Long-Term Changes in Stratospheric Age Spectra in the 21st Century in the Goddard Earth Observing System Chemistry-Climate Model (GEOSCCM)

Li, Feng1,2, Darryn W. Waugh3, Anne R. Douglass2, Paul A. Newman2, Susan E. Strahan1,2, Jun Ma4, J. Eric Nielsen5,2, and Qing Liang1,2

1Goddard Earth Sciences Technology and Research, Universities Research Space Association, Columbia, Maryland, USA
2NASA Goddard Space Flight Center, Greenbelt, Maryland, USA
3Johns Hopkins University, Baltimore, Maryland, USA
4Computational Physics Inc., Springfield, Virginia, USA
5Science Systems and Application Inc., Lanham, Maryland USA

Correspondence to: Feng Li (feng.li@nasa.gov)
Abstract

In this study we investigate the long-term variations in the stratospheric age spectra using simulations of the 21st century with the Goddard Earth Observing System Chemistry-Climate Model (GEOSCCM). Our purposes are to characterize the long-term changes in the age spectra and identify processes that cause the decrease of the mean age in a warming climate. Changes in the age spectra in the 21st century simulations are characterized by decreases in the modal age, the mean age, the spectral width, and the tail decay timescale. Our analyses show that the decrease in the mean age is caused by two processes: the acceleration of the residual circulation that increases the young air masses in the stratosphere, and the weakening of the recirculation that leads to the decrease of tail of the age spectra and the decrease of the old air masses. The weakening of the stratospheric recirculation is also strongly correlated with the increase of the residual circulation. One important result of this study is that the decrease of the tail of the age spectra makes an important contribution to the decrease of the main age. Long-term changes in the stratospheric isentropic mixing are investigated. Mixing increases in the subtropical lower stratosphere, but its impact on the age spectra is outweighed by the increase of the residual circulation. The impacts of the long-term changes in the age spectra on long-lived chemical traces are also investigated.
Coupled Chemistry-Climate Models (CCMs) consistently simulate an acceleration of the stratospheric circulation in the recent past and the 21st century [Butchart et al., 2006, 2010]. The strengthening of the stratospheric circulation in a warming climate is reflected in two diagnostics: the increase of the mean meridional mass circulation [Butchart and Scaife, 2001; Butchart et al., 2006, 2010; Li et al., 2008; Garcia and Randel, 2008; McLandress and Shepherd, 2009] and the decrease of the mean age of stratospheric air [Austin and Li, 2006; Oman et al., 2009; Butchart et al., 2010]. These two diagnostics are strongly correlated [Austin and Li, 2006], but they have different physical meanings. The mean meridional mass circulation, often approximated by the Transformed Eulerian Mean residual circulation, represents the mean advection part of the stratospheric transport circulation [Andrews et al., 1987]. In this paper the mean meridional mass circulation is referred as the residual circulation, although in other literatures it is also called the Brewer-Dobson circulation or diabatic circulation [e.g., Andrews et al., 1987; Shepherd, 2002]. The mean age of air is the average time for an air parcel to transport from troposphere to a stratospheric sample region. It is a measure of the strength of the stratospheric transport circulation. The mean age is determined not only by the residual circulation, but also by other processes such as isentropic mixing and recirculation [Waugh and Hall, 2002].

There are very few observational studies to verify the simulated mean age changes. Engel et al. [2009] examined a long-term record of mean age of air derived from CO$_2$ and
SF₆ measurements in the northern midlatitudes and found no significant trend in the last three decades, contrary to CCM simulations. However, Garcia et al. [2011] pointed out that the results of Engel et al. [2009] have serious caveats due to sparse sampling and the nonlinear growth rate of CO₂ and SF₆. Nevertheless, there are still doubts on the model projected mean age changes.

A major concern of the model results is that the mechanism for the decrease of the mean age is not clear. Previous studies have shown that the increase of the residual circulation plays an important role in driving the trend of the mean age [Austin and Li, 2006; Garcia et al., 2007; Oman et al., 2009], but there is not a complete understanding how these two processes are related. Strahan et al. [2009] demonstrated that in the tropical pipe the timescale of the residual circulation is significantly smaller than the mean age. The differences between the two timescales are caused by recirculation of air parcels between the tropics and midlatitudes. An air parcel could make multiple circulates between the tropics and midlatitudes. This recirculation process depends on mixing through the subtropical transport barriers [Neu and Plumb, 1999]. Thus changes in recirculation and mixing could also impact the trend of the mean age. But it is not clear how recirculation and mixing respond to greenhouse gas increases and how these changes impact the mean age.

Investigating the long-term changes in the age spectra will help to clarify the roles of changes in the residual circulation, recirculation and mixing in driving the decrease of the mean age. The age spectrum is the probability distribution function of transit times
between a source region in the troposphere or tropopause and a sample region in the stratosphere [Hall and Plumb, 1994; Waugh and Hall, 2002]. The mean age is the first moment of the age spectrum, or the average of all the possible transit times. In addition to the mean age, other important parameters that characterize the age spectrum include the modal age and spectral width. The modal age corresponds to the time of the spectral peak. It represents the most probable transit time and is directly associated with the timescale of the bulk velocity of tracer transport [Waugh and Hall, 2002]. The modal age agrees very well with the timescale of the residual vertical velocity within the tropical pipe region [Strahan et al., 2009]. The spectral width is related to the second moment of the age spectrum and is a measure of the strength of the recirculation [Strahan et al., 2009]. The age spectrum contains complete information on transit times and is more useful than the mean age in understanding the distribution of photochemically important trace species in the stratosphere [Schoeberl et al., 2005; Waugh et al., 2007]. While the decrease of the mean age in the 21st century has been extensively documented, no previous studies have investigated the long-term changes in the age spectra.

