

Responses to Referee 3:

Thank you very much for your significant and useful comments on the paper “Secular change in atmospheric Ar/N₂ and its implications for ocean heat uptake and Brewer-Dobson circulation” by Ishido et al. We have revised the manuscript, considering your comments and suggestions. Details of our revision are as follows;

The one minor comment I have is that the one-box ocean model is somewhat over-emphasized, and perhaps the description could be simplified and shortened, because we know very well that the troposphere does not perfectly mix on a timescale of a year, nor does the ocean mix perfectly on a timescale of one year, so it is not really surprising that a one-box ocean model fails to match the observations of Ar/N₂ at surface stations around Japan. I understand that the authors constructed the one-box model as a “straw man”, to be shot down, but they could greatly simplify and shorten the discussion, while making it clear that they do not expect to be able to match their observations with a one-box model of the ocean. In fact, some readers may be confused, the way the authors have written about this one-box model (they seem to imply that they expected it to be able to match their observations). But of course the surface-ocean temperature anomalies are by far the largest source of noise on yearly timescales, for tracers like Ar/N₂, measured in near-surface air. They should clarify that a true tropospheric- average Ar/N₂, measured from aircraft (which is of course too expensive and so is prohibitive), would be needed to actually compare Ar/N₂ observations with modeled Ar/N₂. (So in some sense they are raising a false expectation with their one-box model exercise.)

On the whole, this is a groundbreaking and important paper, and will make a fine contribution to ACP. This is clearly excellent, highest-quality work!

Lines 25-28, 196 and 205-215: Thank you very much for giving high evaluation to the present study. As you pointed out, the one-box ocean model is not enough to evaluate responses of the atmospheric $\delta(\text{Ar}/\text{N}_2)$ to changes in the air-sea heat flux in detail. 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 $\delta(\text{Ar}/\text{N}_2)$ 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 $\delta(\text{Ar}/\text{N}_2)$ 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 $\delta(\text{Ar}/\text{N}_2)$ trend and the new concept of $\delta(\text{Ar}/\text{N}_2)_\Omega$.

Minor editorial comments:

(1) line 35 “. . .Argo floats”.

Line 39: The words “Argo float” have been changed to “Argo floats”, as suggested.

(2) line 47 - “. . .since it has been believed that the gravitational separation near the surface is too small to be detected...” This isn’t really accurate, in the sense that there is no need for the gravitational settling process IN THE troposphere to be significant, in order for the stratosphere to affect the troposphere. Perhaps instead you could say something like, “. . .since it has been believed that the gravitational settling in the stratosphere is fairly small and constant in time, along with the fact that the troposphere has 10x more molecules than the stratosphere.”

Or perhaps you meant to say, “. . .the gravitational separation signal from the stratosphere is too small to be detected at the surface.”? This is accurate.

Line 50-51: The words “since it has been believed that the gravitational separation near the surface is too small to be detected” have been changed to “since it has been believed that the gravitational separation signal from the stratosphere is too small to be detected at the surface”, as suggested.

(3) line 51 “. . .long-term changes in the Ar/N2 ratio near the surface are expected to be extremely small..”

Line 54: The word “near the surface is” have been changed to “near the surface are”.

(4) line 53 again, it sounds like you are saying there there is gravitational separation near the surface, but this is not accurate. Maybe you mean the stratospheric gravitational separation signal near the surface?

Lines 56-57: We have revised the phrase as “a very small secular change in the stratospheric gravitational separation signal near the surface may...”, considering your comments.

(5) line 58 “. . .secular trend of the Ar/N₂ ratio. . .”

Line 62: The words “trend of Ar/N₂ ratio” have been changed to “trend of the Ar/N₂ ratio”

(6) line 60 “Atmospheric Ar/N₂ has been observed. . .” (there is no need to include “ratio” here)

Line 65: We removed the word “ratio”, as suggested.

(7) line 74 “. . .and we usually use the average of 550 data values as the reported Ar/N₂ ratio obtained from the continuous observations (about 11 hours of averaged data).”

Lines 77-79: The sentence has been modified, considering your suggestion.

(8) Line 101 “. . .glass flasks.”

Line 105: The words “glass flask” have been changed to “glass flasks”.

(9) Line 102 “per meg units as follows.”

Line 106: The words “per meg unit” have been changed to “per meg units”.

