Reply to the referee #2

Our point-by-point response is typed in *italics: referee's comments*, roman: authors' response. Text in the revised manuscript is shown in red with page and line numbers of the new revised manuscript in squared brackets.

This paper extends the investigation in earlier papers, by several of the same authors, of the gravitational separation of various species in the stratosphere, and its possible implications for atmospheric transport. What is novel, and welcome, about this paper is its focus on tropical data. In principle, the paper appears to be worthy of publication, subject to attention to a few issues, outlined in what follows.

Gravitational separation does indeed, as the authors claim, provide a new perspective on stratospheric transport, but it is not made very clear just what that perspective is. Some "ad hoc" experiments are illustrated in the 2D model, in which transport parameters are changed, but things would be made much more clear if there were a theoretical exposition of the problem. For example, one could use simplified models to show how separation would manifest itself in the presence of upwelling alone, or vertical eddy diffusion alone. These would not reproduce the real world, but would provide some theoretical baseline to strengthen understanding of what a more complete model shows. One further shortcoming of the model perturbation experiments discussed in section 3.4 is that the tropics are considered in isolation from the rest of the globe. The authors may get better fits with the tropical data by changing parameters (upwelling, mixing) but that could be at the expense of agreement in the extratropics.

The relationship with the vast literature on stratospheric transport could be better illustrated by citing some of this literature more extensively than has been done. Some suggestions are outlined in the following.

We would like to thank the referee for many critical and constructive comments. As the referee suggested, the physical processes of gravitational separation will be clarified by using a simple model, such as one-dimensional model, rather than 2D model. Indeed, we used 1D steady-state model in the previous study (Ishidoya et al., 2013) and we have also developed 1D dynamical diffusion model with vertical eddy diffusion and advection flux. Those were useful to know basic properties of gravitational separation, such as time constant. We would like to publish them elsewhere. In this study, we focused on the different sensitivities between gravitational separation and mean age of air to possible perturbations of the tropical upwelling and the eddy diffusion. Because the model study of gravitational separation in the stratosphere has just begun, further investigation will be needed. For the better understanding of gravitational separation, we also started 3-D model simulations.

We have obtained a lot of knowledge of gravitational separation and the molecular diffusion processes from studies on the polar firn air, because theoretical basis is the same. To make clear the theoretical background of gravitational separation, we have added an equation that is commonly used in firn air study.

We have tried to address all of referee's comments as detailed below.

Other issues, as they arise in the text (page, line):

(1,32): Determination of the BD circulation from age observations has been explicitly discussed in a recent paper by Linz et al. (Nature Geosci., 2017).

We have added many references to introduction and discussions for mean age, including Linz et al. (2017). We have added some sentences as follows.

[p2, L9-14] Bönisch et al. (2011) suggested that the increased upwelling in the tropics after 2000 enhanced the lower stratospheric transport from the tropics into the extra-tropics. From an analysis of the ERA-Interim dataset, Diallo et al. (2012) also showed a negative trend over the 1989–2010 period in the lower stratosphere below 25 km. Linz et al. (2017) discussed the strength of the meridional overturning circulation of the stratosphere by using satellite observation data of SF_6 and N_2O, and suggested that a mesospheric SF_6 loss is important for age estimation using SF_6 mole fraction in the upper layer.

(2,10): The separate effects of circulation and mixing on age distributions are discussed in Garny et al. (J. Geophys. Res., 2014) and Linz et al. (J. Atmos.Sci., 2016). The strength of the circulation determines the horizontal gradient of age, rather than age itself; both mixing and circulation determine the vertical structure.

We have added these studies as references as follows.

[p2, L17-19] Therefore, to discuss a change in the mean age estimated using the clock tracer, it is important to separately evaluate the respective effects of mean circulation and mixing processes on the air age (Garny et al., 2014; Ploeger et al., 2015; Linz et al., 2016).

(5,31): Is it obvious that gravitational separation depends on mass number difference, rather than, say, mass ratio? If this is a theoretical prediction, please describe it.

We have revised and added some sentences and equation to make clear the theoretical background about the relationship between gravitational separation and mass number difference as follows.

[p6, L28-32] From a theoretical investigation of the molecular diffusion in polar firn air, the magnitude of the gravitational separation is proportional to mass number difference (Etheridge et al., 1996), which can be expressed as,

$$\Delta\delta = \Delta \mathrm{m} \times \Delta\delta_0 \qquad (2)$$

Here, Δm and $\Delta\delta_0$ are the mass number difference and the difference of δ values for Δm=1, respectively.

(6,3): The claim that Figure 3 suggests a linear relationship seems a little exaggerated. There are basically only two points (to be sure, there are 3, but two of them are very close together). Empirically, one could fit any number of curves to the data shown. If there is an a priori expectation of a straight line (from theory, or from more extensive observations) then you should say so. Further, the line does not appear to pass through the origin, which surely it should?

As the referee pointed out, the proportionality between the molecular separation and mass number difference is theoretically expected and usually observed in polar firn air. In accordance with the theoretical equation (2) described above, the regression line in Figure 3 has been changed to pass through the origin. We have also added some sentences about small deviations from the regression line as follows.

