

To Referee 3:

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;

Responses to major comments

1. To clarify what we infer from changes in gravitational separation ($\langle\delta\rangle$ value) about the changes in BDC, discussions about vertical profiles of the $\langle\delta\rangle$ value and CO₂ age for the Control Run as well as the Enhanced BDC (Fig. 6 (b)) have been added in the revised manuscript (they have been also attached to the end of this file).

Almost all 3D climate models have projected an intensifying stratospheric circulation in global warming scenarios. On the other hand, attempts to clarify changes in the BDC by using observational age data, including our CO₂ age presented in this manuscript, have yielded contradictory results. However, the long-term trend of age estimated from SF₆ and/or CO₂ data have been still ambiguous and inconclusive mainly because of somewhat large uncertainties and interannual variations and also sparseness of measurements. In this study, we found out the gravitational separation as a previously unknown stratospheric nature. The gravitational separation has different information from CO₂ age because of strong height dependency of the molecular diffusion. Therefore, we presented a new idea to detect a change in the correlation between age and the gravitational separation.

Gravitational separation and CO₂ age at the same altitude are weakened and decreased in response to the enhancement of BDC, respectively, as seen in Fig. 6 (b). Therefore, both the CO₂ age and the $\langle\delta\rangle$ value can be used to detect changes in the BDC as you pointed out. However, the observed CO₂ age and $\langle\delta\rangle$ value could show no significant long-term change due to year-to-year variability superimposed on their secular trends (see Fig. 5). Such the variability would affect significantly to the spatiotemporally discrete balloon-borne observations. On the other hand, the results

given in Fig. 6 also indicate the relationship between the $\langle\delta\rangle$ value and the CO₂ age will clearly change when the BDC is changed. Therefore, long-term changes in the BDC can be examined not only by the respective secular trends of the $\langle\delta\rangle$ value and the CO₂ age but also by the secular changes in the relationship between the $\langle\delta\rangle$ value and the CO₂ age. In addition, as seen from Fig. 5, the observed year-by-year variability of the $\langle\delta\rangle$ value is inversely correlated with that of the CO₂ 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 CO₂ age would provide more reliable information about a long-term change in the BDC than that of only the CO₂ age.

As you pointed out, the simulation by changing the mixing in the model will be important and probably give us different results. In future study, we will carry out the simulation and examine whether the changes in the relationship between the $\langle\delta\rangle$ value and the CO₂ age give information of the mixing such as the fast horizontal mixing in the lower stratosphere simulated by Tropical Leakey Pipe models (e.g. Ray et al. 2010).

2. As you pointed out, the “CO₂ age” in Fig. 5 is not consistent with the time lag from Fig. 3 because of different tropospheric time series. In Fig. 3, we simply calculated the time lag from the middle stratospheric values corrected for gravitational separation and the upper tropospheric values over Japan shown in the figure. We have recognized that the equatorial tropospheric $\delta(\text{O}_2/\text{N}_2)$ is needed to calculate the “ $\delta(\text{O}_2/\text{N}_2)$ age” corresponding to commonly-used “CO₂ age”, but unfortunately we don’t have the equatorial tropospheric $\delta(\text{O}_2/\text{N}_2)$ data. To make these facts clearer, we have added some sentences in the revised manuscript and changed the “ $\delta(\text{O}_2/\text{N}_2)$ age” and “CO₂ age” in section 3-3 to “time lag between the middle stratospheric and upper tropospheric values over Japan” to avoid confusion with our “CO₂ age” calculated using the stratospheric and the equatorial tropospheric data. The related revised sentences have been also attached to the end of this file.

As you also pointed out, temporal expanse of the differences between the middle stratospheric and upper tropospheric corrected $\delta(\text{O}_2/\text{N}_2)$ is also noticeable in Fig. 3.

However, we cannot make out the significance of the expanse of time lag between the middle stratosphere and the troposphere because the precision of the time lag calculated from the $\delta(\text{O}_2/\text{N}_2)$ is significantly worse than that of CO_2 concentration. Therefore, we have limited the use of the time lag from $\delta(\text{O}_2/\text{N}_2)$ only for the validation of the gravitational separation correction.

Responses to minor comments

1. Pg 4840, line 21: Might help to put approximate altitude of turbo pause.

=>The approximate altitude of turbo pause (~100 km) has been added in the revised manuscript.

2. Pg 4841, line 11: "intrusion from the troposphere"? How about "after it enters the tropical stratosphere from the troposphere".

=>We have changed "after its intrusion from the troposphere" to "after it enters the tropical stratosphere from the troposphere" in the revised manuscript, as suggested.

