

To Referee 2:

Thank you very much for your significant and useful comments on the paper “Gravitational separation in the Stratosphere – A New Indicator of Atmospheric Circulation” by Ishidoya et al. We have revised the manuscript, considering your comments and suggestions. Our responses to your comments and suggestions are as follows (your comments are highlighted and followed by our answers);

**1. Page 4840, Line 12: change “air age” to “age of air”**

=>We have changed “air age” to “age of air” in the revised manuscript.

**2. Page 4840, Line 14: change “the gravitational separation” to “gravitational separation” Actually...do this throughout the paper. Change “the gravitational separation” to simply “gravitational separation” in most cases. Minor copyediting would help.**

=>We have changed “the gravitational separation” to “gravitational separation” throughout the paper in the revised manuscript, as suggested.

**3. Page 4841, Line 1 : delete “a” before “new concrete” and “the” before “gravi-“**

=>We have deleted “a” before “new concrete” in the revised manuscript.

**4. Page 4841, Line 3: Is this an effect on O<sub>2</sub> concentration, or on the O<sub>2</sub>/N<sub>2</sub> ratio?**

=> “O<sub>2</sub> concentration” has been changed to “O<sub>2</sub>/N<sub>2</sub> ratio” in the revised manuscript.

**5. Page 4842: could you explain in the text what your reference sample is? (i.e., is it like Standard Mean Ocean Water used to compute depletions for HDO?)**

=>Following sentence has been added in the revised manuscript to denote the reference air used in this study.

“Here,  $n$  means the amount of each substance, and “reference” is dried natural air filled in a high-pressure cylinder as a primary standard of our measurements (Ishidoya et al., 2003).”

**6. Page 4843, Discussion of Figure 1: The model shows a smooth decrease with**

**altitude (and I assume age of air). Why do the observations show coherent oscillations about the model line? Does that oscillation reflect seasonal variation in the strength of the BDC? Or seasonal variations in something about the input airmass or eddy mixing strength?**

=>Following sentences have been added in the revised manuscript associated with the fluctuations of the vertical profiles.

“Such a good correlation is not well interpreted yet, but causes may be due to seasonally-varying strength of the BDC or short-term variations in the stratospheric air transport.”

**7. Page 4846, Line 18: (and Figure 3) I’m still confused what “corrected for the gravitational separation” means.” Why does CO<sub>2</sub> need a correction? And are the corrected values for  $\delta(\text{O}_2/\text{N}_2)$  what you think the surface ratio would be?**

=> Following sentences have been added in the revised manuscript to make the meaning of the “correction” clearer.

“Equations (3) and (4) are similar to equation (3) in Etheridge et al. (1996) and equation (10) in Severinghaus and Battle (2006), respectively, which were used to evaluate gravitational separation of atmospheric components in polar firn. It is expected from the above discussion that the tropospheric air intruded into the tropical stratosphere is affected by gravitational separation during poleward transport. To exclude this effect, the data of  $\delta(\text{O}_2/\text{N}_2)$  and CO<sub>2</sub> concentration taken at altitudes above 18-25 km since 1999 were corrected for gravitational separation using equations (4) and (3), respectively, and their averages for each year are shown in Fig. 3.”

The relative molecular mass of CO<sub>2</sub> (~44) is larger than that of air (~29), so that the gravitational separation correction is needed to determine the CO<sub>2</sub> age precisely. The surface  $\delta(\text{O}_2/\text{N}_2)$  decreases secularly in contrast to CO<sub>2</sub> concentration, so that we can estimate the time lag between the stratosphere and the troposphere by using the  $\delta(\text{O}_2/\text{N}_2)$  corrected for gravitational separation.

**8. Figure 3: why does the tropospheric value decrease with time?**

=>The tropospheric O<sub>2</sub>/N<sub>2</sub> ratio decreases mainly due to fossil fuel combustion at the surface, and the sentence has been modified in the revised manuscript to show it explicitly.

**9. Page 4847, first paragraph: How is the age calculation with the  $\delta(\text{O}_2/\text{N}_2)$ ? Are you just shifting the solid line to match the dashed line? And, don't you then have to assume that the upper tropospheric value given here matches what is seen in the tropics at the tropopause?**

=> Following sentences have been added in the revised manuscript to make the calculation method clearer.

