mixing ratios at the tropical tropopause.

  Thank you for the remark. The suggested text was added in the revised version (p11, L287).
  "Outside of the Southern high-latitude regions and in the proximity of the extratropical tropopause (>30°N/S), the overall differences shown in..."

- L.279-280: *"And, for instance, at the same FRF level of 0.3, the air at the Northern tropics (30°N-40°N) is younger than at the Southern tropics (30°S-40°S) by almost half a*

year."
Is there a (simple) explanation for this? This is an interesting finding and maybe it is worth to discuss it (or speculate about it).

This is likely related to the hemispheric asymmetry in the BDC, which is stronger in the NH during boreal winter; the figure you are referring to is shown for January, 2000. We have added clarifications to the text (p12, L308).
"It is likely due to stronger and deeper BDC in the Northern hemisphere during boreal winter (e.g., Rosenlof, 1995; Butchart, 2014) causing air parcels of the same age to travel deeper pathways through the stratosphere and experience more chemical depletion compared to the Southern hemisphere."

- L.294: *"...(e.g., Schoeberl et al., 2000, 2005; Ehhalt et al., 2007; Hegglin et al., 2014)."*
Would you please add Fritsch et al. (2020) here, because this is in my opinion also a highly relevant work on the topic – limitations (and improvements) for deriving AoA from real-world age tracers.

  The suggested reference was added in the revised version (p14, L325).
  "... about the age spectrum and its shape (e.g., Schoeberl et al., 2000, 2005; Ehhalt et al., 2007; Hegglin et al., 2014; Fritsch et al., 2020)."

- L.335: *"Thus, the accuracy of the estimated AoA changes from APPROX largely depends on the considered period."*
It is true that AoA trend depends on the period, but it is likely that the main criteria is that the period has to be long enough to cover the internal variability. If the period is too short there will be random results for different sub-periods.

  Thank you for the remark. We rephrased the sentence as suggested (p15, L365).
  "Thus, the accuracy of the estimated AoA changes from APPROX largely depends on the considered period, which should be long enough to ensure that the effects of variability is small."

- L.357-358: *"Hence, the good performance of the FULL method can be related to the fact that stratospheric entry H2O mixing ratios are not influencing the calculation."*
To avoid misunderstandings, I would add here: *"... provided that some regions are excluded (as explained in section 3.1)."*

  Thank you for the remark. We have added the suggested text in the revised version of the paper (p16, L387).
  "Hence, the good performance of the FULL method can be related to the fact that stratospheric entry H_2O mixing ratios are not influencing the calculation, provided that the polar regions are excluded (as explained in Sec. 3.1)."