In this paper we investigate the long-term changes in the stratospheric age spectra in the 21st century using simulations with the Goddard Earth Observing System Chemistry-Climate Model (GEOSCCM). The main purposes of this study are to characterize the long-term changes in the age spectra and to identify processes that cause the decrease of the mean age. This paper is organized as following. A brief review of the age spectrum theory and a detailed description of our method to calculate the age spectra are given in Section 2. This is followed by an introduction of the GEOSCCM and the experiment
setup in Section 3. Results are presented in Section 4. Discussions are given in Section 5. Section 6 is the conclusion.

2 Method

The age spectrum is a Green's function, or a boundary propagator, that solves the continuity equation for the mixing ratio of a conserved and passive tracer [Hall and Plumb, 1994]. It is also called the Transit-Time Distribution (TTD) in ocean and tropospheric transport literatures [Holzer et al., 2003; Haine et al., 2008]. The age spectrum is expressed by

\[ \chi(r,t) = \int_0^\infty \chi(\Omega, t - \xi)G(r,t|\Omega,t - \xi)d\xi \]  

(1)

where \( \chi(r,t) \) is the tracer mixing ratio at a sample region \( r \) and sample time \( t \), \( \xi \) is the elapse of time between the sample time \( t \) and source time \( t' \) (i.e., \( \xi = t - t' \)), the source time \( t' \) is when the tracer had last contact with the boundary resource region \( \Omega \), and \( G(r,t|\Omega,t-\xi) \) is the age spectrum. The physical meaning of the age spectrum is clear in Equations 1: \( G(r,t|\Omega,t-\xi)d\xi \) represents the mass fraction of the air parcel at \( r \) and \( t \) that was last in contact with \( \Omega \) between \( \xi \) and \( \xi + d\xi \) ago.

Different methods have been used to calculate age spectra [e.g., Hall and Plumb, 1994; Schoeberl et al., 2003; Haine et al., 2008]. Among all the methods that have been used, the pulse tracer method is the most direct approach. It should be emphasized that the pulse tracer method does not directly produce the age spectrum. Instead it generates another kind of boundary propagator \( G(r,t'|t-\xi|\Omega, t') \), which is called the Boundary
Impulse Response (BIR) [Haine et al., 2008; Li et al., 2012]. Once the BIR is obtained, the age spectrum can be calculated from the BIR.

Replace $t$ with $t'+\xi$, then Equation 1 becomes

$$
\chi(r, t'+\xi) = \int_0^\infty \chi(Q, t')G(r, t'+\xi|Q, t')d\xi
$$

(2)

Note that if we set the boundary condition of $\chi(Q, t')$ as a Dirac delta function, then Equation 2 yields $\chi(r, t'+\xi) = G(r, t'+\xi|Q, t')$. Thus the BIR is the time evolving response to a delta function boundary condition. The BIR can be easily calculated in models using the pulse tracer: release a pulse of a conserved and passive tracer at a chosen source region and source time, and the time series of the tracer’s mixing ratio at any interior point $r$ is $G(r, t'+\xi|Q, t')$.

The mathematic relationship between the BIR and the age spectrum is simple, but Equation 2 does not have the same clear physical meaning as Equation 1. It is important to recognize that the age spectrum and the BIR are different due to their time dependence.

Only in the special case of steady flow the age spectrum is the same as the BIR. In this case we calculate a single realization of the BIR as the age spectrum [Haine et al., 2008]. Computing the age spectrum in unsteady flow is more complicated and requires a series of BIRs that are launched in different source times [Holzer et al., 2003; Haine et al., 2008; Li et al., 2012]. There are two approaches. If one is interested in the seasonal and interannual variability of the age spectra, then one needs to reconstruct time varying age spectra from BIRs [Li et al., 2012]. But if one is mainly interested in the time-averaged properties of the age spectra, one can use the mean of an ensemble of BIRs as a time-
averaged age spectrum. This is because the BIR and the age spectrum share the same
boundary propagator distribution (Haine et al. [2008]).

The stratospheric transport has large seasonal and interannual variations. These
variations have to be accounted for in order to correctly capture the annual mean or the
seasonal properties of the age spectra. Li et al. [2012] investigated the seasonal
variations of the stratospheric age spectra in the GEOSCCM. They reconstructed
seasonally varying age spectra from twelve BIRs released in each month of the annual
cycle. Here, we focus on the long-term changes in the time-averaged properties of the
age spectra, and take the second approach introduced above to use the mean of an
ensemble of BIRs launched in different times as the time-averaged age spectra.

The method of Hall et al. [1999b] is followed to conduct the pulse tracer experiment.
The tropical lower troposphere between 10°N and 10°S and from the surface to about 800
hPa is chosen to be the boundary source region. As an approximation of the delta
function boundary condition, the mixing ratio of the tracer is set to an arbitrary positive
value in the first month of the experiment and then held as zero through the rest of the
experiment in the boundary source region. There are no other sources or sinks for the
tracer. The pulse experiment runs for 20 years.

We perform an ensemble of ten pulse tracer experiments in a 20-year period and use the
mean of the resultant ten BIRs as the time-averaged age spectra in this period. The ten
pulse tracers are released respectively in January and July in each of the first five years of
the 20-year period. The different release times of the pulse tracers are chosen to represent the seasonal and interannual variability of stratospheric transport. We conduct these ten pulse tracer experiments for each of the five 20-year periods in a 21st century simulation with the GEOSCCM. A total of fifty BIRs are calculated and five age spectra are obtained. The five age spectra cover the model year 2000-2019, 2020-2039, 2040-2059, 2060-2079, and 2080-2099, respectively.

