Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-95-RC2, 2020

Atmospheric Chemistry and Physics
Discussions

Interactive comment on “Gravitational separation of Ar/N_2 and age of air in the lowermost stratosphere in airborne observations and a chemical transport model” *by* Benjamin Birner et al.

Anonymous Referee #2

Received and published: 27 March 2020

This paper uses measured and simulated ratios of Ar/N2 to deduce gravitational separation in the stratosphere and their relationship to the stratospheric age of air. The Ar/N2 relationships to AoA in this study are relatively compact compared to the previous Belikov et al. study on gravitational separation and the agreement between the observations and model relationships are improved. This suggests that gravitational separation of Ar/N2, and also potentially the ratios of heavier noble gases, could be useful as an additional diagnostic of transport. The measurements, model runs and methodology are well described and the improvement in the treatment of gravitational

separation in both the measurements and model compared to previous studies is an important step forward.

My main concern with this study is in the calculation of age of air from the measurements. The technique used, although rigorous, is based on assumptions that are not consistent with our current understanding of the shape of age spectra and the transport of air into the lowermost stratosphere. The technique and assumptions are based on a study that is nearly two decades old and there have been numerous studies since that time refining our understanding of age spectra and how they can be inferred from measurements. The results may not change substantially by using more realistic age spectra and transport assumptions but that isn't entirely clear. Regardless, the more recent studies on this topic and the newer techniques should be acknowledged and shown to be in agreement with, or ideally replace, the results from the technique used in this study. Specific references and more detail on this topic are included below.

The results of this study are significant and relevant to ACP so I recommend publication after modification of the age of air calculation and consideration of the specific comments below.

Specific comments:

Line 35: should add ‘however’ before ‘observational’

Lines 40-41: should add references here to studies that have used tracers to infer age spectra (Andrews et al., JGR, 1999; Schoeberl et al., JGR, 2005; Hauck et al., ACP, 2019; Podglajen and Ploeger, ACP, 2019).

Line 94: did you mean ‘CO2’ instead of ‘CO’ or should ‘CO2’ be added here?

Line 97: ‘sample flask i to sample flask i+1’

Line 108: change ‘as a combined result of’ to ‘due to’

Line 113: ‘used’ instead of ‘use’

Section 2.2: This section is my main concern with the paper as I mentioned above. I have two issues: (1) the assumed shape of the age spectra and (2) the tropical single-entry point assumption for the lowermost stratosphere. The first issue, the shape of stratospheric age spectra has been shown in a number of studies derived from various model and reanalysis output based on trajectories or boundary impulse functions (Reithmeier et al., Clim. Dyn., 2008; Li et al., JGR, 2012; Diallo et al., ACP, 2012; Ray et al., JGR, 2014; Ploeger and Birner, ACP, 2016; Podglajen and Ploeger, ACP, 2019; Hauck et al., ACP, 2019). In these studies, it is clear that the dominant mode of variability in the age spectra in the lower stratosphere is the seasonal cycle, and for those models with a QBO, that is the second largest mode of variability. None of these results show a peak in the 5-7 year range relative to the 2-5 year range of the spectra. The use of two peak age spectra with one of the peaks in the 5-7 year range is not realistic for any part of the stratosphere and especially not in the lower stratosphere where the seasonal cycle is so large and the 5-7 year portion of the age spectra has much smaller values (often by an order of magnitude) compared to those at ages less than 5 years. The lack of reference in the manuscript to any of the papers listed above and the use of an age spectra shape that is not consistent with any of them is a clear deficiency.

The use of a single trace gas, such as CO2, can only reveal a limited amount of the age spectra. It has been shown by Hauck et al. (2019) that at least 5-10 tracers with a range of lifetimes are necessary to resolve the important features and the true shape of the age spectra. The Andrews et al. (1999, 2001) studies were important steps at the time in our understanding of how trace gas observations could be used to infer features of the age spectra. But now that we have a detailed understanding of age spectra from models and reanalysis it isn't appropriate to use age spectra defined only by the fit to CO2.

The second issue is related to the transport of air into the lowermost stratosphere (LMS) where most of the measurements in this study were taken. Many studies have shown that the LMS consists of a seasonally varying mixture of tropospheric and strato-

spheric air with some portion of the tropospheric air originating from the extratropics or tropical air that has bypassed the tropical tropopause (e.g. Ray et al., JGR, 1999; Hoor et al., ACP, 2004; Olsen et al., JGR, 2004; Boenisch et al., ACP, 2009; Skerlak et al., ACP, 2014). This precludes the use of a single-entry tropical tropopause time series to derive age of air in the LMS.

I highly recommend a technique considering the above points be used here. Even if the resulting mean age values do not differ significantly, the technique would be consistent with our current understanding of transport in the lower stratosphere.

Line 152: Gamma1 is repeated, the second one should be Gamma2.

Line 179: the second '(ii)' should be '(iii)'

Line 203: 'relationships'

Line 262: 'descends' instead of 'is sinking'

Line 328: Is the Ar/N2 ratio 'mostly unaffected by seasonality' throughout the stratosphere?

Lines 330-331: It's not so much that changes in AoA in the upper stratosphere and mesosphere are difficult to resolve from transient tracers but more that AoA becomes nearly constant in this region as you show in Figure 6. So AoA loses sensitivity in this region where gravitational separation gains sensitivity. This is a difficult region to make measurements of any kind so it may also be challenging to obtain enough measurements of noble gases to monitor gravitational separation.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-95, 2020.