To Prof. Severinghaus:

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;

To clarify the mechanism of the divergent behavior of gravitational separation with age of air between the Control Run and Enhanced BDC shown in Fig 6 (a), we have added Fig. 6 (b) showing the vertical profiles of the <δ> value and the CO_2 age for the Control Run as well as the Enhanced BDC and related discussion 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_2 age is decreased by enhancing the BDC. This indicates that mean circulation, represented by stream functions in SOCRATES model, increases with enhancement of the BDC. It is also seen from Fig. 6 (b) that the <δ> value of about -50 per meg and the CO_2 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 <δ> value and 5.0 years for the CO_2 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).

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

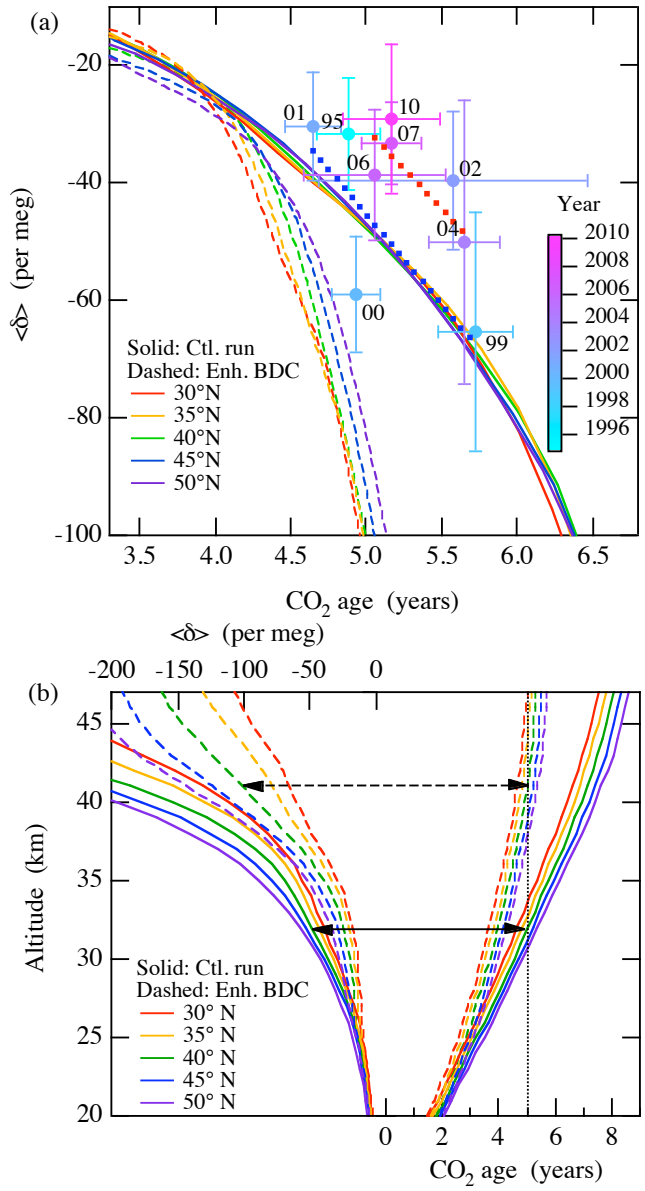

Figure 6. (a) Plots of the <δ> value at 29 km against the average values of CO_2 ages at heights above 18-25 km for the respective observations over Sanriku and Taiki, Japan (closed circles). Color bar and Arabic numerals near the symbols indicate the observation years. The results calculated using the 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 CO_2 age estimated for that year is significantly larger compared to the other years, due to large variability in the

vertical CO_2 profile observed in that year. It is also noted that the observed <δ> values plotted are the values obtained by linearly interpolating the measured <δ> values of the corresponding observations for 29 km, which is approximately the highest altitude covered by all our observations. (b) Vertical profiles of the <δ> values and the CO_2 ages calculated using the SOCRATES model for Control Run (solid lines) and Enhanced BDC (dashed lines). Black solid (dashed) line denotes the <δ> value for the CO_2 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_2 age and the <δ> value for Control Run at northern mid-latitudes are fairly close to the observational results over Japan, which implies that both the CO_2 age and the <δ> 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_2 age and the <δ> 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_2 age is decreased by enhancing the BDC. It is also found that the <δ> value of about -50 per meg and the CO_2 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 <δ> value and 5.0 years for the CO_2 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_2 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_2 age but also the <δ> 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 <δ> value is inversely correlated with that of the CO_2 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 $<\delta>$ value and the CO_2 age would provide more reliable information about a long-term change in the BDC than that of only the CO_2 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_2 and SF_6 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_6 age might have increased for the period 2002-2010. Our long-term record of the middle stratospheric CO_2 concentration over Japan for the period 1985-2010 also shows a slight secular increase in the CO_2 age (our unpublished data, but the CO_2 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)."