

Reviewer (Comments):

Review of "Estimating Brewer-Dobson circulation trends from changes in stratospheric water vapour and methane" by Liubov Poshyvailo-Strube et al.

Recommendation: Publication after minor revision

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. AoA is maybe still the most important proxy for estimating Brewer-Dobson circulation (BDC) trends from observations. Understanding BDC changes is still of high interest, because the BDC controls the distribution of trace gases (and also aerosols to some extent) in the stratosphere and therefore affects radiation (especially by changes of water vapour and ozone in the tropopause region). How BDC changes under changing climate conditions and how BDC changes impacts climate (the feedback mechanisms) are still not well known yet. This work advertises the use of a practical method for obtaining more reliable AoA trends from H₂O and CH₄ observations. This is valuable for two reasons. First, long time series from satellites observations exist (and will be continued hopefully) for both tracers and second, the uncertainties and limitations of the method(s) are very thoroughly analysed here – a general prerequisite for trend analysis from observations.

However, some open questions remain and some points should be clarified/changed/added. The paper should be submitted after addressing the comments below.

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. The argumentation based on the same general approach that Fritsch et al. (2020) used for analysing the sensitivity of AoA trends to the derivation method for non-linear increasing inert SF₆ – carrying out proof-of-concept of the method(s) applied to observations within the closed (or self-consistent) “model world” of CLaMS or EMAC, respectively.

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 H₂O_{entry} + 2•CH₄. This definition is often used, but it might make sense to include also hydrogen (H₂) 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.

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 $H_2O|_{\text{entry}}$ and $CH_4|_{\text{entry}}$ improves the APPROX method in respect of a more reliable AoA trend estimation.

However, it should be clarified that the reconstructed quantities ΔH_2O 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.

Finally a suggestion:

It might be worth to think about changing the title of this very sound paper to a question: “Estimating Brewer-Dobson circulation trends from changes in stratospheric water vapour and methane?”. To my point of view, the answer is yes, but one have to take into account the uncertainties and limitations that you elaborated in this work.

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.

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.”

L.79-81: *”The strength of the chemical source of H_2O ...”*

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.

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?

L168-170: *”Trends in AoA... by using the conservation property of total hydrogen in the stratosphere, namely that the sum of H_2O and two times CH_4 mixing ratios...”*

How about hydrogen (see general comments)?

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.)).

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.).

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

L232-234: “*The location of entry to the stratosphere is approximated as the 390-400 K 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 ($>25^\circ$) (see also comment for L.92-93 above)?

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 ($>30^\circ\text{N/S}$), 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.

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).

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.

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.

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).”

References:

Bönisch, H., Engel, A., Birner, T., Hoor, P., Tarasick, D. W., and Ray, E. A.: On the structural changes in the Brewer-Dobson circulation after 2000, Atmos. Chem. Phys., 11, 3937-3948, 10.5194/acp-11-3937-2011, 2011.