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Key Points:

- Reconcile long-term mean age decrease and observed decadal variability
- Mixing effect is crucial to explain mean age trends
- Mixing effect on mean age is partly coupled to the residual circulation

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Quantifying the effects of mixing and residual circulation on trends of stratospheric mean age of air

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Abstract It is an outstanding issue to what degree trends in stratospheric mean age of air reflect changes in the (slow) residual circulation and how they are affected by (fast) eddy mixing. We present a method to quantify the effects of mixing and residual circulation on mean age trends, based on simulations with the Chemical Lagrangian Model of the Stratosphere (CLaMS) driven by ERA-Interim reanalysis and on the integrated tracer continuity equation. During 1990–2013, mean age decreases throughout most of the stratosphere, qualitatively consistent with results based on climate model simulations. During 2002–2012, age changes show a clear hemispheric asymmetry in agreement with satellite observations. We find that changes in the residual circulation transit time cannot explain the mean age trends, and including the integrated effect of mixing is crucial. Above about 550 K (about 22 km), trends in the mixing effect on mean age appear to be coupled to residual circulation changes.

1. Introduction

The chemical composition of the lower stratosphere is strongly affected by the Brewer-Dobson transport circulation [*Dobson et al.*, 1929; *Brewer*, 1949] and has been shown to be highly relevant for surface climate [e.g., *Shine and Forster*, 1999; *Riese et al.*, 2012]. However, no consensus has been reached regarding changes of the Brewer-Dobson circulation, which may be considered to consist of a residual mean mass circulation and additional two-way eddy mixing. Current general circulation models consistently predict a strengthening of the circulation, deduced from an increasing tropical upward mass flux [e.g., *Butchart et al.*, 2010], related to greenhouse gas-induced climate change [*Rind et al.*, 1990]. Although, the simulated trends may depend on the specific model configuration [*Bunzel and Schmidt*, 2013]. In terms of mean age of air, the average transit time of an air parcel since entering the stratosphere [*Hall and Plumb*, 1994; *Waugh and Hall*, 2002], the models predict a related negative trend throughout the stratosphere [e.g., *Olsen et al.*, 2007; *Garcia and Randel*, 2008; *Garcia et al.*, 2011; *Butchart et al.*, 2010]. This negative mean age trend has been shown to correlate with the increasing tropical mass flux [*Austin and Li*, 2006] and is commonly interpreted as a decreasing mean transit time due to an accelerating circulation.

However, observationally based mean age estimates in the Northern Hemisphere subtropics and midlatitudes between about 24 and 35 km show no indication for a decrease of mean age between 1975 and 2005 [*Engel et al.*, 2009] and hence no indication for an accelerating circulation. Furthermore, satellite observations by the MIPAS instrument (Michelson Interferometer for Passive Atmospheric Sounding) show an inhomogeneous pattern of mean age trends from 2002 to 2012, with mainly decreasing age in the Southern Hemisphere (SH) and increasing age in the Northern Hemisphere (NH), respectively [*Stiller et al.*, 2012]. Recently, *Diallo et al.* [2012] and *Monge-Sanz et al.* [2012] showed that mean age simulations based on European Centre for Medium-Range Weather Forecasts ERA-Interim reanalysis data [*Dee et al.*, 2011] show some consistency with observed mean age trend patterns. It should be noted that the use of reanalysis data may introduce artifacts to trend estimates, related to inhomogeneities in data assimilation and spurious transport [e.g., *Schoeberl et al.*, 2003; *Tan et al.*, 2004]. However, biases in the heat and momentum budget as well as spurious dispersive transport appear to be significantly reduced for ERA-Interim compared to previous reanalysis [*Fueglistaler et al.*, 2009; *Monge-Sanz et al.*, 2012].

Possible explanations for the discrepancy between observed mean age trends and trends simulated by general circulation models involve the sparse sampling of in situ observations [*Garcia et al.*, 2011], as well as differences in the changes between deep and shallow Brewer-Dobson circulation branches [*Birner and*

Bönisch, 2011; *Bönisch et al.*, 2011]. Based on conceptual one-dimensional models, a strong sensitivity of mean age to atmospheric mixing processes has been found [*Neu and Plumb*, 1999; *Ray et al.*, 2010]. Recently, *Garny et al.* [2014] argued based on an indirect estimate that the mixing effect is largely coupled to the residual circulation.

