

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 #2

The authors conducted numerical simulation of carbon dioxide isotopes, $^{12}\text{C}_{16}\text{O}_2$ and $^{13}\text{C}_{16}\text{O}_2$ by using the NIES global tracer transport model (CTM), and clarified its gravitational separation (GS) due to the difference in the molecular diffusion. The CTM, which is an offline 3-D passive tracer transport model based on isentropic vertical coordinates, was driven by the reanalysis dataset, JMA Climate Data Assimilation System (JCDAS). Results of the 3-D CTM were found to be much more realistic than the results of a 2-D transport model. The CTM showed the GS apparently increasing with increasing altitude and latitude in the stratosphere, and also suggested a unique relationship of GS with the age of air (AoA). This work made an important progress in understanding of the 3-D distribution of GS in the stratosphere. However, there are many incomplete discussions, particularly of physical interpretations of simulated results. I recommend to publish the manuscript in ACP after addressing the following comments.

C1:

Generally speaking, the turbulent eddy diffusion is much greater than the molecular diffusion in the troposphere. The eddy vertical diffusions may be enhanced by the large wave activity in the extratropical stratosphere. The authors should describe how the CTM treats the eddy vertical diffusion in the troposphere and stratosphere, and discuss how they affect the GS distributions in the stratosphere.

The eddy vertical diffusion in the troposphere is described by Belikov et al. (2013a): The parametrization of turbulent diffusivity follows the approach used by Hack et al. (1993), with transport processes in the planetary boundary layer (PBL) and free troposphere evaluated separately. Turbulent diffusivity above the top of the PBL is calculated from local stability as a function of the Richardson number and is set to a constant value of $40 \text{ m}^2 \text{ s}^{-1}$ under an assumption of well-mixed air below the PBL top. Three-hourly PBL heights are taken from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim Reanalysis.

Hack, J. J., Boville, B. A., Briegleb, B. P., Kiehl, J. T., Rasch, P. J., and Williamson, D. L.: Description of the NCAR community climate model (CCM2), NCAR/TN-382, 108, 1993.

Added (p.5, l.7–11): “The eddy vertical diffusion in the stratosphere is often neglected in CTMs. However, it should be considered along with molecular diffusion here. The turbulent diffusion coefficient is estimated from parameterizations of gravity wave dissipation (Lindzen et al., 1981) similar to the SOCRATES model. In general, the eddy diffusion mixes concentrations in the volume, reduces vertical stratification and thereby weakens the molecular diffusion effect, as discussed by Kockarts et al. (2002).”

Lindzen, R. S.: Turbulence and stress owing to gravity wave and tidal breakdown, J. Geophys. Res., 86, 9707–9714, 1981.

C2:

The CTM adopts isentropic vertical coordinates, where the diabatic heating assessed in the reanalysis is used to estimate the vertical velocity. I think the choice of isentropic coordinates is adequate, because its vertical motions are free from the gravity wave noises. Note that conventional pressure coordinates tend to overly express the vertical mixing due to gravity wave noises. A problem in this work is that the vertical velocity was assessed from the seasonal mean diabatic heating. It neglected the short-term temporal variation of actual instantaneous diabatic heating, and underestimated their contributions to the vertical diabatic mixing.

The NIES model was developed to simulate greenhouse gases in near-surface layers, so the priority was to reproduce processes in the troposphere (seasonal cycle, inter-hemispheric gradient, moisture convection, etc.). Modeling of the stratosphere was tuned on the basis of the global balance of the tracer and the reproduction of AoA (Belikov et al., 2011, 2013a,b). Climatological heating rate meets these requirements and reduced noisy perturbations in the stratosphere. Although a certain proportion of short-scale changes may be lost in this case, however, the vertical profiles of the parameters studied are reproduced quite confidently as shown in Table 3.

C3:

The characteristics of Brewer-Dobson circulation in a reanalysis have been recognized to vary significantly depending on the reanalysis, and to be subject to systematic errors of the reanalysis. In this experiment, the CTM was driven by using a reanalysis. The systematic errors of reanalysis may degrade the simulated GS. How do the authors think about this problem?

Indeed, due to the features (grid type, horizontal and vertical resolutions, dynamical core, advection algorithm and etc.) modern reanalysis show very different performance in reproduction of the BDC characteristics.

The following sentence added (**p.14, l.32 – p.15, l.2**): “Chabrillat et al. (2018) presented a consistent intercomparison of AoA according to five modern reanalyses (ERA-Interim, JRA-55, MERRA, MERRA-2 and CFSR) and found significant diversity in the distributions which were obtained with BASCOE transport model, depending on the input reanalysis. They have also found large disagreement between the five reanalyses with respect to the long-term trends of AoA. Thus, an ambitious multi-reanalyses approach is needed to distinguish what is robust in the current GS results from what is not.”

C4:

Figure 4 showed that the geographical distribution of $\langle \delta \rangle$ value is significantly different between the northern and southern hemispheres. According to the authors, the stronger polar vortex enhances the GS in JJA-SH compared with DJF-NH. Furthermore, the GS differences may be caused by the Brewer-Dobson circulation and horizontal diffusions on isentropic surfaces. The authors should clarify major mechanisms causing the actual differences in GS distributions.

Discussion of the Figure 4 (now Figure 3) was revised **(p.7, l.11 – p.8, l.5)**: “The enhancement of Li^{-1} does not readily result in a remarkable GS, because it is the difference of Li^{-1} between 13C16O2 and 12C16O2 that creates GS in our case. For all that, we could expect that the enhancement of Li^{-1} combined with the long stratospheric transit time in the polar stratosphere will be favorable for GS. Figure 3 compares the horizontal distributions of the seasonal mean $\langle \delta \rangle$ on 10 hPa pressure surface in polar projections. We can see remarkable GS (small values of $\langle \delta \rangle$) in both Polar Regions exhibiting surprisingly clear axial symmetry. In the present analysis, the physical processes that drive GS (Eq. 1) have been rearranged in the form of Eq. 5 to separate the contribution to GS in two factors, one the concentration gradient (the first term) and the other the temperature structure. A stronger seasonal variability of GS in the southern hemisphere is caused by changes in vertical pressure gradient (Eq. 1) reflected to those in scale height difference between species.”

C5:

Figure 10 showed that the $\langle \delta \rangle$ value decreases very rapidly after the age exceeding 4 years. It means that the GS is highly nonlinear to the residence time of “Age” in the stratosphere. We would like to know the mechanisms for the GS acceleration in layers with an age of 4 or more years in the model.

The following section is added **(p.14, l.13–15)**: “The $\langle \delta \rangle$ value decreases very rapidly after the age exceeding 4 years, as the molecular diffusion coefficient increases with increasing height due to its pressure dependence (Eq. 4), which causes the enhancement of gravitational separation with increasing height. The mechanism does not affect AoA significantly, in the stratosphere (Ishidoya et al., 2008, 2013; Sugawara et al., 2018). This emphasizes a nonlinearity in the GS-AoA relationship in the stratosphere.”