(10) Line 112 “. . . corresponds to an uncertainty of. . .”

Lines 115-116: The words “the uncertainty of” have been changed to “an uncertainty of”.

(11) Line 116 “. . . to +1,800”

Line 120: The word “1,800” has been changed to “+1,800”.

(12) Line 122 “..but they did not correlate..”

Line 125: The words “but did not” have been changed to “but they did not”.

(13) Line 124 “. . . must have been superimposed. . .”

Line 127: The words “must have superimposed on” have been changed to “must have been superimposed on”.

(14) Line 126 “. . . due to a temperature. . .”

Line 129: The words “due to temperature” have been changed to “due to a temperature”.

(15) Line 138 “. . . by a fundamental sine-cosine. . .”

Line 141: The word “fundamental” have been changed to “a fundamental sine-cosine”.

**(16) Line 143 “. . .reach seasonal maxima. . . due to the larger relative...”
(enhancing is not really the correct word, sine the solubility is a physical constant
and cannot be enhanced)**

Line 151: The word “enhancing” have been changed to “due to”.

(17) line 146 “Similar increases. . .”

Line 162: The word “increase” has been changed to “increases”.

(18) line 150 “..found a seasonal. . .”

Line 166: The words “found seasonal” have been changed to “found a seasonal”.

(19) line 154 “. . .than the 14 ± 6 . . .”

Line 170: The words “than 14 ± 6 ” have been changed to “than the 14 ± 6 ”.

**(20) line 160 this sentence is very long and hard to read. Perhaps you could
simplify and shorten it. Also the verb “are shown” comes at the very end of the
sentence, which is awkward. Instead you could write, “Variations in the 0-2000
m global OHC are shown (Fig. 4), reported by. . .”**

Lines 175-180: Considering your suggestion, we have revised the sentence as “Variations in the 0-2000 m global OHC are shown (Fig. 4), reported by the National Oceanographic Data Center (NOAA)/National Centers for Environmental Information (NCEI) (updated from Levitus et al. 2012, https://www.nodc.noaa.gov/OC5/3M_HEAT_CONTENT/). The OHC values are shown as anomalies from the baseline value observed in mid-1980s.

In the figure we also plot a interannual variation of the OHC values obtained by using the same digital filtering technique used in Fig. 2, and globally averaged surface temperature anomalies (Japan Meteorological Agency, http://www.data.jma.go.jp/cpdinfo/temp/nov_wld.html).

(21) line 180 “We boldly modeled. . .” This is not usual scientific language. Perhaps say “As a first approximation we modeled . . .”

Line 197: The words “We boldly modeled” have been changed to “As a first approximation we modeled”, as suggested.

(22) line 187 “..was estimated to drive. . .”

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

(23) line 196 “As mention in the Introduction,”

Line 235: The words “in Introduction” have been changed to “in the Introduction”.

(24) line 201 “. . .caused by changes in gravitational separation.” It is not clear whether you intend to say gravitational separation that occurred in the stratosphere, or gravitational separation that occurred in the troposphere. Please clarify. For example, “. . .caused by changes in stratospheric gravitational separation that influence the whole troposphere.” OR “. . .caused by changes in gravitational settling within the troposphere itself.”

Lines 239-241: The sentence has been modified considering your suggestion as “Therefore, we need to explore the possibility of tropospheric $\delta(\text{Ar}/\text{N}_2)$ variations caused by changes in the stratospheric gravitational separation that influence whole troposphere.”.

(25) line 261 “..seesaw..” perhaps you mean to say “inverse”?

Line 306: The word “seesaw” has been changed to “inverse”.

(26) line 265 “inputted” is an awkward word. Perhaps instead use “heat is added to a . . .”

Line 310: The words “is inputted” have been changed to “is added to”.

(27) line 269 “ is non-negligible. . .”

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

(28) line 275 “the derived. . .”

Line 317: The word “derived” has been changed to “the derived”.

(29) line 342 “. . .there is no method so far to validate OHC based on ocean temperature measurements.” You will find a lot of oceanographers objecting to this statement. I would suggest you temper it somewhat, to something like “there is no method yet to adequately measure OHC via ocean temperature observations in the full-depth volume of the ocean”.

Lines 386-388: The sentence has been modified, as suggested.

(30) 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.