3. Pg 4842, I think the equations should be at the start of the paragraph when $\delta(^{15}\text{N})$ first used.

=>The paragraph has been modified in the revised manuscript to show the definition of $\delta(^{15}\text{N})$, $\delta(^{18}\text{O})$, $\delta(\text{O}_2/\text{N}_2)$, $\delta(\text{Ar}/\text{N}_2)$ and $\delta(^{40}\text{Ar})$ at the start of the paragraph.

4. Pg 4844: Can you include the equation 33 from Lettau (1951)? Accessing very oldarticles is not always easy.

=>The equation (33) in Lettau (1951) has been included in the revised manuscript as follows;

$$v_i = v_{i0} \exp\left(-\frac{\mu_i}{H} \int_0^z \frac{gT_0}{g_0 T} Q_i dz\right) \left[1 - \frac{\gamma_0}{v_{i0}} \int_0^z \frac{Q_i}{N d_{iN_2}} \exp\left(\frac{\mu_i}{H} \int_0^z \frac{gT_0}{g_0 T} Q_i dz\right) dz\right],$$

$$Q_i = \frac{d_{iN_2}}{d_{iN_2} + D}, \quad \mu_i = (m_i - m_{N_2})/m_{N_2}, \quad \text{and} \quad H = kT_0/m_{N_2}g_0, \quad (2)$$

where subscript i denotes the species of air. v_i , g and T (v_{i0} , g_0 and T_0) are the number concentration, the gravitational acceleration and absolute temperature at the altitude z

km (at the surface), respectively. N is the number density of air, and γ_0 is the strength of the continuous external source (or sink). m_i and m_{N_2} are the relative molecular masses of the species i and N_2 , respectively. k is Boltzmann's constant, and d_{iN_2} and D denotes the coefficient of mutual (i and N_2) molecular diffusion and the vertical eddy-diffusion coefficient, respectively.

5. Pg 4845, line 6: "will be described somewhere" is not very useful. If a publication on measurements not at least submitted then I think there needs to be some information in this manuscript (Appendix?).

=>Some additional information of the measurement has been added and the reference under preparation of a manuscript has been removed. The added sentences in the revised manuscript are as follows;

“During this period, the air in the flask with room temperature was introduced into an inlet system of the mass spectrometer equipped with a dual inlet system (Thermo Scientific DELTA-V). The sample air introduced from the flask was exhausted from the inlet system with a flow rate of 4 mL min^{-1} and only a smidgen of air was transferred to an ion source of the mass spectrometer through a thermally-insulated fused silica capillary, to continuously measure $\delta(^{15}\text{N})$, $\delta(^{18}\text{O})$, $\delta(\text{O}_2/\text{N}_2)$, $\delta(\text{Ar}/\text{N}_2)$ and $\delta(^{40}\text{Ar})$. The reference air was also introduced into the mass spectrometer by the same procedure using the other inlet. The pressure imbalance between the sample air and the reference air, as well as the influences of CO_2 concentration and O_2/N_2 ratio of the sample air on the measured values of the respective variables, were experimentally corrected (e.g. Bender et al., 1994; Ishidoya et al., 2003). The sample air and the reference air were alternately analyzed, taking 80 seconds for one cycle analysis of reference-sample-reference. By analyzing the same air sample repeatedly, the precision of measured values of $\delta(^{15}\text{N})$, $\delta(^{18}\text{O})$, $\delta(\text{O}_2/\text{N}_2)$, $\delta(\text{Ar}/\text{N}_2)$ and $\delta(^{40}\text{Ar})$ were estimated to be ± 8 , ± 20 , ± 5 , ± 10 and ± 70 per meg, respectively.”

6. Section 3.3 on: I found it a bit confusing to have "delta" as the average because easily confused with "delta" for different gases. I would suggest using delta with an overbar or "<delta>", as overbar or <> often used to indicate averages.

=>The “ δ ” has been changed to “δ” throughout the paper in the revised manuscript, as suggested.

7. Pg 4846 line 17-19: I am bit confused about the correction for gravitational separation. "using the above-mentioned equations" is a bit vague. I think this needs to be better described.

=>We have changed “using the above-mentioned equations” to “using the equation (4) and (3), respectively” in the revised manuscript, to specify the corresponding equations to the respective corrections for the $\delta(\text{O}_2/\text{N}_2)$ and the CO_2 concentration.

8. Pg 4849: What is meant by "magnitude depends on observation"?

=> “its magnitude depends on the observation” has been changed to “the vertical gradient is different depending on observation” in the revised manuscript to make the meaning clearer.

