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Reply to RC2

Thank you for the time and attention you have given to our paper.

1) The relation between mean age of air and fractional release does not become clear without being familiar with prior studies. Here, the authors could greatly enhance the understandability of their argumentation by providing better explanations and/or examples. This is all the more important since the main conclusions (impact of horizontal mixing on MERRA-2 traces gas simulations) are partially drawn from the relationship. Statements in section 4.1.2 need more detailed information, e.g., Page 9, line 1: where does mixing lead to an increase of age? Only in the tropics? Or in the whole stratosphere? Why does this not change the fractional release? Also, it is hard to follow the argumentation since once of the major figures is not clear: Do figures 2c and d show the fractional release (as indicated in the caption and text) or the change of the fractional release (as indicated in the labeling of the y-axis)?

We have made several changes to clarify the relationship between mean age and fractional release.

First, in addition to fixing an error with section numbers, we have completely rewritten the section titled “mean age and fractional release”. This revision has received a friendly review from GSFC colleagues and we feel it is more clear. We have also revised Figure 2. We replaced line plots that compare the fractional release of N2O from GEOSCCM and GMI CTM at constant pressure and at single mean age, with a six panel figure, comparing contours of fractional release at fixed mean age for GMI CTM (two top panels, NH and SH) with a similar contour plot for GEOSCCM (middle panels, NH and SH). The bottom two panels compare the standard deviation of fractional release as a function of mean age for 1990 – 2000 with that obtained for 2005 – 2015 (SH and NH). In both hemispheres, this profile is nearly the same for both periods for GEOSCCM, but changes for GMI CTM. We make the point the physical consistency is guaranteed in the coupled model but not in the assimilation system where data insertion and large increments can introduce additional mixing (e.g., Schoeberl et al., 2004?, Douglass et al., 2004?).

The age spectrum for a stratospheric air parcel is the distribution of transit times between entry to the stratosphere to the parcel location for each of the elements that comprise the parcel. The mean age is the average of this distribution, and comparisons of simulated mean age with that obtained from measurements such as SF_6 (a long-lived trace gas that is increasing in the troposphere) provide information about the realism of the advective and mixing processes that control the parcel paths. *Hall et al.* [1999] used mean age comparisons to show lack of realism in the transport produced by various models that participated in Model and Measurements Intercomparison II [*Park et al*., 1999]. *Hall* [2000] *and Schoeberl et al.* [2000] used trajectory calculations to show on average, the oldest elements have risen to highest altitudes, thus evidence of loss of a long-lived gas in a parcel found in the lower stratosphere indicates that the age spectrum includes parcel elements that have experienced high altitude where source gas destruction occurs. These older elements of the age spectrum contribute to the mean age and determine the amount of destruction of long-lived gases. This relationship between the oldest elements of the age spectrum and the probability of destruction leads to compact, inverse relationships between mean age and gases with tropospheric sources. Indeed, aircraft observations show that the destruction of long-lived gases including chlorofluorocarbons, N_2O and CH_4 is related to the mean age obtained from SF_6. *Schauffler et al.* [2003] use aircraft observations to compute the fractional release f_r

$$f_r = \left(1 - \frac{X(\mathbf{x})}{X_i}\right)$$

where $X(\mathbf{x})$ is the mixing ratio of a source gas in a parcel at location **x** (latitude, altitude, pressure, time) and X_i is the mixing ratio at entry to the stratosphere, finding a near linear relationship between *fr* and the SF_6 mean age. *Waugh and Hall* [2002, and references therein] discuss the relationship between tracer distributions and transport timescales in the stratosphere, noting the connection between the BDC and the wave-driven quasi-horizontal mixing that control distributions of stratospheric trace gases. Because the fractional release depends on destruction of the source gas and therefore on the portion of the age spectrum that reaches high altitude where destruction is possible, the relationship between fractional release and mean age is a stronger test of the realism of simulated transport than the simple comparisons of mean age distributions. The older elements of the age spectrum that contribute to the mean age make the largest contribution to the fractional release. Mean age is negatively correlated with source gases and positively correlated with the products of their destruction (e.g., HCl and HNO_3).