“In this study, the data of equatorial tropospheric  $\delta(\text{O}_2/\text{N}_2)$  are not available. Therefore, we simply calculated average time lags between the corrected middle stratospheric and upper tropospheric values of  $\delta(\text{O}_2/\text{N}_2)$  and  $\text{CO}_2$  concentration shown in Fig. 3, by shifting the relevant solid line to match the corresponding dashed line. The average time lag, thus obtained, is  $(3.9 \pm 0.9)$  years for  $\delta(\text{O}_2/\text{N}_2)$  and  $(4.0 \pm 0.4)$  years for the  $\text{CO}_2$  concentration, both values being consistent with each other. This agreement strongly suggests that the present gravitational separation correction is appropriate. The time lags obtained by this method are shorter by about 1 year than our “ $\text{CO}_2$  age” to be appeared in Figs. 5 and 6, which were calculated from the middle stratospheric and equatorial tropospheric  $\text{CO}_2$  concentrations, reflecting different tropospheric  $\text{CO}_2$  concentrations between the tropics and northern mid-latitudes.”

**10. Page 4848: In the model, do you look at the gravitational separation for the species measured with the cryogenic sampler? Why do you model with a different ratio?**

=> Following sentences have been added in the revised manuscript to explain the reason why we use the  $\delta(^{45}\text{CO}_2)$  to simulate the  $\langle \delta \rangle$  value instead of the  $\delta(^{15}\text{N})$  observed in this study.

“In this study, we used the isotopic ratios of  $\text{CO}_2$  to simulate the  $\langle \delta \rangle$  value instead of those of  $\text{N}_2$ ,  $\text{O}_2$  and Ar obtained from our measurements, since most chemistry-climate models including SOCRATES cannot calculate the concentrations and isotopic ratios of major components of the atmosphere. However, as seen from equation (4), the effect of gravitational separation on the isotopic ratio depends not on atmospheric component but on  $\Delta m$ . We also confirmed using SOCRATES that variations in the simulated vertical profile of  $\delta(^{46}\text{CO}_2)$  ( $\Delta m=2$ ) are just twice larger than those of  $\delta(^{45}\text{CO}_2)$  ( $\Delta m=1$ ). Therefore, gravitational separation of major stratospheric components can be discussed

using the  $\langle \delta \rangle$  value simulated for  $\delta(^{45}\text{CO}_2)$ .”

**11. Model discussion: You need a reference for the model. Is a BDC somehow imposed, or calculated by the model through some imposed eddy parameterizations. How does eddy mixing impact the ratios being examined? And, why is the model circulation “too fast”? Can you show that is the case (i.e., via examining propagation of the water vapor tape recorder...comparison with other**

=> We have added Park et al. (1999) and Khosravi et al. (2002) for references of the SOCRATES model. The purpose of model simulations in our study is a first attempt to examine basic structure of gravitational separation in the stratosphere, and we simply reproduce the changes in the BDC by arbitrarily changing the mean mass stream function. Differences of global mean age between various models, including SOCRATES, have been studied by “Model and Measurements Intercomparison II” in detail (Park et al., 1999). They found that almost all models show lower mean ages for the lower and middle stratosphere, compared to observations. They also suggested that the mean age should be increased by reducing the residual circulation magnitude and/or by enhancing the mixing of extratropical and tropical air. In this connection, the latter may partly explain the age underestimated by models, since the  $\text{CO}_2$  ages estimated from our balloon observations are consistent with those reported by Ray et al. (2010) using a Tropical Leaky Pipe (TLP) model with fast horizontal mixing in the lower stratosphere. These facts have been noted in the revised manuscript.

**12. Page 4849 (and figure 5): Could you explain how you did the average del profile? Is that done by averaging all the values shown in Figure 1 at a given level?**

=> We have rewritten the related sentence as follows in the revised manuscript to make the calculation method of the average  $\langle \delta \rangle$  profile clearer.

“To examine the temporal variations of the vertical gradient, we calculated an average vertical profile of  $\langle \delta \rangle$  by applying a linear least squares fit to all the profiles observed in this study and then obtained deviations of the respective  $\langle \delta \rangle$  values from the average profile.”

**13. Figure 6: Is the del value shown here from the model based on the  $\text{CO}_2$  ratio shown in equation 6? Does that give the same del you have calculated from  $\text{O}_2$ ,  $\text{N}_2$**