Here we present a direct calculation of the effect of mixing on trends of mean age of air. Based on chemistry transport model simulations and on the tracer continuity equation, we separate the effects of the residual circulation and of eddy mixing (in the following simply "mixing") and show that mixing plays a crucial role in determining the mean age trends.

2. Method

Mean age of air has been simulated with the Lagrangian chemistry transport model CLaMS (Chemical Lagrangian Model of the Stratosphere) [see *McKenna et al.*, 2002; *Konopka et al.*, 2004], driven by ERA-Interim winds for the period 1990–2013. Above about 300 hPa, vertical transport in the model is based on isentropic coordinates and is driven by ERA-Interim total diabatic heating rates. Note that ERA-Interim includes evolving greenhouse gas concentrations, set to observed 1990 values plus a linear trend [see *Dee et al.*, 2011]. Model mean age is calculated from a "clock tracer," an inert tracer with a linearly increasing source in the lowest model layer (for details about the CLaMS simulation see *Ploeger et al.* [2014] and *Pommrich et al.* [2014]).

Using the property of mean age as the time lag between local and tropospheric clock-tracer mixing ratios [*Hall and Plumb*, 1994], the isentropic zonal mean tracer continuity equation [e.g., *Andrews et al.*, 1987, equation (9.4.21)] can be formulated for mean age of air [e.g., *Plumb*, 2002]. This equation can be formally solved by integration along its characteristics which correspond to the residual circulation trajectories (the hypothetical paths of air parcels if they were only advected by the residual circulation). The resulting equation for mean age Γ at location *x* and time *t* is [e.g., *Ploeger et al.*, 2014]

$$\overline{\Gamma}(x,t) = \tau_{\text{RCTT}}(x,t) + \int_{t_0}^t \mathcal{M}(x,t') \,\mathrm{d}t' \,. \tag{1}$$

Here τ_{RCTT} is the residual circulation transit time (RCTT) for transport from the tropical tropopause (crossed at time t_0) and \mathcal{M} is the local mixing tendency for mean age, including the local effects of eddy mixing, given by

$$\mathcal{M} = \frac{1}{\overline{\sigma}} \nabla \cdot \mathcal{M} - \frac{1}{\overline{\sigma}} \partial_t (\overline{\sigma' \Gamma'}) \,. \tag{2}$$

Our notation follows Andrews et al. [1987], with overlines denoting zonal means and primed quantities fluctuations therefrom, $\sigma = -g^{-1}\partial_{\theta}p$ is the density in isentropic coordinates, p pressure, and g acceleration due to gravity. The components of the eddy flux vector are defined via $M_{\phi} = -(\sigma v)'\Gamma'$ and $M_{\theta} = -(\sigma Q)'\Gamma'$, with θ the potential temperature, v the meridional velocity, and Q the diabatic heating rate. Note that the transient (second) term on the right-hand side of equation (2) is negligible for all results shown here (for instance, calculation of the mixing effect on AoA (Figure 1d) including the transient term causes relative differences well below 1%, except at polar latitudes). Equation (1) provides a separation of the mean age value at each location and time into the residual circulation transit time τ_{RCTT} , which describes the pure effect of the residual circulation on mean age and an additional contribution by mixing. Note further that in the isentropic formulation used here "residual circulation" denotes the diabatic (mass-weighted) zonal mean circulation ($\overline{v}^*, \overline{Q}^*$), with $\overline{v}^* = (\overline{\sigma v})/\overline{\sigma}$ and $\overline{Q}^* = (\overline{\sigma Q})/\overline{\sigma}$.

