Responses to Referee 2:

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;

The manuscript is challenging to review as it touches on a wide range of topics but information provided is often (too) sparse. As far as I can see, the main points of the manuscript are:
(i) Ar:N2 measurements are accurate enough to detect trends, which was not possible previously (Line 37).
(ii) Variations in the ratio over a decade are well correlated with OHC variations as reported by NOAA/NCEI.
(iii) There is a quantitative mis-match if only OHC is considered.
(iv) A model simulation with imposed change in the stratospheric BD circulation suggests that the near surface Ar:N2 ratio is as sensitivite to the BD circulation as to OHC variations (for the observed magnitude of changes in either of these).
(v) If one assumes that the OHC measurements and their impact on Ar:N2 are correct, one infers that over the period of measurements (2012-2019) the BD circulation slowed down.
The authors discuss the last point in some detail, and seem to be concerned that the sign of the change in the BD circulation is the opposite of a supposedly long term strengthening of the BD circulation. There is no reason, however, why a short period such as the one studied here should show the long term trend given the large interan nual variability of the stratospheric circulation. I recommend to shorten this somewhat misleading discussion (the authors are most likely not observing "trends"), and instead strengthen the points (i)-(iv).

Thank you for your comments and suggestion. As you pointed out, the observational period of 8-years in the present study is not enough to discuss the long-term trend of the BDC. In addition, the troposphere, and not only the ocean, does not mix perfectly on a timescale of a year, and that the surface-ocean temperature anomalies would be a large source of interannual variation on a yearly timescale for the observed δ(Ar/N_2) in the near-surface air. Therefore, we recognize the possibility that the secular trends observed at the surface and simulated by the 2-D model do not represent a long-term trend but is a part of the large interannual variation. Nevertheless, it would of interest to see if it is possible to obtain a scientifically "meaningful" OHC change based on the firstly-reported secular δ(Ar/N_2) trend at the surface and the new concept of δ(Ar/N_2)$_{\Omega}$. Taking these into consideration, we have revised the related sentences throughout the manuscript, and stated clearly in the revised sentences that the 2-D model simulations are not enough to represent the mechanism to drive AoA change in the real atmosphere but it does serve as a kind of sensitivity test (the revised sentences have been highlighted by blue color : for example, lines 216-232, 293-304, and Appendix A and B).

Specifically:

(a) This reviewer is not sufficiently familiar with the measurement technique to be able to assess claim (i); the data as presented in Figures 2,3 and 4 does look, however, reasonable; as does the calibration (Figure 1). Nonetheless, I hope that another reviewer may be in a position to comment.

Thank you for your comments. As you expected, another reviewer commented on this matter and we have added the sentences not only to describe the uncertainties for seasonal cycles (lines 154-161) but also to show the detail method to extract the secular trend of δ(Ar/N_2) observed at the surface stations (lines 216-232).

(b) The time filtering and frequency separation method needs to be better explained - based on the information provided in this manuscript, it is not possible to reconstruct what precisely the authors have done. This is an extremely important part of the argument as the claim is that seasonal variations are due to local SST variations, whereas the longer-term changes are due to global OHC changes (i.e. the seasonal cycle at TKB and HAT differ (Fig 3), but their longer-term variations (Fig 4) are highly correlated). Also, the analysis then uses largely

only TKB and HAT - but why not also show COI in Figure 4? Please use 2 different colors for AHT and TKB in Figure 4a - the separation with "+" and "o" does not allow to see the differences (Looking at Figure 2; it is surprising that these two timeseries align as well as shown in Figure 4).

Lines 140-147 and Fig. 4: The sentences have been added to explain the time filtering and frequency separation method more in detail. Also, we have added the data at COI in Fig. 4 and used three different colors for TKT, HAT and COI, as suggested.

(c) The SOCRATES model is not sufficiently described. Please provide more information on vertical and horizontal resolution, time steps, and parameterization of atmospheric transport and mixing. Please show the model's mean vertical profile of the Ar:N2 ratio, along with the measurements thereof in the stratosphere. Do you have other information (e.g. CO2 mixing ratio profile) that could serve as validation?

Lines 245-246, 281-282, 295-296, 391-405, 407-430, Figs. A1, A2 and A3: Considering your suggestions, the sentences and figures have been added to show the detail descriptions of the SOCRATES model (lines 391-405), and meridional distribution of the $\delta(Ar/N_2)_\Omega$ calculated using the model and its comparison with the stratospheric $\delta(Ar/N_2)$ over Japan and equatorial region observed in our previous studies (lines 407-420, Figs. A1 and A2). We have also added the sentences and figure to discuss the annual mean meridional distribution of the AoA trend (yrs yr^{-1}) calculated using the SOCRATES model for weakened BDC simulation (lines 421-430 and Fig. A3). 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 serve as a kind of sensitivity test. Therefore, we have clarified the limitation of the 2-D model (lines 293-304). We consider our simulation is worthwhile as the sensitivity test since it constitutes the first step to investigate the effect of gravitational separation of the whole atmosphere on the surface $\delta(Ar/N_2)$.

Additional points

1) **Figure 5: My understanding is that this is simply an idealized calculation for 10 years; if so, please change x-label (labels 2000 - 2010 suggest that this is specifically for this period; which is confusing also because OHC changes are taken from this period, see next item).**

Figure 6: Thank you for your comments, we have revised the figure as suggested. It is noted that the number of the figure has been changed from “Fig. 5” to “Fig. 6” since we have added a new figure as Fig. 5 to discuss the secular trends of the $\delta(Ar/N_2)$ observed at TKB and HAT.

2) **OHC: Why do you discuss OHC change for 2000-2010 (Line 267)? Why are you not using the data for 2012-2019? The text here is confusing; Figure 4b suggests you use the data for the correct period.**

Lines 310-312: The sentence has been modified to discuss OHC change for 2012-2019, considering your suggestion.

3) **Please indicate the baseline period for OHC (i.e. the figures shows departures from a baseline, not absolute OHC as one would think based on labels and caption).**

Lines 177-179 and caption of Fig. 4: The sentence and figure caption have been modified to show the baseline period for OHC, as suggested.

4) 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.