Kinetic fractionation of noble gases in the stratosphere over Japan, Sugawara et al., EGUsphere 2025

Main objective of manuscript: Evaluate the vertical distribution of noble gases (Kr, Xe, and Ne) over the stratosphere, and processes governing it.

Main approach: Measurements of noble gases elemental and isotopic composition at different stratospheric altitude collected using balloon borne cryogenic air samples over Japan. Combined with two dimensional atmospheric model simulation (SOCRATES).

Main results: Elemental ratios increase, and isotopic ratios decrease for all noble gases with increasing altitude.

Implication: Kinetic fractionation occurred in the stratosphere because of difference in molecular diffusivity, can be a new tool to diagnose stratospheric mass transport processes.

Review

Major Comments:

- Section 2.3: Mean age of air: this section was a little hard to follow and could be improved with more information related to the statements made in this paragraph. Below are examples of lines where more clarification would be very helpful:
    - Line 178-179: "*certain relationship between the gravitational separation of the major atmospheric components and the mean age of stratospheric air*".
      What the relationship is, is unknown to the reader, and should be stated clearly.

    - Line 180-183: "*We also measured the mole fractions of CO2 and SF6 in our stratospheric air samples…. Because this method of estimation has already been described in previous studies… only a brief description is presented here*."
      It should be stated clearly why CO2 and SF6 are species specifically chosen to measure the age of air. That information is currently not available here and may not be known to all.

    - Lines 188-200: "*The mean age was estimated…. ratio of moments to be 1.25years*"
      This section is mostly unclear to me, although my expertise in this is limited. Some clarifications would be helpful though. First, along with clarifying why CO2 and SF6 mole fractions are chosen specifically, clarifying what the convolution method is, would also be very helpful. Specifically, clarifying what the tropospheric reference curve and the hypothetical age spectrum represent physically/temporally.
      Additionally, describing where the ratio of moments relationship comes from and its physical significance would be helpful for the reader. It seems important in this context. Also, why the 1.25years is the choice made here should be explained. The authors mention

that this was reported in a previous study but, clarifying why they think that this is accurate to use, would be helpful.

- Section 3.1:
  - General suggestion on Figure 2: This figure is an important figure for this manuscript as it shows the measurement results of this study. The figure is hard to read and decipher both in print and on the computer. The legends are too small to read, and it would be more useful to use different color/bigger markers to show these findings. The mode result lines are also hard to distinguish in the midst of all the measurement results.

    I would recommend improving the readability of this figure, as it reports the major findings of this study. Additionally, it would be useful to have a visual representation of the uncertainty in these measurements, either as errorbars in the figures or just as a legend that represents the uncertainty of the data.

  - Line 213-215: "*relative to the values observed in the lowest layers of the balloon observations…. negligibly small*"

    Clarify why this would be the case.
  - Line 216: "*clearly apparent... decreased with increasing altitude".*

    Although this statement is broadly true and the data shown in figures 2 &3 clearly highlight that, the elemental and isotopic composition of all 3 noble gases in figures 2&3 highlight excursions around specific altitudes. This is also apparent in all the data. For Xe, its between 20-25 km, Kr 25-30km, and Ar 15-20km.

    This does not seem included in the first section of the results, although they seem quite consistent. It would be useful to also note these in the results, and including them in the ensuing discussions.
  - Line 228: "*The fluctuations of the Xe isotopic ratios were irregular and larger than those of the Ar and Kr isotopic ratios"*

    Why?

- Section 3.2: I thought this section is really well written and clear, and easy to understand and follow. Lines 379-385 particularly do a good job of summarizing simply the key findings of this section.

- Section 3.4:
  - Lines 505-507: " *if Brewer Dobson circulation strengthened over time… reduce the mean age of stratospheric air*."

    Does this change refer to the BDC as a whole? If my understanding is correct, there is shallow arm of the BDC that causes mass exchange between the statosphere and troposphere across the 380 K isentrope, and a deeper arm of the BDC that exchanges mass with greater altitudes of the atmosphere.

    Does the model result distinguish between these? Would a difference between this matter? Presumably if only the shallow branch changed, it would have a different magnitude of impact on the troposphere than if the deeper branch changed? Would it be possible to disentangle this information from the model results? There may be other existing 3-D modeled results (for e.g. https://link.springer.com/article/10.1007/s00382-

006-0162-4) that could shed more insights on this. This would be relevant and helpful information to have I believe.

- Lines 511-520: It is unclear to me why the authors chose to only simulate increased mean age of air in this study, if it is unclear whether the mean mid-stratospheric age in increasing or decreasing. It would make more sense to simulate both increased and decreased mean ages, or explain more clearly why this choice was made.

- Line 528-530: "*significant influence on estimates of ocean heat content..*"

  Is there any way to get some first order constraints on this? For e.g., would it be possible to make a statement like "if stratospheric circulation changed by X%, we would expect it to have Y% impact on estimates on long term ocean heat content" or something along these lines? This seems like a very important constraint to have on this proxy. Noble gas thermometry for mean ocean temperatures from ice cores (for e.g.: https://www.researchsquare.com/article/rs-5610580/v1) seem to fundamentally assume no other processes are affecting it. If there is indeed an effect from circulation, it would be very important to know how large this effect could be, and how that translates to effects on MOT reconstructions.