To apply equation (1), we calculate backward trajectories advected by daily mean ERA-Interim residual circulation wind velocities, starting on the 15th of each month during 1990–2013 on a regular latitude/ θ grid, until they cross the tropical tropopause (here 380 K isentrope, 20°S–20°N). Figure 1a exemplarily shows the backward trajectories starting at 400 K from tropics to polar latitudes on 15 July 2012 (initialized with a 4° latitude spacing). The back trajectories are color coded by their RCTT, the transit time since crossing 380 K in the tropics (20°S–20°N). The integrated mixing contribution is calculated by integrating the daily local mixing tendencies numerically along the trajectories (background shading in Figure 1a shows the climatological local mixing tendency). For that purpose, the daily local mixing tendencies have been



Figure 1. (a) Residual circulation back trajectories, starting at 400 K (4° latitude spacing) on 15 July 2012, with transit time (RCTT) from the tropical tropopause (black dashed, 380 K) color coded. Background shading shows climatological local eddy mixing tendencies for mean age (blue: negative and orange-red: positive). (b) Mean age for 1990–2013 from CLaMS, (c) reconstructed as sum of RCTT and integrated mixing effect aging by mixing, (d) aging by mixing, and (e) RCTT (white areas with gray contour shading indicate negative values). White arrows in Figure 1d show mixing flux of mean age, in Figure 1e residual circulation velocities, and dashed lines show altitude levels in kilometers. (f) Time series of mean age (black), reconstructed from RCTT and mixing (gray), aging by mixing (red), and RCTT (blue), at 32°S/500 K. Linear trends in years per decade are given in the legend (the uncertainty is the standard deviation of the regression).

calculated first from the model mean age distribution, using equation (2). As shown in Figure 1a, mixing locally increases (orange and red shading) mean age at low latitudes equatorward of about 40°N/S and decreases (blue shading) mean age at higher latitudes, because mixing exchanges young air from low latitudes with old air from high latitudes. This fact is consistent with the mainly horizontal nature of isentropic eddy mixing. To compare the mean age reconstructed as the sum of residual circulation and mixing with the simulated mean age based on the boundary layer clock tracer, we add the (time-dependent) mean age value at the tropical tropopause to the RCTT, for the rest of the paper.

As recently proposed by *Garny et al.* [2014], the mixing effect on mean age might be calculated as the residual between mean age and RCTT. This estimate is easier to deduce than the exact calculation presented here but may include additional effects due to unresolved processes and numerics [see *Garny et al.*, 2014]. For trend analyses it needs to be shown that these additional effects are small.

3. Results

Figures 1b and 1c confirm that the CLaMS-simulated mean age and the reconstruction of mean age by adding the RCTT and the integrated mixing effect agree very well, providing confidence in the method. Only at high latitudes the reconstructed age appears to be weakly biased old. As already discussed by Birner and Bönisch [2011], the RCTT largely differs from the mean age (compare Figures 1b and 1e). In most regions the RCTT is younger than the mean age. Only in the high-latitude lower stratosphere below about 20 km (500 K) the RCTT is older than the mean age. Hence, mixing is crucial for establishing the observed mean age distribution, causing a recirculation of air into the tropics and an additional aging throughout most of the stratosphere, termed "aging by mixing" by Garny et al. [2014]. Largest aging by mixing occurs in the subtropics around 24 km (Figure 1d). The negative aging by mixing in the polar lower stratosphere results from the particular transport history of the air parcels in this region, reaching high into the stratosphere and sampling mainly regions of negative local mixing tendency at high latitudes while avoiding regions of strong positive mixing tendencies in the subtropics (see Figure 1a). As aging by mixing results from integration of the local mixing tendency along the residual circulation (mapping the integrated mixing effect to the location where mean age is calculated), its distribution (Figure 1d) differs significantly from that of the local mixing tendency (Figure 1a) and from the related mixing flux (for a similar discussion regarding ozone, see Abalos et al. [2013]). The information where the mixing has occurred is included in the eddy mixing flux of mean age (white arrows in Figure 1d), which is strongest (and equatorward) in the lower stratosphere below about 500 K, below the region of largest aging by mixing. This recirculation in the lowest part of the stratosphere affects mean age globally (for details about the mixing seasonality, see Konopka [2015]).

