

We thank the reviewers for their constructive and helpful suggestions. We have provided our responses to the reviewers' comments and believe that our manuscript is much improved as a result.

The main paper improvements are:

1. The AoA analysis was revised. We used the idealized linearly growing "surface" tracer proposed in the Age of air intercomparison project (Krol et al., 2018). Therefore, all figures that depict AoA were updated: the zonal mean plot and the comparing with observations (Figures 1b, 5-8).
2. Figure 2 was removed.
3. The script for calculating averages for 3D distributions was revised, therefore Fig. 1a and Fig. 3 were updated.
4. The script for calculating seasonal averages was revised, therefore panels at Fig. 2 (the diffusion velocity of SON and MAM are exchanged) and Fig. 4 (less spread, large values around the level of 35km) were updated.
5. Due to scripts revision, inaccuracies in figures were eliminated. Thus, consistency between vertical profiles and 2D distributions has improved. All figures show $\langle \delta \rangle$ values around -100 per meg in the high-latitudes, -70 per meg in the middle-latitudes and -50 per meg in tropics.

The reviewer's specific comments (shown in blue) are addressed below.

Anonymous Referee #1

This paper shows results of a CTM with molecular diffusion added to diagnose gravitational separation (GS) in the stratosphere. The different processes that control the diffusive velocity due to GS are discussed and the model distributions are compared to modeled age of air as well as observed profiles of derived GS and mean age. The model theory is clearly described and the comparisons to observations are interesting.

C1:

My main concern with the paper is the lack of clear evidence that the GS calculation adds significantly to our understanding of the stratospheric circulation. The conclusion section states that the model GS characteristics provide useful insights into structure changes in the UTLS, particularly over mean age.

Please see C3.

C2:

Mean age from measurements is ill-defined in the UTLS so it is not used as a measurement-based indicator of circulation changes in that region. There are a number of other trace gases, such as ozone, water vapor and CO, that are commonly used to define the transport and structural changes of the UTLS circulation. Even in the lower stratosphere above 100 hPa, where the seasonal cycle of CO₂ can impact mean age estimates, a careful consideration of the boundary conditions can alleviate much of the uncertainty. The simple lag technique used to calculate mean age in this paper is inappropriate for a trace gas with nonlinear growth such as CO₂.

The mean age calculation method was updated. For details please see C7.

C3:

The topic is appropriate for ACP and the model simulations of GS are novel. The benefits of these simulations to help interpret stratospheric circulation variability should be more clearly shown and described in my opinion.

As mentioned above, this work is a continuation and expansion of earlier research. So we summarized the limitations that need to be overcome and those properties that should be studied in future works (**p.16, l.19–23**): “However, due to the simplified approach and parameterizations, the presented simulation of the GS using the NIES model could not fully achieve the potential of 3D modeling. Modern reanalysis dataset and recently developed transport models effectively simulated the upper atmosphere can be employed to address

these issues. Since this work is the first in 3D modeling of GS, we believe this insight is useful for the scientific community working in the field of the UTLs studies.”

C4:

There are also a number of grammatical errors so I would suggest a more thorough proofreading of the paper is necessary.

English editing company managed by native speakers rechecked English grammar in the original manuscript and didn't find any other errors except shown below. Therefore, if errors are indicated, we will be grateful.

Specific comments:

C5:

Pg. 2, line 4: add TM abbreviation here since you refer to it on line 7.

Done.

C6:

Pg. 2, line 23: This is a confusing sentence, the mean meridional circulation is part of the BDC.

Revised as: “is perhaps best known for being a proxy for the rate of the stratospheric mean meridional circulation and the whole BDC.”

C7:

Eq. 7: This is an oversimplified way to calculate the mean age from a non-linearly growing trace gas such as CO₂. Why not compute the true model mean age using an idealized linearly growing tracer?

The age of air calculation method was updated. The text was revised (**p.6, l.10–13**): “Along with the $\langle \delta \rangle$ value we analyze the AoA (Fig. 1). For this, we used the idealized linearly growing “surface” tracer proposed in the Age of air intercomparison project (Krol et al., 2018). To fit with our analysis period we extended the original simulation period (1988–2014) to 29 years (1988–2016) with a shorter (10 years) spin-up, as less time required to reach equilibrium for the AoA analysis.”

C8:

Figs 2-4: Very little discussion of these figures in the text. Figure 2 is interesting but the scales are different on each plot and the terms are labeled 1-3 rather than by the physical

mechanism responsible for each term so it's difficult to understand what's the important take away. There is only one sentence describing Figure 3, that's not enough. Better to fully discuss the important features of the figure or it should be removed.

The scale of the figures is quite diverse, so a selection of common color bar is complicated.

Figure 2 was removed.

Discussion of the Figure 3 (now Figure 2) was revised **(p.7, l.1–10)**: “To estimate the contribution of atmospheric conditions to molecular diffusion, we consider the sum of the three terms in the square bracket of Eq. 5. Because the contribution of the first term (concentration gradient) is relatively small, the second term (originated from pressure gradient in Eq. 1) is the major contributor among the three terms. Therefore, sum of three terms can be approximated by the difference between the reciprocals of two scale heights (hereafter referred to as Li^{-1}). It has a dimension reciprocal to the length, and is interpreted as a measure of the efficiency of vertical molecular diffusion under gravity. In view of the essentially one-dimensional nature of GS, it is interesting to consider how Li^{-1} distributes in the troposphere and stratosphere. Figure 2 shows the latitude-height distribution of Li^{-1} averaged in each season for the case of $^{12}C^{16}O_2$. Here the positive values indicate that $^{12}C^{16}O_2$ molecules descend relative to major constituents. The temperature fields necessary for the calculation are taken from the JCDAS reanalysis. Since Li^{-1} is in inverse proportion to temperature, it is generally small in the troposphere and takes maxima in the cold region such as the tropical tropopause region and the winter time stratosphere.”

C9:

Pg. 11, line 20: change “part” to “altitudes

Done

C10:

Pg. 12, line 1: change “sampling” to “sample”

Done

C11:

Pg. 12, line 6: change “the model” to “a model”

Done

C12:

Pg. 16, lines 20-22: As mentioned above, these statements aren't necessarily supported in the paper.

See C3