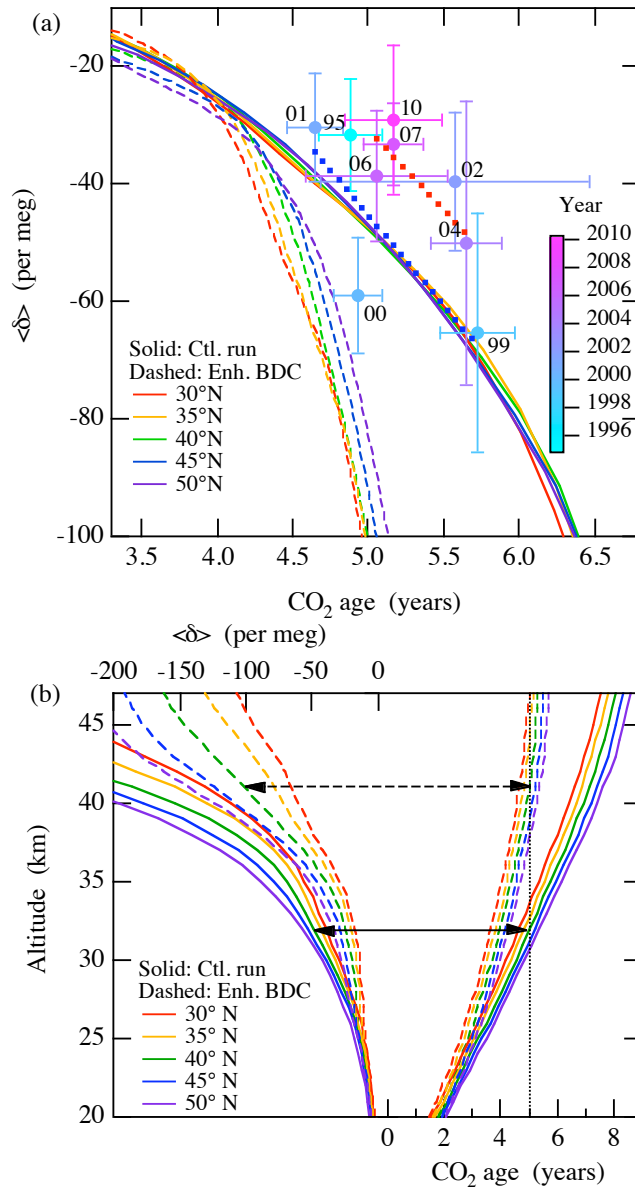
**and AR and I assume is what is plotted with the data points on figure 6? And, why is 2002 considered an outlier but 2000 is not?**

=>The  $\langle\delta\rangle$  values shown here from the model are based on the isotopic ratio of CO<sub>2</sub> as you pointed out, and they are considered to give the same  $\langle\delta\rangle$  from N<sub>2</sub> as the reply to the comment No. 10. The caption of Fig. 6 has been rewritten in the revised manuscript to make the reason clearer why the data in 2002 is considered to be an outlier. The error in the CO<sub>2</sub> age in 2002 is significantly larger compared to the other years, due to large variability in the vertical CO<sub>2</sub> profile observed in that year.

**14. Figure 6 (and Page 4850): Could you explain why the gravitational separation is enhanced when the BDC is faster? Are the parcels getting to an age of 4.25 years (where the lines split) following a drastically different path (possibly going much higher)? Is your imposed enhanced BDC anything like what is modeled in 3D climate models?**

=>To clarify the mechanism for the changes in the relationship between gravitational separation and age of air, the vertical profiles of the  $\langle\delta\rangle$  value and the CO<sub>2</sub> age for the Control Run as well as the Enhanced BDC (Fig. 6 (b)) and related discussion have been added in the revised manuscript (they have been also attached to the end of this file). It is clearly seen from Fig. 6 (b) that gravitational separation is weakened and the CO<sub>2</sub> age is decreased by enhancing the BDC. It is also found that the  $\langle\delta\rangle$  value of about -50 per meg and the CO<sub>2</sub> age of 5.0 years are found at 31-34 km over the northern mid-latitudes for Control Run, while Enhanced BDC shows about -100 per meg for the  $\langle\delta\rangle$  value and 5.0 years for the CO<sub>2</sub> age at 38-47 km over the same latitude region. This phenomenon is caused by a strong height dependency of gravitational separation due to the fact that the molecular diffusion coefficient increases with increasing height. Therefore, gravitational separation on the iso-age surface could be enhanced when the BDC is faster as shown in Fig. 6 (a). Our imposed enhanced BDC simulation is a simple mimic of the changes in the BDC simulated by recent 3D climate models. Almost all 3D climate models have projected an intensifying stratospheric circulation in global warming scenarios, but our attempt to estimate changes in the BDC by using observational data have suggested contradictory results, i.e., temporal changes in the relationship between the observed gravitational separation and CO<sub>2</sub> age (Fig. 6 (a)) are opposite of the changes in the BDC based on the 3D models simulation.