We consider an exemplary SH subtropical region (32°S, 500 K), representative for the most significant simulated 1990–2013 mean age changes (as shown below). Comparison of mean age reconstructed from RCTT and mixing using equation (1) with the model mean age shows that the mean age budget of equation (1) is very well closed even locally at a single grid point (compare gray and black lines in Figure 1f). A similarly good closed budget holds globally, and slightly larger imbalances occur only near the tropopause. In the SH subtropical stratosphere mean age significantly decreases from 1990 to 2013, with a linear trend of -0.36 ± 0.04 yr/decade (uncertainty denotes 2σ standard deviation of the linear regression). The RCTT shows only a very weak trend, and the mean age trend appears to be clearly linked to the effect of mixing. Specifically, aging by mixing causes about 90% of the mean age trend in this region.

Figure 2 presents the global view of the mean age trends from 1990 to 2013, calculated as linear trends after deseasonalizing the monthly time series. Most evident is a mean age decrease, strongest in the SH (Figure 2a). Reconstruction of the trend from RCTT and aging by mixing yields a globally consistent result (Figure 2b). In view of the complex calculation (integration of the mixing tendencies over several years) we consider the agreement between the simulated and reconstructed mean age trends reliable (Figures 2a and 2b), with differences potentially related to missing processes in the mean age budget of equation (1) (e.g., parameterized CLaMS small-scale mixing). Clearly, the trend pattern of mean age is largely caused by the effect of mixing (Figure 2c), whereas changes in the RCTT alone produce a small reduction of mean age throughout the stratosphere (Figure 2d). Mean age decadal changes during 2002–2012 corroborate this result of a large effect of aging by mixing trends on mean age trends (Figure 3).



Figure 2. Trends for 1990–2013 of (a) mean age of air, (b) reconstructed as sum of residual circulation and mixing effects, (c) trend of aging by mixing, and (d) trend of RCTT. Gray shading shows regions where trends are not significant at the 2σ level (σ the standard deviation of the linear regression). Dashed lines show altitude levels in kilometers.



Figure 3. Same as Figure 2 but for the 2002–2012 period.

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Figure 4. (a) Local mixing tendency trend for 1990–2013 (background color shading), together with residual circulation back trajectories starting in the NH and SH subtropics on 15 January of each year (color coded with the respective year, starting locations as crosses). Black contours show climatological local mixing tendencies (solid positive, dashed negative, compare Figure 1). (b) Eddy mixing flux of mean age ($M_{\phi} = -\overline{\sigma}^{-1}(\sigma v)'\Gamma'$) from the SH into the tropics across 30°S (black solid, equatorward flux positive) and its linear trend (red dashed). (The vertical black dashed line in Figure 4a illustrates 30°S.)

4. Discussion and Conclusions

Mean age trends for 1990–2013 simulated by CLaMS driven by ERA-Interim winds are mainly negative throughout the lower stratosphere (Figure 2), in qualitative agreement with the climate model simulated response to increasing amounts of greenhouse gases [e.g., *Butchart et al.*, 2010]. *Engel et al.* [2009] have found no indication for decreasing mean age in the NH subtropics and midlatitudes above about 24 km. The CLaMS-simulated mean age trends confirm this result—we find insignificant (or weakly positive) trend values in this region. A recently published analysis by *Ray et al.* [2014], based on the in situ mean age data set of *Engel et al.* [2009] in combination with model simulations, shows negative trends in the NH below about 24 km, again in good agreement with the global CLaMS results (Figure 2). We showed that the effect of mixing integrated over the air parcel history is crucial to explain the simulated mean age trends.

Furthermore, CLaMS-simulated mean age changes during the last decade (2002–2012) significantly differ from the 1990–2013 trends (compare Figures 2 and 3), reflecting strong decadal variability, in reasonable agreement with satellite observations from MIPAS [*Ploeger et al.*, 2014], and also with decadal changes in HCl mixing ratios [*Mahieu et al.*, 2014]. As shown in Figure 3, the strong hemispheric asymmetry with decreasing age in the SH and increasing age in the NH between 2002 and 2012 is clearly reflected in the integrated effect of mixing. Only below about 20 km the net mean age decrease coincides with decreasing



Figure 5. Trend in aging by mixing for (a) 1990–2013 and (b) 2002–2012 resulting from changes in the residual circulation alone. This sensitivity integration has been carried out with constant local mixing tendencies (climatology over 1990–2013), and resulting trends are coupled to changes in the residual circulation. Dashed lines show altitude levels in kilometers.