In GEOSCCM the BDC is fully consistent with the planetary wave breaking that results in horizontal mixing and plays a role in driving the BDC. This consistency is not guaranteed in the MERRA-2 fields. Comparison of fractional release and its relationship to mean age with values obtained from observations tests the balance between horizontal mixing and vertical transport. *Waugh et al*. [2007] apply these concepts to the simulated amounts of inorganic chlorine (Cl_y) released from source gases in a CTM using different grid resolution and meteorological fields, finding large differences in Cl_y for the same mean age. Within the same CTM, different meteorological fields or different implementation of meteorological fields may produce the same mean age but different values for the fractional release because the mean transport pathways differ.

In GMI CTM the fractional release for fixed mean age varies substantially between 1990 and 2000 in both hemispheres (Figure 2a and b). In contrast, in GEOSCCM the fractional release at fixed mean age varies slowly (Figure 2c and d). The changing relationship between fractional release and mean age in the GMI simulation reveals decadal variations in the relationship between horizontal and vertical transport processes in MERRA-2. After about 2000, the small variations in *fr* for fixed mean age in GMI CTM are comparable to the variations obtained throughout the period for GEOSCCM. We illustrate this by comparing the ratio of the *fr* standard deviation to its mean for 1990 – 2000 and 2005 – 2015 for each simulation for both hemispheres (Figure 2 e and f). In GEOSCCM the ratio is about 2% for both time periods and for both hemispheres. For GMI CTM the ratio for 1990 – 2000 is between 6% and 8% for the age range 2 – 3.5 years in both hemispheres. For 2005 – 2015 the GMI CTM ratio is comparable to that obtained from GEOSCCM, although still larger than GEOSCCM in the southern hemisphere. The smaller values of *fr* at fixed mean age found during the early 1990s suggest that the balance between horizontal and vertical transport processes up until about 2000 is substantially different in both hemispheres in the first half of the 1990s than in subsequent years. This changing relationship contributes to the lower correlations of mean age with long-lived tracers in GMI CTM compared with GEOSCCM noted in 4.1.1 and also affects the relationship of mean age with reservoir gases discussed below.

2) It does not become clear why the GEOSCCM simulations are included. It seems that all the conclusions can be made without the use the CCM model runs, at least they are not mentioned in the discussion or conclusion section at all.

We feel that the role of GEOSCCM is made clear by the discussion of fractional release and replacement of Figure 2. In addition, we have added this sentence to the conclusions:

Although the relationship between the fractional release and mean age is expected to change in the midlatitude lower stratosphere if the BDC strengthens due to climate change [*Douglass et al.,* 2008], the observations are not consistent with the large changes in fractional release for fixed mean age obtained during the 1990s using GMI CTM compared with the changes in GEOSCCM.

3) *There seems to be some offset in the timing of the different characteristics described in the text. The shift in agreement between observations and CTM simulations of HNO3/HCl seems to appear between 2000 and 2005. However, the largest changes of fractional release relative to age of air seem to appear between 1990 and 1995 (if I understand Figure 2c correctly). Later in the manuscript this discrepancy between the two analyses is avoided by talking about a shift around 2000. Please clarify.*

The new Figure 2 makes it clear that the fractional release for fixed mean age settles down sometime in the early 2000's. The prior figure showing only a single line was less directly related to the column comparisons.

Minor comments:

1) *Page 6, line 24: MERRA should be MERRA-2*

   Changed

2) Page 11, line 21-27: Could this shift occurs also without a change in the fractional release – mean age comparison due to too old/young age in MERRA-2 before 2000? Should it be Figure 2c instead of 2b?

The referee has noticed the most difficult point in this paper. My first thought on seeing the comparison for the columns was that the age should be older. When I compared the CH4 in the 1990s – the sense of the comparison was that the age should be younger. This led me to consider the fractional release. I think that the summaries after discussing each data set support the final conclusions, the conclusion of each data set is meant to describe what can be drawn from that comparison by itself.

