Author Responses to the Referee #2 Comments

Thank you very much for your significant and useful comments on the paper "Kinetic fractionation of noble gases in the stratosphere over Japan" by Sugawara et al. We have revised the manuscript, considering your comments and suggestions. Details of our revision are as follows. The line numbers denote those of the revised manuscript.

The paper by Sugawara et al. presents observation of noble gases in the stratosphere and discusses the effect of kinetic fractionation in explaining observed vertical gradients. The paper is well written and presents interesting data with a solid interpretation of the effects controlling the vertical gradients and in particular the effect of gravitational settling caused by kinetic fractionation of different isotopes of the noble gases. Overall, I found the paper well written, the data of high quality and the interpretation thorough. I found the section 3.3. about the two-dimensional model somewhat difficult to follow, but this may also be caused by my lack of expertise in this area. The sections 3.4. and 3.5. could be sharpened by deriving clearer conclusions, e.g. on magnitudes of changes that could be detectable. I also suggest summarizing clearer conclusions in the abstract. I recommend the paper for publication after addressing the issues mentioned here and some modifications as detailed below.

As you pointed out, we have revised abstract and conclusions to make it clear that an impact of a changing Brewer-Dobson circulation on the noble gas elemental ratios in the modern troposphere would be hard to detect. Please see the last item of specific comments for details.

Specific comments:

1. *160 and following: what is the rf gas which was used?*

Lines 166-167:

We have added a sentence:

"Here, "rf" is the reference gas which is dried natural air filled in a high-pressure cylinder (cylinder no. CRC00045) (Ishidoya and Murayama, 2014)."

2. *172: please clarify how the difference was calculated: is this the mean absolute difference (MAD)?*

Lines 176-177:

We have revised as follows:
"The mean absolute difference of the values in 2007 was 50 ± 24 per meg."

3. *201: how was the CO2 mole fraction corrected for gravitational separation, how large is this effect? Please briefly explain here, even if you reference other literature.*

We have added some sentences to explain how we corrected CO_2 mole fraction for gravitational separation.
Lines 225-229:
"The correction of the CO_2 mole fraction for gravitational separation is non-negligible and can be estimated by $C \times (m - m_{\mathrm{air}}) \times \langle \delta_G \rangle$, where C is CO_2 mole fraction, m and m_{air} are the respective mass numbers of the molecule and air, $\langle\delta_G\rangle$ is the average gravitational separation (Ishidoya et al., 2006; Sugawara et al., 2025). The maximum depression of the CO_2 mole fraction due to the gravitational separation amounts to about 0.4 μmol mol^{-1} at the altitudes over 30 km, assuming C = 400 μmol mol^{-1} and $\langle\delta_G\rangle = -60$ per meg."

4. *245: In the case of 29N and 28N the delta-values and the normalised data values are equal. Neverthe, when comparing to normalised values, I suggest to denote the value with a subscript n.*

Line 289 and relevant descriptions, Figure 4:
All relevant descriptions of $\delta(^{29}N_2/^{28}N_2)$ in the main text and Figure 4 have been revised. Also, we added a short sentence to make it clear as follows:
"$\delta_n(^{29}N_2/^{28}N_2)$ (= $\delta(^{29}N_2/^{28}N_2)$)"

5. *242-257: would it make sense to summarize these values in a table?.*

Table 1 and Line 290:
As you pointed out, we have summarized δ_n values in Table 1 and add a sentence:
"The average values of $\delta_n(X/Y)$ at altitudes above 30 km are summarized in Table 1."

6. *436: I think it would be good to distinguish more clearly between molecular diffusion and eddy diffusion. In my understanding, the latter would certainly not lead to any kinetic separation and*

should be distinguished more clearly.

We have added some sentences to the beginning of this paragraph to clarify the difference between molecular and eddy diffusions:

Lines 481-484:

"To investigate the kinetic fractionation, the relative contributions of molecular and eddy diffusion are important. In an environment dominated by molecular diffusion alone, gravitational separation according to the mass number difference of molecules occurs, whereas in an environment dominated by eddy diffusion alone, separation according to molecular species is not expected to occur."

Section 3.4.: I understand that the overall conclusion from this is that an impact of a changing Brewer-Dobson circulation on the tropospheric isotope ratio would be hard to detect. The authors have focussed on a slowing-down of the BDC, however, models consistently predict an increasing BDC. It might be worthwhile discussing the detectability of an increasing BDC.

We completely agree with your comment. We have added some sentences to abstract and conclusions to make it clear that an impact of a changing Brewer-Dobson circulation on the tropospheric noble gases in the modern atmosphere would be hard to detect, except for Ar/N_2 ratio. Considering RC#1, we also performed additional simulations of the glacial condition and discussed about influences on the noble gas elemental ratios in ice core samples in Sect. 3.6. We added as follows:

Lines 28-30 (abstract):

"In the modern atmosphere, it is difficult to detect the long-term change of the stratospheric circulation from noble gases, except for Ar/N_2 ratio, in the troposphere. However, it was suggested that changes in the stratospheric circulation during glacial and interglacial cycles may have affected the noble gas elemental ratios in ice core samples."

Lines 741-744 (conclusions):

"At present, it is difficult to detect variations of noble gases elemental ratios in the troposphere caused by variations in stratospheric circulation, except for $\delta(^{40}Ar/^{28}N_2)$. However, when considering longer timescales such as glacial-interglacial cycles, variations in stratospheric circulation may have a significant impact on ice core data."

We have conducted additional simulations of a decreasing mean age scenario (enhanced RMC scenario). The resulting trend is the opposite of that of the weakened RMC scenario, but the magnitude is almost the same. We have added some sentences as follows:

Lines 581-584:

"We have also conducted simulations of enhanced-RMC scenario so that the mean age of air decreased by 0.15 years decade^{-1} at an altitude of 35 km over the northern mid-latitudes. The resulting trends in δ_{Ω} at the ground surface and mid-stratosphere were the opposite of those of the weakened-RMC scenario, but their magnitudes were almost the same. Similar results have been also shown in trends simulated for $\delta(Ar/N_2)$ by Ishidoya et al. (2021)."