RCTT, indicating an acceleration of the residual circulation in the lowest part of the stratosphere during 2002–2012, in agreement with *Aschmann et al.* [2014]. Remarkably, for the 1990–2013 period the residual circulation shows acceleration (negative RCTT trend) extending to higher altitudes (up to ~26 km, Figure 2d), compared to the 2002–2012 period.

As discussed by *Garny et al.* [2014], trends in aging by mixing may result both from trends in the local mixing intensity and from trends in the residual circulation. In the latter case, the RCTT may change, causing changes in the time exposed to mixing, and the residual circulation structure may change, such that the sampling of mixing regions (by air parcel trajectories) changes over time. In the following, we present evidence that the CLaMS aging by mixing trend arises from a combination of these different effects.

First, the local mixing tendencies *M* show a clear trend pattern, mainly below about 550 K (Figure 4a). Exemplary back trajectories from the SH subtropical lower stratosphere (launched at 32°S/500 K on 15 January of each year, Figure 4a) illustrate that the pathway to this region is not changing over time. Therefore, the significant negative aging by mixing trends in the SH for 1990–2013 (compare Figure 2c) are related to air masses sampling regions of significantly decreasing local mixing tendencies in the SH subtropics below about 500 K (compare trajectories in Figure 4a). In the climatology, the subtropical lower stratosphere is characterized by recirculation of air into the tropics, indicated by a strong equatorward eddy mixing flux of mean age (black line in Figure 4b). This eddy mixing flux in the SH weakens over time (red dashed). Hence, the decreasing local mixing tendencies in the SH subtropics (Figure 4a) are related to a weakened recirculation in the SH (Figure 4b). In the NH, the situation is more complex with back trajectories showing significant spread over time (Figure 4a).

Furthermore, we carried out a sensitivity integration with constant local mixing tendencies (fixed to 1990–2013 climatological values). This sensitivity calculation shows that above about 550 K trends in aging by mixing (Figure 2c) can be largely explained without taking trends in local mixing tendencies into account (compare, e.g., Figure 5a to Figure 2c). Below about 550 K, on the contrary, trends in local mixing tendencies are essential for the reconstruction of the aging by mixing trend. These results are consistent with the findings of *Garny et al.* [2014] that the effect of the local mixing intensity is strongest in the lowest part of the stratosphere, whereas at higher altitudes the aging by mixing changes are largely coupled to the residual circulation. Note that the mixing tendency depends on both the local mixing intensity and the background mean age gradient, which in turn is affected by the residual circulation. Hence, the effect of local mixing tendency trends (Figure 5) is not unambiguously related to changes in the local mixing intensity and also related to changes in background gradients. To further analyze trends in the local mixing intensity, independent mixing estimates are needed (e.g., effective diffusivity) [see *Nakamura*, 1996; *Haynes and Shuckburgh*, 2000].

Given the strong net effect of mixing on trends of mean age of air (via the net aging by mixing) implies that differences in mean age trends between models or between models and observations are likely related to differences in aging by mixing. The diagnostic method presented here may help to understand the differences between simulated and observed mean age trends in future work.

References

Abalos, M., F. Ploeger, P. Konopka, W. J. Randel, and E. Serrano (2013), Ozone seasonality above the tropical tropopause: Reconciling the Eulerian and Lagrangian perspectives of transport processes, *Atmos. Chem. Phys.*, *13*, 10,787–10,794.

Andrews, D. G., J. R. Holton, and C. B. Leovy (1987), Middle Atmosphere Dynamics, Academic Press, San Diego, Calif.