Figure 1 shows examples of the BIRs and their ensemble mean in 3 different locations in the 2000-2019 period. The BIRs released in January and July are shown in red and blue, respectively. The BIRs have strong seasonal and interannual variability. In the tropics, the interannual variability reflects the impacts of the quasi-biennial oscillation (QBO) on the BIRs. In the extratropics, seasonal differences of the BIRs stand out, although there are considerable interannual variations. The age spectrum (thick black line), i.e. the ensemble mean of the BIRs, is different from the ensemble members. Therefore it is important to use an ensemble of BIRs in order to accurately capture the time-averaged property of the age spectra.

3 GEOSCCM and Simulation

The model we use in this study, the GEOSCCM [Pawson et al., 2008], couples the GEOS5-AGCM [Rienecker et al., 2008] with a comprehensive stratospheric chemistry package [Douglass et al., 1997]. The GEOSCCM has 72 vertical levels with a model top at 0.01hPa. The horizontal resolution in the GEOSCCM is adjustable and a grid of 2°
latitude by 2.5° longitude is used in this study. The pulse tracer experiments were carried out with a sensitivity simulation of the 21st century in which CO₂ increases under the IPCC (2001) A1b scenario, but the amount of ozone depleting substances (ODSs) and other greenhouse gases are fixed at the year 2000 level conditions. The simulation uses modeled sea surface temperature and sea ice in the 21st century under the A1b scenario from the NCAR Community Climate System Model 3.0. In this sensitivity simulation, the model climate change is solely driven by increases in CO₂ and sea surface temperature. As described in detail in the previous section, a total of fifty pulse tracer experiments are carried out with the simulation. Fifty BIRs are generated and five age spectra are computed. In the rest of the paper, the five age spectra are referred as 2000, 2020, 2040, 2060, and 2080 spectra, respectively. All results presented in this paper are zonally and monthly averaged.

The GEOSCCM has participated in the Chemistry-Climate Model Validation Activity 2 (CCMVal-2) and is one of the best models in CCMVal-2 [SPARC CCMVal, 2010]. GEOSCCM simulations of the recent past compare well with observations in stratospheric chemistry, transport, and dynamics [SPARC CCMVal, 2010; Strahan et al., 2011]. Quite realistic stratospheric transport characteristics, such as the mean age, the tropical ascent rate, and the lower stratospheric mixing rate, are captured by the GEOSCCM. But the Antarctic polar vortex is more isolated in GEOSCCM than observed, a common bias in CCMs. The version of the GEOSCCM used in this study is slightly different from the one participated in CCMVal-2 in that it produces a QBO by increasing the non-orographic gravity wave source in the tropics.
4 Results

Our simulation projects a decrease in the mean age of air and an increase in the residual circulation in the 21st century, consistent with previous CCM studies [e.g., Butchart et al., 2006, 2010]. Figure 2a shows the differences in the mean age of air between the 2080 and 2000 spectra, where the mean age is computed from the age spectrum by

$$\Gamma(r,t) = \int_0^\infty \xi G(r,t; \Omega, t - \xi) d\xi.$$  

The mean age is younger in 2080 than in 2000 everywhere in the stratosphere. The rate of decrease is larger in the midlatitudes than in the tropics, indicating a reduced mean age gradient between these two regions and an enhanced tropical ascent rate [Neu and Plumb, 1999]. Strong decrease in the mean age is found in the subtropical and midlatitude lower stratosphere in both hemispheres, suggesting an increase of the quasi-horizontal transport in this region. In the northern hemisphere lower stratosphere the area of large mean age decrease extends to the high latitudes. The largest decrease in the mean age is found in the Arctic lower stratosphere (over 0.8 years, or 20%). This suggests the acceleration of the quasi-horizontal transport is particularly strong in the northern hemisphere. Overall these results agree well with those produced by the version of the GEOSCCM that participates in CCMVal-2 [Butchart et al., 2010], although the decrease of the mean age is larger in the current simulation. Note that in CCMVal-2 the GEOSCCM simulates ozone recovery in the 21st century. A stronger decrease in the mean age without ozone recovery is consistent with the findings of Oman et al. [2009] that ozone recovery in the 21st century acts to reduce the rate of mean age decrease.
The decrease in the mean age of air is consistent with the acceleration of the residual circulation (Figures 2b and 2c). The changes in the residual vertical velocity ($w'$) and meridional velocity ($v'$) clearly show two cells in each hemisphere. The increase in the residual velocities is much stronger in the lower and upper stratosphere than in the middle stratosphere. Within each cell increase in the tropical upwelling is balanced by increase in the poleward mass transport and extratropical downwelling. Changes in the lower branch of the residual circulation are confined to the tropics and midlatitudes, whereas the increase of the upper branch of the residual circulation extends all the way to the high latitudes.