[p7, L6-8] It is not clear what caused the small deviations of $\Delta\delta$ from the proportional relationship shown in Figure 3. The thermal diffusion is one of the plausible causes, but its effect on our observational data taken by using our traditional cryogenic sampler was negligibly small (Ishidoya et al., 2013).

(8, 25) and elsewhere: Nowhere is it acknowledged that the authors are trying to draw conclusions from data that are highly localized in time, and whose representativeness is therefore open to question. So, e,g., the differences with the Brazil data may due to temporal, rather than spatial, variations. In general, the limitations of the temporal sampling should be prominently acknowledged.

As the referee suggested, we have revised a sentence to acknowledge the limitations as follows.
[p10, L7-9] This would be partly due to the different time and observation location, although the balloon data observed in the equatorial mid-stratosphere are still relatively sparse and not representative in time and space.

(9,18): The differences between CO2- and SF6-based age calculations are also discussed in, e.g., Hall and Waugh (J. Geophys. Res., 1998), Ray et al. (J. Geophys. Res., 2017), Linz et al. (Nature Geosci., 2017).

We have revised and added some sentences by adding these references as follows.
[p11, L4-5] The difference in the CO_2 and SF_6 ages has also been discussed in previous studies (Harnisch et al., 1998; Hall and Waugh, 1998; Strunk et al., 2000, Andrews et al., 2001).
[p12, L8-9] More recently, Ray et al. (2017) also reported that the SF_6 age in the stratosphere must account for a potential influence from the polar vortex air.
[p12, L11-14] Linz et al. (2017) compared the MIPAS SF_6 age with the N_2O age calculated with the N_2O data from the Global OZone Chemistry And Related trace gas Data records for the Stratosphere (GOZCARDS), and showed that the MIPAS SF_6 age is larger than the N_2O age in the tropics. The CO_2 and SF_6 ages observed in this study are consistent with the N_2O age rather than the MIPAS SF_6 age, although the observation period is different.

(10,1): Since the CO2 seasonal cycle, like the water vapor "tape recorder" signal, propagates into the tropical lower stratosphere as a decaying sinusoid in the vertical, I do not understand why "the age difference should be larger in the lower stratosphere . . .". It depends on time of year of the data being compared.

As the referee pointed out, our description was incorrect. We have corrected and added some sentences to make this clear as follows.
[p11, L16-26] For an ideal clock tracer that has increased or decreased monotonically in troposphere, $x(\Gamma, t)$ will be a single-valued function of Γ, which allows us to determine the mean age of air from the clock tracer mole fraction. On the other hand, if the CO_2 seasonal cycle is still significantly large at the observation altitude, it is not necessarily guaranteed that $x(\Gamma, t)$ is a single-valued function of Γ, depending on the season. In such a case, the CO_2 age will be underestimated or overestimated, depending on the time of year, and it is difficult to estimate the CO_2 age precisely from the mole fraction at that altitude. If that is the case, then the difference between the CO_2 and SF_6 ages caused by the CO_2 seasonal cycle might be significant in the season when the CO_2 mole fraction takes seasonal maxima and minima in the upper troposphere and the lower stratosphere. However, our results showed good agreement between the CO_2 and SF_6 ages in the TTL and the lower stratosphere. This is probably due to the fact that the seasonal CO_2 variation in the equatorial upper troposphere takes an intermediate concentration value in February, a level between its maximum and minimum (Sawa et al., 2008).

(11,15): If it is the mass dependence of Dmi that matters most, could you show us what that dependence is?

The molecular diffusion coefficient, D_{mi}, strongly depends on the atmospheric pressure. Since the mean free path of a specific molecule increases with decreasing pressure, D_{mi} increases rapidly with increasing altitude. We have revised a sentence as follows.

[p13, L12-14] In addition, the separation effect by molecular diffusion is enhanced with increasing altitude due to the rapid increase of the molecular diffusion coefficient, D_{mi}.

(11,25): Given the relatively small variation of T in the stratosphere, does this term matter much in practice?

As described in this paragraph, we neglected the thermal diffusion flux in our model calculation (i.e. α_{Ti} =0). This term is related with total number density and the atmospheric scale height and derived from the hydrostatic equation (Banks and Kockarts, 1973). Although the effect of temperature variations on the gravitational separation is not evaluated in this study, its effect was included in our calculations and it would be small at least in the stratosphere. Seasonal variations of gravitational separation shown in Figure 7 might include small temperature effect, while the seasonal change of atmospheric circulation would affect dominantly.

Figure 7: Why are the altitude scales different on the two frames? And can you comment on the negative ages in the northern lower stratosphere?

We have corrected the altitude scale in Figure 7. Age of air calculated here was adjusted so that the age values at 17 km are equal to CO_2 age observed at 17.2 km (0.4 years). Because SOCRATES does not have a good resolution for the tropospheric modeling (1 km for the vertical coordinate), it seems that the vertical differences of the CO_2 mole fraction around the tropopause are not resolved. In addition to this, the vertical transport is so fast in troposphere, which resulted that the CO_2 mole fraction was almost constant vertically in the tropics. Therefore, the CO_2 mole fraction around and just above tropopause in the northern hemisphere seems to be overestimated in our model.