

## ***Interactive comment on “Secular change in atmospheric Ar/N<sub>2</sub> and its implications for ocean heat uptake and Brewer-Dobson circulation” by Shigeyuki Ishidoya et al.***

### **Anonymous Referee #3**

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This is an excellent paper. It breaks open a whole new idea: namely that the stratosphere's well-known gravitational separation (in which heavy gases such as argon settle out relative to lighter gases such as N<sub>2</sub>), may have a small but perceptible impact on the Ar/N<sub>2</sub> ratio of the troposphere, by the simple logic of whole-atmosphere mass conservation. Variations in the amount of gravitational settling over time, due for example to variability in the Brewer-Dobson circulation in the stratosphere, are proposed to have a very small impact of opposite sign in the troposphere. Excellent quality long-term data from air monitoring stations is shown to back up this claim. However, it remains to be demonstrated that the signals seen in the data are entirely due to the stratospheric effect; other minor processes such as imperfect mixing within the troposphere

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and shallow-ocean temperature anomalies may play a partial role, and so further work is needed to track down these possible issues. But the authors have made a very important contribution and this paper should be published with only minor revisions.

The authors furthermore expand and improve on the Blaine and Keeling approach to decadal-scale ocean heat content variations deduced from tropospheric Ar/N<sub>2</sub> measurements. To my eye, this is the best Ar/N<sub>2</sub> data ever published on this critical topic of ocean heat, and the authors are to be commended for this important contribution to understanding the Earth's net heat budget under the current anthropogenic forcing. These type of data may eventually make it possible to better constrain the climate sensitivity to a doubling of atmospheric CO<sub>2</sub>, because the ocean may be masking the true top-of-the-atmosphere energy imbalance, due to the fact that something like 93% of this excess energy goes into the ocean and so may be escaping our detection. The present paper is an important step in this fundamentally critical direction.

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/N<sub>2</sub> 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/N<sub>2</sub>, measured in near-surface air. They should clarify that a true tropospheric-average Ar/N<sub>2</sub>, measured from aircraft (which is of course too expensive and so is prohibitive), would be needed to actually compare Ar/N<sub>2</sub> observations with modeled

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Ar/N<sub>2</sub>. (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!

Minor editorial comments:

line 35 "...Argo floats"

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 51 "...long-term changes in the Ar/N<sub>2</sub> ratio near the surface are expected to be extremely small.."

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?

line 58 "...secular trend of the Ar/N<sub>2</sub> ratio. ..."

line 60 "Atmospheric Ar/N<sub>2</sub> has been observed. ...." (there is no need to include "ratio" here)

line 74 "...and we usually use the average of 550 data values as the reported Ar/N<sub>2</sub> ratio obtained from the continuous observations (about 11 hours of averaged data)."

line 101 “. . .glass flasks.”

line 102 “per meg units as follows.”

line 112 “. . . corresponds to an uncertainty of. . .”

line 116 “. . .to +1,800”

line 122 “. . .but they did not correlate..”

line 124 “. . .must have been superimposed. . .”

line 126 “. . .due to a temperature. . .”

line 138 “. . .by a fundamental sine-cosine. . .”

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

line 146 “Similar increases. . .”

line 150 “. . .found a seasonal. . .”

line 154 “. . .than the  $14 \pm 6$ . . .”

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. . .”

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

line 187 “. . .was estimated to drive. . .”

line 196 “As mention in the Introduction, “

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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.”

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

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

line 269 “ is non-negligible. . .”

line 275 “the derived. . .”

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”.

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