Previous studies have shown that the mean age is strongly correlated with the tropical upward mass flux in the lower stratosphere, a measure of the overall strength of the residual circulation [Austin and Li, 2006; Butchart et al., 2010]. But the timescale of the residual circulation should not be confused with the mean age. The residual velocity approximates the bulk velocity of tracer transport. Waugh and Hall [2002] showed that the timescale of the residual circulation (or bulk tracer transport) is closely associated with the modal age in regions of weak mixing such as the tropical pipe. The modal age and the timescale of the residual circulation are smaller than the mean age because the stratospheric age spectrum has an asymmetric shape with a long tail [Hall and Plumb, 1994]. Schoeberl et al. [2008] calculated the vertical velocity for water vapor advection from the tape recorder signal in the tropical pipe and found that it agrees very well with the residual vertical velocity. Strahan et al. [2009] further showed that the modal age is a
lower limit of the timescale of the residual vertical velocity and both are shorter than the mean age.

In order to illustrate the relationship between the mean age, the modal age, and the timescale of the residual circulation, Figure 3 compares these three timescales in the tropical pipe region between 10°N and 10°S and from 70 hPa to 1 hPa. First we note that the transit time of the mean vertical advection is closely associated with the modal age and is significantly shorter than the mean age (Figure 3a), confirming the results of Strahan et al. [2009]. The mean age, modal age, and the timescale of the vertical advection are all shorter in 2080-2099 than in 2000-2019 (Figure 3b). The decreases in the vertical advection timescale and modal age are comparable to each other in most of the stratosphere, and they are distinctly smaller than the decrease of the mean age. In terms of the absolute value, decreases in the mean age are more than twice those in the transit time of the vertical advection. The relative changes in the mean age are also larger than that in the transit time of the vertical advection. This example shows that the decrease of the mean age can only be partly explained by the acceleration of the residual velocities.

Although the decrease in the mean age in a warming climate has been well documented [Garcia and Randel, 2009; Oman et al., 2009], no previous studies have investigated the long-term changes in other important age spectral parameters such as the modal age and width. Figure 4 shows the distribution of the modal age and spectral width of the 2000 spectra (contour) and the differences between the 2080 and 2000 spectra (color).
distribution of the modal age is somewhat similar to the mean age, but the modal age has strong gradients in the high latitude lower stratosphere. The modal age increases by up to 2-4 times in a very narrow latitudinal band in this region. From 2000 to 2080 the modal age decreases in most of the stratosphere. The changes in the modal age are less smoothly distributed than the changes in the mean age. In general the decrease in the modal is smaller in the tropics than in the high latitudes. The largest decrease is seen in the polar lower stratosphere, especially in the Arctic. We will show later that the large change of the modal age in the polar lower stratosphere is caused by change in the multi-mode spectral shape in this region.

The spectral width is related to the square root of the second moment of the age spectrum by
$$\Delta(r,t) = \sqrt{\frac{1}{2} \int_0^\infty (\xi - \Gamma(r,t))^2 G(r,t | \Omega,t - \xi) d\xi}. $$

It quantifies the spread of the transit time distribution [Waugh and Hall, 2002]. Qualitatively, the width indicates how important the tail of the spectrum contributes to the mean age. The wider the width, the longer the tail, and the larger the fraction of the tail contributes to the mean age. The tail of the age spectrum is related to the strength of the recirculation, thus the width can also be viewed as a measure of the strength of the recirculation [Strahan et al., 2009]. Figure 307b shows that the distribution of the spectral width is similar to that in the mean age below about 10 hPa, but the width becomes uniform with a value of about 2 years above 10 hPa. Throughout the stratosphere, the spectral width becomes narrower at 2080 than at 2000. The largest decreases in the width are found in the subtropical (20°-40° N and S) lower stratosphere.
We now examine the distribution of the age spectra and their changes at 50 and 10 hPa. The focus is on the lower stratosphere because the largest changes in the mean age, modal age, and spectral width occur below 10 hPa. Figure 5 shows the 2000 age spectra at 50 hPa as a function of latitude and the differences between the 2080 and 2000 age spectra. The age spectra have an asymmetrical shape with a young peak and a long tail. Only the first 10 years of the age spectra are shown because the tail of the spectra decays rapidly with increasing transit time. The age spectra have large latitudinal variations with younger and stronger peaks and more compacted distribution in the tropics than in the extratropics. The modal age has very sharp gradients around 70° latitudes in both hemispheres with values increasing by more than 2 times in a narrow latitudinal band.

The 2080 age spectra have higher percentages of young air and lower percentages of old air compared to the 2000 age spectra (Figure 5b). The transition from positive (more young air) and negative (less old air) differences follows approximately the modal age of the 2000 age spectra. This indicates that the spectral peaks become younger and stronger in the 2080 spectra. These changes lead to decreases in the mean age and spectral width. Furthermore, a younger and stronger spectral peak together with a narrower width means that the 2080 age spectra have a shorter tail. There are multiple peaks at high latitudes, suggesting the age spectra in this region have a multi-model shape [Li et al., 2012].

Overall the age spectra at 10 hPa are similar to those at 50 hPa, although they have smaller latitudinal variations, especially in the spectral width (Figure 6a). The changes between the 2080 and 2000 age spectra at 10 hPa are also similar to those at 50 hPa.
(Figure 6b). Again the 2080 spectral have a larger fraction of young air than the 2000 age spectra and the change from more young air to less old air occurs at about the time of the modal age of the 2000 spectra. A notable discrepancy is that the age spectra differences at high latitudes at 10 hPa do not show multiple peaks.