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

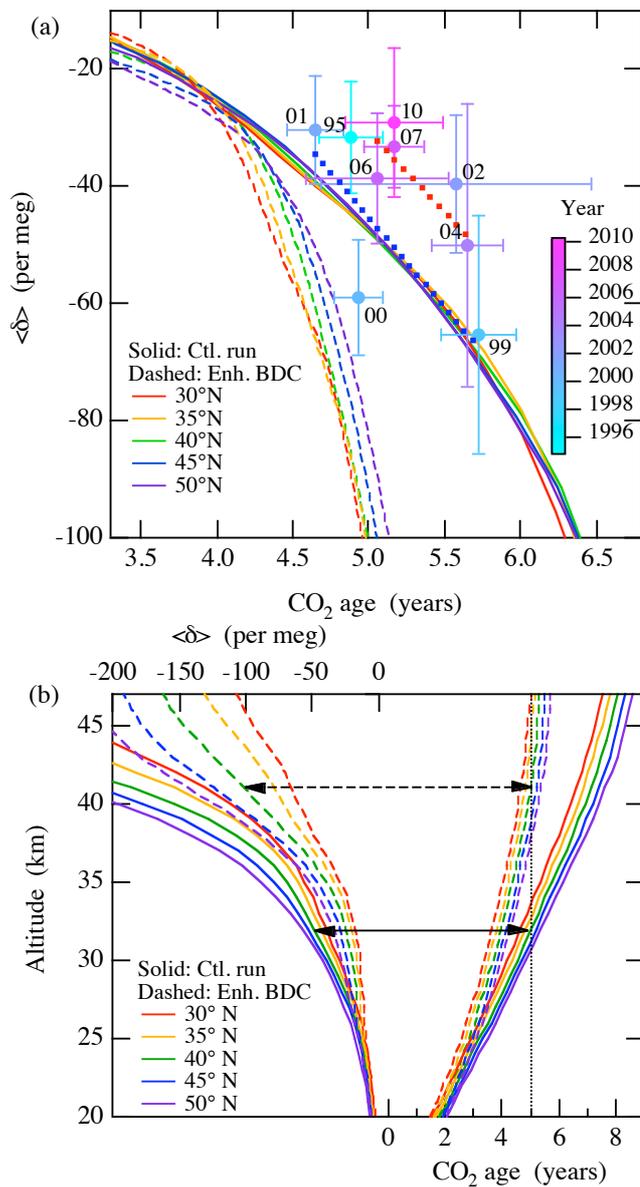


Figure 6. (a) Plots of the $\langle \delta \rangle$ value at 29 km against the average values of CO_2 ages at heights above 18-25 km for the respective observations over Sanriku and Taiki, Japan (closed circles). Color bar and Arabic numerals near the symbols indicate the observation years. The results 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 CO₂ age estimated for that year is significantly larger compared to the other years, due to large variability in the vertical CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ age is decreased by enhancing the BDC. It is also found that the $\langle\delta\rangle$ value of about -50 per meg and the CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ 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 CO₂ age would provide more reliable information about a long-term change in the BDC than that of only the CO₂ 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 CO₂ and SF₆ ages in the stratosphere over northern mid-latitudes showed no significant trend over the last 30 years, while the satellite measurements indicate that the SF₆ age might have increased for the period 2002-2010. Our long-term record of the middle stratospheric CO₂ concentration over Japan for the period 1985-2010 also shows a slight secular increase in the CO₂ age (our unpublished data, but the CO₂ 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).”

<Sentences about the time lag between the middle stratospheric and the upper tropospheric $\delta(\text{O}_2/\text{N}_2)$ in the revised manuscript>

It is also obvious in Fig. 3 that the stratospheric $\delta(\text{O}_2/\text{N}_2)$ value, corrected for the gravitational separation, decreases secularly following a similar temporal change in the upper troposphere over Japan, mainly due to anthropogenic fossil fuel combustion (Ishidoya et al., 2012). By comparing the stratospheric CO₂ and SF₆ concentrations with

their concentration histories in the equatorial troposphere, Waugh and Hall (2002) found that the middle stratospheric air is older by several years than the tropospheric air. If the gravitational separation effect is reasonably corrected by the above method, then the obtained stratospheric $\delta(\text{O}_2/\text{N}_2)$ values should yield a mean age similar to that derived from the CO_2 concentration data. 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 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 ± 0.9) years for $\delta(\text{O}_2/\text{N}_2)$ and (4.0 ± 0.4) years for the 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 “ CO_2 age” to be appeared in Figs. 5 and 6, which were calculated from the middle stratospheric and equatorial tropospheric CO_2 concentrations, reflecting different tropospheric CO_2 concentrations between the tropics and northern mid-latitudes.