<Figure 6 (a) and (b) in the revised manuscript>



**Figure 6.** (a) Plots of the  $\langle \delta \rangle$  value at 29 km against the average values of CO<sub>2</sub> ages at heights above 18-25 km for the respective observations over Sanriku and Taiki, Japan (closed circles). Color bar and Arabic numerals near the symbols indicate the observation years. The results calculated using the SOCRATES model for Control Run (solid lines) and Enhanced BDC (dashed lines) are also shown. Blue and red dotted lines represent the results obtained by applying a linear regression analysis to the data

for the respective periods 1995-2001 and 2004-2010. It is noted that the result for 2002 is not used in the regression analysis, since the error in the CO<sub>2</sub> age estimated for that year is significantly larger compared to the other years, due to large variability in the vertical CO<sub>2</sub> profile observed in that year. It is also noted that the observed  $\langle\delta\rangle$  values plotted are the values obtained by linearly interpolating the measured  $\langle\delta\rangle$  values of the corresponding observations for 29 km, which is approximately the highest altitude covered by all our observations. (b) Vertical profiles of the  $\langle\delta\rangle$  values and the CO<sub>2</sub> ages calculated using the SOCRATES model for Control Run (solid lines) and Enhanced BDC (dashed lines). Black solid (dashed) line denotes the  $\langle\delta\rangle$  value for the CO<sub>2</sub> age of 5 years at 40°N for Control Run (Enhanced BDC).

We revised a related paragraph in our manuscript as follows.

“As seen in Fig. 6 (a), the relationships between the CO<sub>2</sub> age and the  $\langle\delta\rangle$  value for Control Run at northern mid-latitudes are fairly close to the observational results over Japan, which implies that both the CO<sub>2</sub> age and the  $\langle\delta\rangle$  value can be almost reproduced by SOCRATES. However, the relationships for Enhanced BDC are clearly different from those of Control Run, indicating that the CO<sub>2</sub> age and the  $\langle\delta\rangle$  value respond differently to changes in the stratospheric transport, i.e. gravitational separation for the air molecules with the same age is enhanced when the BDC is accelerated. To see such a behavior in more detail, vertical profiles of the two variables for Control Run are compared in Fig. 6 (b) with those for Enhanced BDC. It is clearly seen from this figure that gravitational separation is weakened and the CO<sub>2</sub> age is decreased by enhancing the BDC. It is also found that the  $\langle\delta\rangle$  value of about -50 per meg and the CO<sub>2</sub> age of 5.0 years are found at 31-34 km over the northern mid-latitudes for Control Run, while Enhanced BDC shows about -100 per meg for the  $\langle\delta\rangle$  value and 5.0 years for the CO<sub>2</sub> age at 38-47 km over the same latitude region. This phenomenon is caused by a strong height dependency of gravitational separation due to the fact that the molecular diffusion coefficient increases with increasing height.

It is not easy to detect a long-term change in the BDC only from the CO<sub>2</sub> age derived from spatiotemporally discrete balloon observations because of its year-by-year

variability superimposed on a secular trend. On the other hand, the results given in Fig. 6 indicate that not only the CO<sub>2</sub> age but also the  $\langle\delta\rangle$  value, as well as their relationship, is clearly changed when the BDC varies. As seen from Fig. 5, the observed year-by-year variability of the  $\langle\delta\rangle$  value is inversely correlated with that of the CO<sub>2</sub> age. This suggests that the influence of year-by-year variability is reduced by inspecting the two variables simultaneously and that a long-term change in the BDC can be detected as a change in the correlation between age and gravitational separation. Therefore, simultaneous observation of the  $\langle\delta\rangle$  value and the CO<sub>2</sub> age would provide more reliable information about a long-term change in the BDC than that of only the CO<sub>2</sub> age. It is actually found from our observational results shown in Fig. 6 (a) that gravitational separation for the air with the same age was slightly weakened with time for the period 1995-2010. This tendency is just the opposite of that expected from the Enhanced BDC simulation. Balloon and satellite observations (Engel et al., 2009; Stiller et al., 2012) reported that the CO<sub>2</sub> and SF<sub>6</sub> ages in the stratosphere over northern mid-latitudes showed no significant trend over the last 30 years, while the satellite measurements indicate that the SF<sub>6</sub> age might have increased for the period 2002-2010. Our long-term record of the middle stratospheric CO<sub>2</sub> concentration over Japan for the period 1985-2010 also shows a slight secular increase in the CO<sub>2</sub> age (our unpublished data, but the CO<sub>2</sub> age values for a limited time period of 1986-2001 are available from Engel et al. (2009)). These observational results on gravitational separation and the air age could imply that the BDC has not changed significantly or weakened slightly over the past 10-30 years, in conflict with the model prediction of an enhancement of the BDC due to global warming (Austin and Li, 2006; Li et al., 2008).”