- Aschmann, J., J. P. Burrows, C. Gebhardt, A. Rozanov, R. Hommel, M. Weber, and A. M. Thompson (2014), On the hiatus in the acceleration of tropical upwelling since the beginning of the 21st century, *Atmos. Chem. Phys. Discuss.*, 14, 9951–9973.
- Austin, J., and F. Li (2006), On the relationship between the strength of the Brewer-Dobson circulation and the age of stratospheric air, *Geophys. Res. Lett.*, 33, 221–225, doi:10.1029/2006GL026867.

Birner, T., and H. Bönisch (2011), Residual circulation trajectories and transit times into the extratropical lowermost stratosphere, Atmos. Chem. Phys., 11, 817–827, doi:10.5194/acp-11-817-2011.

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

Brewer, A. W. (1949), Evidence for a world circulation provided by the measurements of helium and water vapour distribution in the stratosphere, Q. J. R. Meteorol. Soc., 75, 351–363.

Bunzel, F., and H. Schmidt (2013), The Brewer-Dobson circulation in a changing climate: Impact of the model configuration, J. Atmos. Sci., 70, 1437–1455, doi:10.1175/JAS-D-12-0215.1.

Butchart, N., et al. (2010), Chemistry-climate model simulations of twenty-first century stratospheric climate and circulation changes, J. Clim., 23, 5349–5374, doi:10.1175/2010JCLI3404.1.

Dee, D. P., et al. (2011), The ERA-Interim reanalysis: Configuration and performance of the data assimilation system, Q. J. R. Meteorol. Soc., 137, 553–597, doi:10.1002/qj.828.

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Diallo, M., B. Legras, and A. Chedin (2012), Age of stratospheric air in the ERA-Interim, Atmos. Chem. Phys., 12, 12,133–12,154.
 Dobson, G. M. B., D. N. Harrison, and J. Lawrence (1929), Measurements of the amount of ozone in the Earth's atmosphere and its relation to other geophysical conditions. Part III, Proc. R. Soc. London, Ser. A, 122, 456–486.

Engel, A., et al. (2009), Age of stratospheric air unchanged within uncertainties over the past 30 years, *Nat. Geosci.*, 2, 28–31, doi:10.1038/ngeo388.

Fueglistaler, S., B. Legras, A. Beljaars, J. J. Morcrette, A. Simmons, A. M. Tompkins, and S. Uppala (2009), The diabatic heat budget of the upper troposphere and lower/mid stratosphere in ECMWF reanalysis, Q. J. R. Meteorol. Soc., 638, 1–276, doi:10.1002/qj.361.

Garcia, R. R., and W. J. Randel (2008), Acceleration of Brewer-Dobson circulation due to increase in greenhouse gases, J. Atmos. Sci., 65, 2731–2739.

Garcia, R. R., W. J. Randel, and E. D. Kinnison (2011), On the determination of age of air trends from atmospheric trace species, J. Atmos. Sci., 68, 139–154.

Garny, H., T. Birner, H. Bönisch, and F. Bunzel (2014), The effects of mixing on age of air, J. Geophys. Res. Atmos., 119, 7015–7034, doi:10.1002/2013JD021417.

Hall, T. M., and R. A. Plumb (1994), Age as a diagnostic of stratospheric transport, J. Geophys. Res., 99(D1), 1059–1070.

Haynes, P., and E. Shuckburgh (2000), Effective diffusivity as a diagnostic of atmospheric transport: 1. Stratosphere, J. Geophys. Res., 105, 22,777–22,794.

Konopka, P. (2015), Hemispheric asymmetries and seasonality of mean age of air in the lower stratosphere: Deep versus shallow branch of the Brewer-Dobson circulation, J. Geophys. Res. Atmos., 120, doi:10.1002/2014JD022429, in press.

Konopka, P., et al. (2004), Mixing and ozone loss in the 1999–2000 Arctic vortex: Simulations with the three-dimensional Chemical Lagrangian Model of the Stratosphere (CLaMS), J. Geophys. Res., 109, D02315, doi:10.1029/2003JD003792.

Mahieu, E., et al. (2014), Recent Northern Hemisphere stratospheric HCl increase due to atmospheric circulation changes, *Nature*, *515*(7525), 104–108.