We investigate the changes in age spectra in the lower stratosphere in more detail by examining the evolution of the age spectra at 50 hPa in different locations. Figure 7 clearly shows that as the integration progresses, the modal ages become younger, the spectral peaks get stronger, the tails are shorter, and the widths are narrower. The age spectra undergo larger changes in 2020-2039 than in other periods. The age spectra at high latitudes, particularly in the Arctic, have different characteristics from those at low and middle latitudes. They have multiple peaks with comparable magnitude, in contrast to the single-mode shape at lower latitudes. For example, in the Arctic the 2000 spectrum (black line) has 3 peaks between 3 and 4 years of transit time (Figure 7e). The strongest peak, which is just slightly stronger than other peaks, occurs at 3.7 years. This multi-mode spectral shape indicates that there are multiple, nearly equally important transport pathways to the polar lower stratosphere. From 2000 to 2080, the percentages of air younger than 2 years increase significantly from 15% to 25%. The percentages of air with transit time between 2 and 4 year increase only slightly from 36% to 38%. The fraction of air older than 4 years decreases from 49% in 2000 to 37% in 2080. In the 2080 spectrum (red line) the peak at 1.8 years becomes the strongest among several comparable peaks, and we obtain a decrease of modal age of 1.9 years from 2000 to 2080. However, the spectral peak at 1.8 years in the 2080 spectrum does not correspond
to the peak at 3.7 years in the 2000 spectrum. The dramatic modal age change reflects the changes of relative strength of the multiple peaks.

The results presented in Figures 5-7 indicate that the decrease of the mean age in the 2080 age spectra is due to an increases in the percentage of the young air and a decrease in the percentages of the old air, or the tail of the age spectra. The changes in the tail of the age spectra can be more easily seen when the age spectra are plotted in the logarithmic scale. Figure 8 is the same as Figure 7, but it uses the logarithmic scale and covers the whole 20-year period. There are several interesting features regarding the distribution and change of the tail of the age spectra. First, the tail can be represented by a linear regression line. That is, the tail is approximated very well by an exponentially decaying mode $\Psi_0(r,t)\exp(-\frac{r}{\tau_0})$, where $\tau_0$ is the decay timescale. Second, the slope of the tail, or the decay timescale $\tau_0$, appears to be independent of locations. And third, the tails are shorter in 2080 than in 2000. The decrease in the tail can be quantified by a decrease in $\tau_0$.

The decay timescale $\tau_0$ has long been known as a fundamental stratospheric transport diagnostic [Prather 1996; Hall et al., 1999a; Ehhalt et al., 2004]. Under the steady state condition, the age spectrum can be decomposed into a set of normal modes, each of which decays exponentially at a timescale that is equal to the reverse of its eigenvalue [Hall et al., 1999a]. The base mode has the longest decay timescale $\tau_0$ and it decays more slowly than the higher modes. For long transit time, only the base mode survives and thus the tail of the age spectrum can be approximated by the base mode. $\tau_0$ is a
unique transport diagnostic because it is independent of location. Physically $\tau_o$ describes
how fast the mixing ratio of a conserver tracer in the stratosphere decays due to transport
alone [Ehhalt et al., 2004]. It can also be viewed as an integrated measure of the strength
of stratospheric recirculation.

The decrease in the mean age of air through the 21st century is strongly correlated with
the decrease of $\tau_o$. Figure 9a plots the evolution of $\tau_o$ from 2000 to 2080 against the
globally and stratospherically (100-1 hPa) averaged mean age. When calculating $\tau_o$, the
tail of the age spectra is regressed onto a single exponentially decay mode. Here we
define the tail as the region with transit time older than 4 years, noting that the age
spectra start to exponentially decay at about 4 years (see Figure 8). The correlation
between $\tau_o$ and the mean age is 0.998, though we only have 5 samples. This strong
correlation indicates that the decrease of the tail makes a significant contribution to the
decrease of the mean age.

The decrease of $\tau_o$ is highly anti-correlated with the increase of the upward mass flux in
the tropical lower stratosphere (Figure 9b), which means that an accelerated residual
circulation acts to weaken the stratospheric recirculation. It is known that changes in
mixing cross transport barriers could affect recirculation [Neu and Plumb, 1999; Strahan
et al., 2009], but our results show that the stratospheric mean meridional circulation has a
significant impact on recirculation and the tail part of the age spectra (also see
discussions in Section 5.1).
The tail of the age spectra has received less attention than other spectral parameters in previous studies, but the tail has a significant impact on the mean age [Schoeberl, 2003, 2005]. The mean age can be regarded as the mixing ratio of an ideal clock tracer that has a linearly increasing stratospheric source and a fixed tropospheric concentration [Waugh and Hall, 2002]. Therefore the tail of the age spectra weights heavily on the mean age.

Figure 10a shows the fractional contribution of the tail (defined as transit times old than 4 years) to the mean age in the 2000 spectra (contour) and the changes of the fractional contribution between the 2080 and 2000 spectra (color). The distributions of the tail contribution look similar to those of the mean age. The tail contributes a small fraction to the mean age in the tropical lower stratosphere where the age spectra are dominated by young spectral peaks. In the rest of the stratosphere, however, the tail accounts for more than 50% of the mean age. From 2000 to 2080, the tail becomes shorter and its fractional contributions to the mean age decrease everywhere in the stratosphere. The largest decrease of the tail contribution occurs in the subtropical lower stratosphere between 20° and 30° latitudes and centered at 70 hPa, which corresponds to the largest decrease in the spectral width (see Figure 4b). This correspondence is not a complete surprise as the width is closely linked with the tail.

The changes in the tail can also be quantified by the changes in the averaged transit time in the tail, referred to as the tail age here. Comparing Figure 10b with Figure 2a reveals that the tail age decreases more than the mean age between 2080 and 2000. This confirms that the decrease in the tail decay timescale indeed makes an important contribution to the decrease in the mean age. The differences between changes in the
mean age and tail age are those from air younger than 4 years. It can be inferred from Figure 10b and Figure 2a that the average transit time for air parcels younger than 4 years is shorter at 2080 than at 2000.

