Responses to Referee 1:

Thank you very much for your significant and useful comments on the paper "Secular change in atmospheric Ar/N_2 and its implications for ocean heat uptake and Brewer-Dobson circulation" by Ishidoya et al. We have revised the manuscript, considering your comments and suggestions. Details of our revision are as follows;

Main Points

1) **Surface observations. TKB has the longest and most dense data record. The observed temporal variations (Figure 2) give a compact annual cycle. In contrast, the sparser observations at the other 3 sites show much more variability (large amplitude variations). What are the reasons for this? Is it a measurement issue or possible real atmospheric variations?**

Lines 154-161: We consider the variability is a measurement issue. As you pointed out, the uncertainties of seasonal amplitude at COI, HAT and SYO are found to be larger than ±7 per meg of the uncertainty expected from the repeated analyses of the same flask air sample (Fig. 3). This would be due to the fact that the uncertainty of each analysis value of the standard air (±5.3 per meg, black dots in Fig. 1), which represents short-term (month-to-month timescale) stability of our $\delta(Ar/N_2)$ scale*, is superimposed on the uncertainty of each analysis of the flask air sample and continuous measurement. Therefore, a mean squared error expected for the observational data from the flask air sample is about ±9 per meg, which is comparable to the uncertainties for the seasonal amplitudes in Fig. 3. Corrections of the $\delta(Ar/N_2)$ values at HAT and COI data prior to September, 2015 and January, 2019 (lines 94-105) could also be contributing to the uncertainties at the sites.

*It is noted that ±1.6 per meg is expected for the long-term (interannual timescale) stability of our $\delta(Ar/N_2)$ scale, as in Fig. 1.

2) **Observed long-term trend. Even the two longest data records (8 years) are short for deriving accurate long-term trends. The trend fit is not explained clearly. The model leaves variations which are > 36 months but the trend is quoted as if**

it is a linear term? The fits to Ar/N2 and OHC in figure 2 are far from linear (but do vary with the long-term variability in the temperature data). I am concerned that the paper is reporting values in the abstract which imply a long-term linear trend, which is (quantitatively) not obvious from the plots.

Lines 140-147, 216-232 and Fig. 5: The sentences and figure have been added to explain how the secular trend were obtained. We have added descriptions for the digital filtering technique and clarify the definition of the interannual variation and secular trend in the present study (Lines 140-147). We regard the average linear increasing/decreasing trend throughout the observation period as the secular trend. The methods used to extract the secular trends at TKB and HAT are described in Lines 216-232 and Fig. 5.

3) **Atmospheric modelling. An updated 2-D model has been used to model surface Ar/N2. There is no evaluation of the model in the stratosphere using available profiles of Ar/N2 to show that the higher altitude gravitational separation is modelled realistically.**

Lines 281-282, 407-420, Figs. A1 and A2: Considering your suggestions, the sentences and figures have been added to show the meridional distribution of the $\delta(Ar/N_2)_\Omega$ calculated using the updated SOCRATES model and its comparison with the stratospheric $\delta(Ar/N_2)$ over Japan and equatorial region observed in our previous studies.

4) **Imposed trend in the BDC. The stratospheric BDC is complicated with deep and shallow branches. A trend in the circulation is imposed in the model and the resulting trend at 35 km is shown. First, more information on the impact at other altitudes should be shown (e.g. latitude-height cross section of the impact on age-of-air). Second and more importantly, the use of the Engel et al paper to support a trend in AoA, which is converted to a correction of surface Ar/N2 is unjustified. Engel et al use a series of sparse balloon observations of CO2 and SF6 to derive an AoA trend (from 24-35km) up to 2005 – so there is no overlap with the observation period in this paper. Moreover, the error bar on their trend is very**

large and the title of their paper gives the headline message of 'AoA unchanged within uncertainties'. Therefore, I cannot see how the imposed trend of 0.02 yrs/yr can be justified as the best estimate which gives the correction used in the abstract.

Lines 293-304, 421-430 and Fig. A3: The sentences and figure have been added to discuss the annual mean meridional distribution of the AoA trend (yrs yr^{-1}) calculated using the updated SOCRATES model for weakened BDC simulation (lines 421-430 and Fig. A3), considering the first comment. For the second comment, we agree with you that the Engel et al. does not support the 0.02 yrs yr^{-1} trend. In addition, we have recognized the simulations using the 2-D model in the present study, made by arbitrarily changing the MMC only with fixed horizontal mixing, are not enough to represent the mechanism to drive AoA change in the real atmosphere but it does served as a kind of sensitivity test. Therefore, we have clarified the limitation of the 2-D model and changed the reference from Engel et al. (2009) to Diallo et al. (2012) to support to use 0.02 yrs yr^{-1} trend (lines 293-304). Of course, we know the data periods of Diallo et al. (2012) are not also overlap with the observational period in this study. However, we consider our simulation is worthwhile as a sensitivity test since it constitutes the first step to investigate the effect of gravitational separation of the whole atmosphere on the surface $\delta(Ar/N_2)$.