McKenna, D. S., P. Konopka, J.-U. Grooß, G. Günther, R. Müller, R. Spang, D. Offermann, and Y. Orsolini (2002), A new Chemical Lagrangian Model of the Stratosphere (CLaMS): 1. Formulation of advection and mixing, J. Geophys. Res., 107(D16), 4309, doi:10.1029/2000JD000114.

Monge-Sanz, B. M., M. P. Chipperfield, D. P. Dee, A. J. Simmons, and S. M. Uppala (2012), Improvements in the stratospheric transport achieved by a chemistry transport model with ECMWF (re)analyses: Identifying effects and remaining challenges, *Q. J. R. Meteorol. Soc.*, *139*, 654–673.

Nakamura, N. (1996), Two-dimensional mixing, edge formation, and permeability diagnosed in area coordinates, J. Atmos. Sci., 53, 1524–1537.

Neu, J. L., and R. A. Plumb (1999), Age of air in a "leaky pipe" model of stratospheric transport, J. Geophys. Res., 104, 243–255, doi:10.1029/1999JD900251.

Olsen, M. A., M. R. Schoeberl, and J. E. Nielsen (2007), Response of stratospheric circulation and stratosphere-troposphere exchange to changing sea surface temperatures, *J. Geophys. Res.*, *112*, D16104, doi:10.1029/2006JD008012.

Ploeger, F., M. Riese, F. Haenel, P. Konopka, R. Müller, and G. Stiller (2014), Variability of stratospheric mean age of air and of the local effects of residual circulation and eddy mixing, *J. Geophys. Res. Atmos.*, *120*, 716–733, doi:10.1002/2014JD022468.
Plumb, R. A. (2002), Stratospheric transport, *J. Meteorol. Soc. Jpn.*, *80*(48), 793–809.

Pommrich, R., et al. (2014), Tropical troposphere to stratosphere transport of carbon monoxide and long-lived trace species in the Chemical Lagrangian Model of the Stratosphere (CLaMS). *Geosci. Model Dev.*, 7, 2895–2916.

Ray, E. A., et al. (2010), Evidence for changes in stratospheric transport and mixing over the past three decades based on multiple data sets and tropical leaky pipe analysis, J. Geophys. Res., 115, D21304, doi:10.1029/2010JD014206.

Ray, E. A., et al. (2014), Improving stratospheric transport trend analysis based on SF₆ and CO₂ measurements, *119*, 14,110–14,128, doi:10.1002/2014JD021802.

Riese, M., F. Ploeger, A. Rap, B. Vogel, P. Konopka, M. Dameris, and P. M. Forster (2012), Impact of uncertainties in atmospheric mixing on simulated UTLS composition and related radiative effects, J. Geophys. Res., 117, D16305, doi:10.1029/2012JD017751.

Rind, D., R. Suozzo, N. K. Balachandran, and M. J. Prather (1990), Climate change and the middle atmosphere. Part I: The doubled CO₂ climate, J. Atmos. Sci., 47, 475–494.

Schoeberl, M. R., A. R. Douglass, Z. X. Zhu, and S. Pawson (2003), A comparison of the lower stratospheric age spectra derived from a general circulation model and two data assimilation systems, *J. Geophys. Res.*, *108*(D3), 4113, doi:10.1029/2002JD002652.

Shine, K. P., and P. Forster (1999), The effect of human activity on radiative forcing of climate change: A review of recent developments, *Global Planet. Change*, 20, 205–225.

Stiller, G. P., et al. (2012), Observed temporal evolution of global mean age of stratospheric air for the 2002 to 2010 period, Atmos. Chem. Phys., 12, 3311–3331.

Tan, W. W., M. A. Geller, S. Pawson, and A. da Silva (2004), A case study of excessive subtropical transport in the stratosphere of a data assimilation system, J. Geophys. Res., 109, D11102, doi:10.1029/2003JD004057.

Waugh, D. W., and T. M. Hall (2002), Age of stratospheric air: Theory, observations, and models, *Rev. Geophys.*, 40(4), 1010, doi:10.1029/2000RG000101.