5 Discussions

5.1 Long-Term Changes in Isentropic Mixing

In addition to the residual circulation, changes in isentropic mixing could also impact the trend of the mean age and age spectra. For instance, enhanced mixing from the midlatitudes to the tropics increases the probability of recirculation within the stratosphere, and thus leads to a longer tail, a wider width, and an older mean age [Neu and Plumb, 1999; Strahan et al., 2009]. However, it is not clear how stratospheric mixing changes in the 21st century, and what are the impacts of the changes in mixing on the age spectra. Here we calculate the equivalent length of N₂O to investigate the long-term changes in isentropic mixing. The equivalent length of a chemical tracer measures the geometry complexity of the tracer contours in the equivalent latitude coordinate on isentropic surfaces [Nakamura, 1996; Ma et al., 2003]. Nakamura [1996] showed that the equivalent length, or more accurately its square, is a useful diagnostic of the efficiency of isentropic tracer mixing.

Figure 11 shows the distribution of the normalized equivalent length squared of N₂O (η) for 2000-2019 (lines) and the relative differences between 2080-2099 and 2000-2019 (colors). Large values of η correspond to strong mixing, whereas small values indicate
The most striking feature in the changes of $\eta$ between 2080-2099 and 2000-2019 is a large increase in $\eta$ in the tropical/subtropical lower stratosphere. Mixing increases up to 50% in the base of the tropical pipe. There are essentially no changes in mixing across the subtropical barriers in the middle and upper stratosphere. Thus the tropical pipe becomes more leaky, but only in the base. Mixing decreases significantly just below the tropical pipe and the area of reduced $\eta$ extends to midlatitudes. Mixing increases in the Arctic lower stratosphere, but it remains the same in the Antarctic stratosphere.

The pattern of changes in $\eta$ in the tropical/subtropical lower stratosphere indicates that the distribution of $\eta$ is shifted upward in this region from 2000-2019 to 2080-2099. The upward shift in the mixing pattern is consistent with the zonal wind changes. Figure 12 shows the changes in temperature and zonal wind between 2080-2099 and 2000-2019. The meridional temperature gradient increases in the subtropical upper troposphere and lower stratosphere (UTLS), due to strong warming in the tropical upper troposphere.
This causes significant westerly acceleration of the zonal wind in the tropical/subtropical UTLS, leading to an upward lift of the subtropical jets and the zero wind line. Shepherd and McLandress [2011] showed that, through critical-layer control, the upward lift of the zonal wind in the subtropical lower stratosphere shifts higher the wave breaking altitude, which drives the acceleration of the lower branch of the residual circulation. Since mixing is generated by wave breaking, we argue that the upward shift of wave breaking could also explain changes in mixing in the tropical/subtropical lower stratosphere.

In order to demonstrate the relationship among changes in isentropic mixing, residual circulation, and zonal wind, Figure 13a plots the evolution of the 20-year mean $\eta$ averaged in 10°-30° latitudes and 440-520 K against the tropical upward mass flux at 70 hPa and the subtropical UTLS zonal wind averaged in 10°-30° latitudes and 200-70 hPa in bother hemispheres. The strong correlations among the three diagnostics support our argument that the increases in the mixing and residual circulation are closely related to each other, and both are driven by zonal wind changes that lead to enhanced wave breaking in the subtropical lower stratosphere.

However, the increase in mixing in the subtropical lower stratosphere does not produce older main age or wider width in this region. Figure 13b shows that as mixing increases, the mean age (black line) and the spectral width (blue line) in the subtropical lower stratosphere decrease. Ray et al. [2010] showed that, using the conceptual tropical leaky pipe model, the trend of the mean age is very sensitive to the relative importance of changes in the upwelling and mixing. One important result in this study is that changes
in mixing and upwelling are not independent to each other; rather they are closely related.

Our model results indicate that the impact of enhanced mixing on the age spectra is outweighed by the acceleration of the residual circulation.

5.2 Relationship between Changes in Age Spectra and Chemical Tracers

The mean age of air is compactly related with long-lived chemical tracers such as N$_2$O and CH$_4$ in the lower stratosphere [e.g., Boering et al., 1996]. This compact relationship can be used to infer the distribution of the mean age. For example, Andrews et al. [2001] derived an empirical relationship between the mean age and N$_2$O in the midlatitude lower stratosphere from NASA ER-2 aircraft measurements. Applying this relationship to all latitudes, they estimated the seasonal distribution of the mean age in the lower stratosphere. However, as the mean age and age spectra change in response to CO$_2$ increases in the 21st century, the relationship between the mean age and chemical tracers also changes.

Figure 14a compares the compact relationship between the mean age and N$_2$O in 2000-2019 and 2080-2099 in the northern hemisphere lower stratosphere 100-50 hPa. For age older than about 1.5 years, the compact line in 2080-2099 is shifted to the left of the line in 2000-2019. This shift means that the mean age and N$_2$O respond differently to circulation change in the 21st century. As the residual circulation speeds up, the distribution of the mean age and long-lived chemical tracers are lifted upward, but the
upward lifting in the mean age is stronger such that a given mean age is associated with a smaller mixing ratio of N$_2$O in 2080-2099 than in 2000-2019.