5) **Ocean model. This is a crude approach (as acknowledged by the authors) and it leads to statements in lines 187-193 that the model maybe too simplistic (i.e. not suitable) and that other factors may need to be considered. Overall, this part of the analysis seems incomplete therefore.**

Lines 196 and 205-215: As you pointed out, the one-box ocean model is not enough to evaluate responses of the atmospheric $\delta(Ar/N_2)$ to changes in the air-sea heat flux in detail. Unfortunately, we cannot use better ocean model to calculate spatiotemporal variations in the air-sea heat flux. Therefore, we have stated the limitation of the one-box ocean model clearer. We have also added a reference showing the renewal time of permanent pycnocline water in the North Pacific, which would make the readers imagine that the secular trend of the $\delta(Ar/N_2)$ for 8-years in the present study mainly reflects the OHC change except deep ocean. We recognize the OHC changes estimated in the present study, based on the observed 8-years $\delta(Ar/N_2)$ trend combined with the 2-D atmospheric model and the one-box ocean model, are insufficient to suggest some revisions of the OHC from ocean temperature measurements. Nevertheless, it would of interest to see if it is possible to obtain a scientifically "meaningful" OHC change based on the firstly-reported secular $\delta(Ar/N_2)$ trend and the new concept of $\delta(Ar/N_2)_\Omega$.

Other Specific Points

1) **Line 21. The uncorrected trend of 0.75 +/- 0.30 per meg yr^{-1} is also consistent with trend derived from ocean temperature, at the limits of the error bars. The correction is not needed, which is what is implied by the text.**

We leave the discussion as they are since the main aim of the correction is not an estimation of the precise OHC change comparable to that from ocean temperature measurements but to suggest that we cannot ignore the secular trend of $\delta(Ar/N_2)$ caused by changes in gravitational separation in the whole atmosphere.

2) **Line 132. 'Fig 2 in 3-1'. What does this mean?**

Lines 133-136: The sentence has been modified to make the meaning clearer.

3) **Line 163. 'Increase rates' (also in caption of Figure 4 and label axis). This should be referred to simply as 'rate of change'. The positive values will imply an increase.**

Lines 180-181 and Fig. 4: The words "increase rate" has been changed to "rate of change", as suggested.

4) **Line 180 'boldly'. This is the wrong word. You must mean something like 'crudely'.**

Line 197 and 381: The words "We boldly" has been changed to "As a first approximation we", considering your comments (line 197). The words "boldly assuming" has also been changed to "crudely assuming" (line 381).

5) Line 183. ‘modern’ – better to say ‘present-day’

Line 200: The word “modern” has been changed to “present-day”, as suggested.

6) Line 184. Insert ‘total atmospheric mass. . ..’.

Line 201: The word “total mass” has been changed to “total atmospheric mass”.

7) Line 187 ‘drives’. Delete s.

Line 204: The word “drives” has been changed to “drive”. Thank you for pointing out.

8) Line 216. ‘molecular mass number’ -> ‘relative molecular mass’.

Lines 254-255: The sentences have been modified. The $m_{A\text{-}air}$ is not a relative molecular mass, but a reciprocal average of m_A and m_{air}.

9) Line 221-223. Add a reference or make it clear that you are referring to this work.

Line 264: We have added references.

10) Line 238. ‘decrease’ (no s)

Line 280: The word “decreases” has been changed to “decrease”.

11) Line 240 (and later). I think ‘idealised’ is better than ‘virtual’.

Line 283, 286 and 287: The word “virtual” has been changed to “idealised”, as suggested.

12) Line 250. Don’t need to say ‘increase’.

Line 293: The word “increase rate” has been changed to “rate”.

13) Line 251. Give the dates that the Engel et al study covered (but see main comment above).

Lines 299-300: We have added the dates that the Fritsche et al. (2016) covered and revised the sentence considering your main comment. Fritsche et al. is the study updated from Engel et al. (2009).

14) Line 255. The word ‘obtained’ is wrong. The perturbation to the model circulation was forced arbitrarily. Change to e.g. ‘forced’?

Line 302: The word “obtained” has been changed to “forced”, as suggested.

15) Line 258. Nb ‘simplistic’ is a negative term which means that the model is too simple to be suitable.

The sentence has been revised, and the word “simplistic” has not been used in the revised sentence.

16) Lines 265. Change ‘increases’ to ‘is estimated to increase’.

Line 310: The words “atmospheric $\delta(Ar/N_2)$ increases” have been changed to “atmospheric $\delta(Ar/N_2)$ is estimated to increase”.

17) Line 267. Change ‘expected’ to ‘is estimated’.

Line 312: The word “expected” has been changed to “estimated”.

18) Line 269. Insert ‘are non-negligible trends compared. . .’.

Line 313: The words “are non-negligible compared” have been changed to “is a non-negligible trend compared”.

19) Line 274 Insert ‘. . . estimated effects..’.

Line 317: The words “the effects” have been changed to “the estimated effects”, as suggested.

20) Caption for Fig. 2

We have removed the incorrect sentence “All data are corrected for the scale drift of the primary standard air shown in Fig. 1 (b).” in the caption for Fig. 2 in ACPD paper since we have not applied such correction to the data.