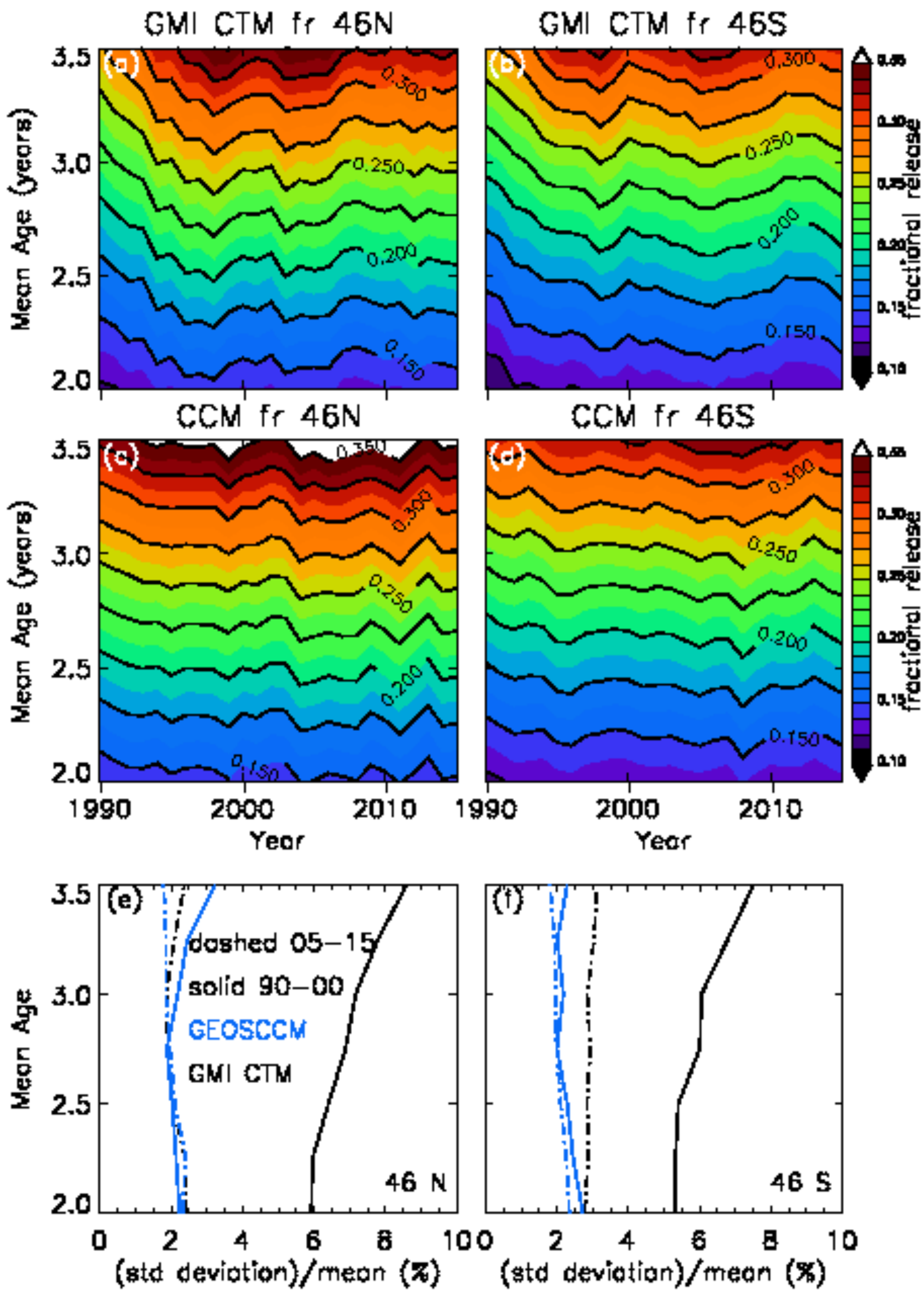

Figure 2: Contours of the evolution of fractional release for N_2O simulated by GMI CTM for 1990 – 2015 as a function of mean age at (a) 46°N and (b) 46°S. Same for the GEOSCCM at (c) 46°N and (d) 46°S. The ratio of the standard deviation of the N_2O fractional release to the mean value from GMI CTM (black) and from GEOSCCM (blue) averaged from 1990 – 2000 (solid) and from 2005 – 2015 at (e) 46°N and (f) 46°S.