The mean age also exhibits a compact relationship with fractional release of chlorofluorocarbons (CFCs) [Shauffler et al., 2003]. The fractional release is defined as $fr = 1 - \chi(r)/\chi_0$, where $\chi(r)$ is the mixing ratio of a CFC at a stratospheric sample region $r$, and $\chi_0$ is the mixing ratio of the same CFC that would have had if there was no chemical loss [Shauffler et al., 2003]. The fractional release provides useful information on photochemical loss of CFCs. Figure 14b shows that the compact relationship between the mean age and CFC12 (CF$_2$Cl$_2$) changes from 2000-2019 to 2080-2099 in a manner similar to the change of the mean age – N$_2$O relationship. Note that when calculating the fractional release, $\chi_0$ is simply taken as the fixed boundary condition value. But in general when the boundary condition changes with time, $\chi_0$ is calculated using Equation 1 with the knowledge of the age spectrum.

Our results are very similar to Douglass et al. [2008], who also showed that the GEOSCCM reproduces the observed compact relationship between the mean age and the fractional release. The shift in the relationship between the mean age and the long-lived tracers indicates that the mean age is more sensitive to the circulation change than the tracers. We argue that this is because the tracers are less sensitive to change in the tail of the age spectra. As discussed in the previous section, changes in the tail of the age spectra significantly impact the mean age (see Figure 10). The chemical tracers, however, are not as sensitive to the tail of the age spectra [Schoeberl et al., 2005].
air parcels in the tail region could have released most of the chemical tracer they carry because they are likely to experience higher maximum altitude and stronger photochemical loss than those with shorter transient times [Waugh et al., 2007; Douglass et al., 2008]. Therefore chemical tracers respond differently to changes in the age spectra.

We use the ideal "radioactive" tracer to test the above hypothesis. Assume the radioactive tracer has a spatially uniform decay rate $\lambda$ in the stratosphere and a fixed surface concentration $c$, its mixing ratio can be written as

$$\chi(r,t) = c \int_0^\infty e^{-\lambda \xi} G(r,t | \Omega, t - \xi) d\xi.$$  (3)

[Schoeberl et al., 2005]. In contrary to the clock tracer, the mixing ratio of the radioactive tracer relies more on the head than the tail of the age spectrum. Since we have the age spectrum, we can calculate the mixing ratio and the mass burden of the radioactive tracer from Equation 3. Figure 15 shows the evolution of the stratospheric (100-1 hPa) mass burden of three radioactive tracers with different decay rate. For comparison the evolution of the stratospheric burden of CFC11 (green), CFC12 (blue) and the mean age (dashed) are also plotted. Except for the mean age, all other tracers' stratospheric burden increases in the 21st century. For the radioactive tracers, the faster the decay rate the larger the mass burden increasing. The mass burden of CFC11 increases more than that of the CFC12.

To better understand the different long-term changes of the mass burden of these tracers, we calculate the changes of stratospheric air masses as a function of transit time. Figure
16a shows the air mass changes between 2080 and 2000 normalized to the total stratospheric air mass burden, and Figure 16b is the relative changes between 2080 and 2000. The stratospheric air mass burden is a constant, but the distribution of air masses changes with time. The air masses younger than 2 years increase and the air masses older than 2 years decrease between 2080 and 2000. The changes of the young and old air masses compensate each other. However, the mass burden of tracers may increase or decrease with time depending on how the tracers weight toward different parts of the age spectrum. The mean age, i.e., the mixing ratio of the clock tracer, decreases because the clock tracer weights more in the old air than the young air. The radioactive tracers are more sensitive to the changes in the young air than the old air, and the mass burden of the radioactive tracers increases. A radioactive tracer with a faster decay rate has a larger weighting in the young air masses than one with a slower decay rate, and therefore its stratospheric mass increases more. This argument can be used to explain different changes in CFC11 and CFC12. CFC11 has a stronger local chemical loss frequency than CFC12 [Douglass et al., 2008], and its stratospheric mass burden increases more than CFC12. Note that all the above arguments are based on the condition that the tracer’s surface boundary condition is fixed. Nevertheless, our analyses clear show that the age spectrum is more relevant to chemical tracers than the mean age.

6 Conclusion

The long-term changes in the stratospheric age spectra in response to CO₂ increases in the 21st century are investigated using the GEOSCCM simulations. Changes in age spectra
are characterized by increases in young air masses and decreases in old air masses, younger and stronger peaks, shorter tails, and more compacted distribution. These changes lead to decreases in the mean age, modal age, spectral width, and tail decay timescale. An important result of this study is that changes in the tail of the age spectra make an important contribution to the decrease in the mean age.

A major purpose of this paper is to identify processes that cause the long-term changes in the mean age of air. Our analyses show that the decrease in the mean age is driven by two processes. The first process is the acceleration of the residual circulation that increases young air masses in the stratosphere. This process has been shown to be directly associated with the decrease of the modal age in the tropical pipe region. The second process is the weakening of the recirculation in the stratosphere, which leads to a shorter tail of the age spectra and a decrease of old air masses. This process is quantified by the decrease in the decay timescale of the tail of the spectra. We have shown that the decrease in the tail decay timescale is strongly correlated with the increase of the residual circulation. An accelerated residual circulation increases the stratosphere-troposphere mass exchange rate, which weakens the recirculation of tracers within the stratosphere. In summary, both processes are related to the strengthening of the residual circulation, but they impact different aspects of the age spectra.

The long-term changes in stratospheric mixing are investigated using the equivalent length of N₂O. In the simulation, the tropical pipe becomes more leaky in its base at the end of the 21st century. If there were no changes in tropical upwelling, the enhanced
mixing would increase the recirculation between the tropics and midlatitudes, which would lead to older mean age, larger width, and longer tail. However, the increase of isentropic mixing in the subtropical lower stratosphere is closely associated with the increase of tropical upwelling. Our model results indicate that the impacts of increased mixing on the age spectra are dominated by the acceleration of the residual circulation.

