

General Remarks

The author has considered most of my concerns in the revised version of the paper. The most substantial new information is to my opinion Figure 6, which provides the overall picture of the CH₄ trends in the middle atmosphere and its statistical significance. However, I think that the interpretation of this picture is still not state-of-the art in the revised version.

Diallo et al. (2012), and Monge-Sanz et al. (2012) also, found a decrease of mean age of air (AoA) in the lower branch and an increase of AoA in the deep branch of the BDC for the ERA-Interim time period. This confirms earlier findings from papers by Engel et al. (2009) and Bönisch et al. (2011) both using in-situ tracer observations to diagnose BDC changes. Stiller et al. (2012) uses satellite (MIPAS) observations of SF₆ to derive AoA trends for the time period 2002 to 2010. The MIPAS satellite data analysis has improved significantly since then and the AoA trend of the 2012 paper must be regarded as too patchy (Stiller, pers. communication). The excellent paper of Ploeger et al. (2015) that is highlighting the fact that „AoA (and long-lived tracer, note of the reviewer) changes are the result of a delicate local balance between the competing effects of residual circulation and mixing”, provides a more actual MIPAS SF₆ based AoA trend. Generally, the findings of this paper also corroborates the hypothesis of a strengthening of the lower BDC branch at first introduced by Bönisch et al. (2011).

In addition to the revised paper, the author came up later on with further material: A comment concerning the interpretation of the overall CH₄ trends. A major point in this comment is that “the distribution of CH₄ trends in Fig. 6 indicates a likely contribution from the effects of horizontal transport and mixing across latitudes”. This is to my opinion the most obvious and the statistical most robust feature in the analysed CH₄ trends and fits into the picture drawn by the above mentioned papers, all based on observations and/or meteorological reanalysis datasets.

However, the argumentation of the author in his additional comment is not really clear to me. He states that the regions in the stratosphere, where CH₄ trends are significantly larger than in the troposphere, correspond to the regions, where AoA show increasing trends and vice versa. This is to my opinion the wrong way round or at least not trivial. In the lower midlatitude stratosphere, where one can see in Figure 6 highly significant CH₄ trends of about 4-8% per decade (especially in the Northern Hemisphere), Bönisch et al. (2011), Diallo et al. (2012), Monge-Sanz et al. (2012) as well as Ploeger et al. (2015) all diagnosed decreasing, not increasing, AoA due to a possible increase of the shallow branch of the BDC.

Consequently, the argumentation in the other way is to my opinion problematic too. It is not straight forward to associate older AoA (positive AoA trends) in the midlatitude middle stratosphere (25-55 degN/S and 10 to 1 hPa) with increasing CH₄. A weakening of the subtropical barrier, as argued by the author, would most likely lead to an increase of CH₄ in the midlatitudes, but it is not self-evident what would happen to AoA (see Garny et al., 2014 and Ploeger et al., 2015). Intuitionally, one would expect a decrease of AoA, but both papers mentioned above show, that recirculation, depending on the balance between residual transport and bidirectional mixing, might lead to “aging by mixing” in the midlatitudes. Furthermore, the breakup of the CH₄-AoA correlations in the upper stratosphere and mesosphere (photochemical lifetime becomes short compared to transport timescales (Plumb and Ko, 1992)) imply that changes in the strength of the subtropical transport barrier in the middle stratosphere might have different effects on temporal evolution of AoA and CH₄.

To summarise the discussion: The paper is in a state that it can and should be published in ACP. I recommend to add the additional comment, concerning mainly the discussion and interpretation of the overall CH₄ trends, which has unusually been submitted by the author directly to the editor, even though I disagree with the conclusion drawn here. However, it is to my personal point of view neither the duty nor the right of the reviewer to dictate the conclusions that should be drawn from scientifically generated facts, i.e. observations. Nevertheless, it would be preferable and good scientific practice, if the author officially submits a finalised version of the section about the interpretation of the CH₄ trends. Maybe, some of the discussion above will change the conclusions drawn here. This new added section should be at least presented to the editor before publication.

References

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