The mean age and chemical tracers respond differently to changes in the age spectra because they are sensitive to different parts of the age spectra. The mean age weights heavily on the tail of the age spectra, whereas the chemical tracers are more sensitive to the head of the age spectra. Because young and old air masses change differently, the mean age and chemical tracers have different long-term changes in the 21st century. Clearly the age spectrum is more useful than the mean age to study chemical tracers.

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Figure Captions:

Figure 1: Examples of the Boundary Impulse Responses (BIRs) in 2000-2019 at three locations. For this 20-year period, five BIRs are released in January (red) and five BIRs are released in July (blue) of 2000-2004. The thick black line is the age spectrum, which is calculated as the mean of the ten BIRs. The unit is 1/month.

Figure 2: (a) Distribution of the 2000 age spectrum mean age (lines) and the differences in the mean age between the 2080 and 2000 age spectra (color). (b) Distribution of the residual vertical velocity for the period 2000-2019 (lines) and the differences in the residual vertical velocity between 2080-2099 and 2000-2019 (color). (c) Same as Figure 2b, but for the residual meridional velocity.

Figure 3: (a) Comparison of the mean age (dashed), modal age (dotted), and the timescale of the residual vertical velocity in the tropical pipe region (10°S – 10°N, 70hPa – 1 hPa) in the period 2000-2019. The error bars represent interannual variations of these three timescales. (b) Difference in the mean age (dashed), model age (dotted) and the timescale of the residual velocity in the tropical pipe region between 2080-2099 and 2000-2019.

Figure 4: The color shadings are differences in the modal age (left) and spectral width (right) between the 2080 and 2000 age spectra. The contours are the modal age (left) and spectra width (right) of the 2000 age spectra.
Figure 5: (a) Distribution of the 2000 age spectra at 50 hPa as a function of latitude. The black solid, dashed, and dotted lines are the mean age, spectral width, and modal age, respectively. (b) Differences between the 2080 and 2000 age spectra at 50 hPa. The black lines are the same as in panel (a). The green solid, dashed, and dotted lines are the mean age, spectral width, and modal age in the 2080 age spectra, respectively. In both panels the color scales are normalized to the maximum value shown at the top of the color bar. The unit is 1/month.

Figure 6: Same as Figure 5, but for age spectra at 10 hPa.

Figure 7: Evolution of the age spectra in the 21st century at different locations at 50 hPa.

Figure 8: Same as Figure 7, but on logarithmic scale.

Figure 9: (a) The scatter plot of the decay timescale of the tail of the age spectra against the globally and stratospheric averaged (100 – 1 hPa) mean age. (b) The scatter plot of the tail decay timescale against the tropical upward mass flux at 70 hPa. The error bars represent the interannual variations of these diagnostics.

Figure 10: (a) The fractional contribution of tail to the mean age in the 2000 age spectra (lines) and the differences between the 2080 and 2000 age spectra (colors). The tail is defined as the region with transit time older than 4 years. (b) Same as Figure 10a, but for the averaged transit time in the tail region.
Figure 11: Distribution of the normalized equivalent length squared of N₂O for the period 2000-2019 (lines) and the percentage changes between 2080-2099 and 2000-2019 (colors). Only differences statistically significant at the 95% confidence level are shown.

Figure 12: (a) Color shadings are differences in temperature between 2080-2099 and 2000-2019. Lines are 2000-2019 mean. (b) Same as Figure 12a, but for the zonal wind.

Figure 13: (a) The scatter plot of the equivalent length squared of N₂O in the subtropical lower stratosphere (averaged in 10°-30° latitudes and 440-520 K) against topical upward mass flux at 70 hPa (black, left axis), and the scatter plot of the equivalent length squared of N₂O against the zonal wind in the subtropical UTLS (averaged in 10°-30° latitudes and 200-70 hPa, blue, right axis). (b) The scatter plot of the equivalent length squared of N₂O in the subtropical lower stratosphere against the mean age (black) and spectral width (blue) in the subtropical lower stratosphere (averaged in 10°-30° latitudes and 70-50 hPa).

Figure 14: (a) Comparison of the compact relationship between the mean age and N₂O in the northern hemisphere 100-50 hPa between 2000-2019 (black) and 2080-2099 (red). (b) Same as Figure 14a, but for the relationship between the mean age and the fractional release of CFC12.
Figure 15: Evolution of the stratospheric mass burden of three radioactive tracers with different decay rate (black solid), the mean age tracer (black dashed), CFC11 (green) and CFC12 (blue) in the 21st century relative to their respective 2000-2019 level.

Figure 16: (a) Changes of stratospheric air masses between the 2080 and 2000 age spectra relative to the total stratospheric mass burden as a function of transit time at 1-year interval. (b) Changes of stratospheric air masses between the 2080 and 2000 age spectra relative to their 2000 level.
(a) Mean Age (Year)

(b) Vertical Velocity (mm/s)

(c) Meridional Velocity (m/s)
Age Spectra 2000 10hPa

Difference 2080-2000 10hPa
(a) Decay Timescale vs. Mean Age

(b) Decay Timescale vs. Tropical Upwelling
Transient Time (Year)

Change Relative to Total Burden (%)

0 2 4 6 8 10 12 14 16 18 20

Percentage Change (%)

0 20 40 60

0 2 4 6 8 10 12 14 16 18 20