1   RC1: 'Comment on amt-2020-481', Anonymous Referee #1, 21 Mar 2021

2

3   **General comments:**

4   Language: in some places the English writing could be improved, I have listed some

5   suggestions in the detailed comments below.

6

7   Abstract: the abstract would benefit for having a number to represent the differences in

8   precision between the laboratories, next to the offsets. The span offsets given in % are not

9   directly clear, and it would be helpful if they could be expressed differently. It is also not clear

10   from the abstract how this span offsets leads to a value of 0.29 PgC/yr in the carbon budget,

11   and in which direction the shift is. It would be good to elaborate on this. It would be helpful

12   to specify what the term zero offsets represents, so that the abstract is easier to read without

13   reading the manuscript first. It would be good to include a quantification of the comparison

14   of the 2 records at HAT in the final sentence. The "temporal drift" in line 21 could also be

15   further explained, e.g. with details on the time period.

16   Response: The description about the offsets was revised in overall to be able to understand

17   what the value is (p1, L25-L28). We simply described how to estimate a value of 0.29 PgC/yr.

18   But this value was update and revised to 0.30PgC/yr. The words of "zero offsets" was revised

19   to "deviations in the measured δ (O2/N2) values on laboratories' scales" (p1, L28). A result

20   of the comparison of the 2 records at HAT was added in the final sentence(p2, L1-L2). Time

21   period was added to the "temporal drift"(p1, L23).

22

23   Page 3, lines 25-28: it is not fully clear to me how the correction for the expansion rates are

24   applied and how these are measured.

25   Response: We cleared how the correction for the expansion rates are applied and how the

26   rates are measured. (p4, L8-L12)

27

28   Page 4, line 10: it would be good to add information on the choice of the range, it seems quite

29   a large range in comparison to observed values.

30   Response: We selected a large range in comparison to observed values in order to evaluate the

31   difference of the respective span sensitivities accurately. We added the information on the

32   choice of range. (p4, L24-L25)

33

34   Page 5, line 3: how are these values determined? (5000, 1000, 500 times).

35   Response: The values were calculated based on the abundances of $^{17}O^{17}O$ and $^{17}O^{16}O$, $^{18}O^{18}O$

36   and $^{18}O^{16}O$, and $^{15}N^{15}N$ and $^{15}N^{14}N$. We added to the sentence about the calculation. (P5,

L21)

Page 5, line 28: what is sufficiently small?
Response: We revised the sentence to "sufficiently smaller than those of $^{17}O^{16}O$, $^{18}O^{16}O$, and $^{15}N^{15}N$". (P6, L20-21)

Page 6, line 13: why are these values constant?
Response: We added why these values are constant. (P7, L6-L7)

Page 7, line 2: can the authors also provide the long-term stability for the other labs?
Response: Because the long-term stability of NIES and AIST already described, the stability of SIO and TU were added. (TU: P8, L5-L7, SIO: P8, L28-P9,1)

Page 7, line 27: it would be good to explain that the SIO scale is defined to be 0 per meg, because it is used internationally.
Response: We added the sentence of "of which scale is defined as δ (O_2/N_2) = 0". (P8, L26)

Page 9, line 11: (and other places in the text): what does the "expanded uncertainty" represent?
Response: We explained the expanded uncertainty in text (P10, L14-L16).
Expanded uncertainty (U) was represented using standard uncertainty (u) and coverage factor (k)by the following equation,
   U=ku
We used the coverage factor of 2(k=2) which means ≈ a 95% level of confidence.

Page 9, line 14: this seems a large drift in a couple of years' time, so "slightly" might not be the appropriate word here. The explanations for the drift because of the oxidation inside the cylinders seems to be different for each cylinders, are the corrections made for each cylinder separately? Are these the regular cylinders, also used for maintaining the NMJI/AIST scale? Or is this only used for the round-robins?
Response: We removed "slightly" and explained that the corrections for each cylinder were performed separately (P10, L19, L28). We used cylinders having inner wall treated for storing O2 standard gases. NIES and EMRI/AIST also use same type of cylinders.

Page 9, line 32: Where do we see the long-term drift of each laboratory's scale?
Response: The sentence about the long-term stabilities of each laboratory's scale were added

in section 2.3 (EMRI/AIST:p6, L8-L9, NIES:p7,L26-L27, TU: p8,L5-L7, SIO: p8, L28-p9, L1).

Page 10, line 10: what do these percentages represent?

Response: This percentages represent relative deviation from span sensitivity of the NMIJ/AIST scale. We revised the sentence. (P11, L19-20)

Page 10, line 12: could you elaborate on the filling years?

Response: We revised this sentence in overall. (P11, L23-L24)

Page 10, line 16: how are the results consistent with the GOLLUM program? Can this be quantified?

Response: Our results were consistent with those of the GOLLIM program within uncertainty. We add the words of "within their uncertainty". (P11, L28-L29)

Page 10, line 17: could you quantify "slightly bigger"?

Response: This sentence was removed. (P11, L29)

Page 10, line 22: can you quantify how this study shows that the labs can be compared?

Response: We revised "shows" to "aims". (P12, L5)

Page 10, line 25: how was it confirmed that the isotope ratios did not differ significantly?

Response: We add some sentences to get good understanding of this part. (P12, L8-L12)

Page 10, line 29: why not both against the SIO scale?

Response: We understand the SIO scale is internationally used. But purpose of this manuscript is the comparison between individual laboratories' scale values and gravimetric values directly. Therefore, we discussed NIES and EMRI/AIST scales based on NMIJ/AIST scale.

Page 11, line 2: how is the value of -6.6 per meg derived, and should it be compared to the goal of intercompatibility of 5 per meg?

Response: We revised the sentence according your comments. (P12, L22-L25)

Page 11, lines 16-20: the GCB paper has been updated in the meantime twice, and it would

be best to use the numbers from Friedlingstein et al. 2020. Line 18 does not seem to be a full sentence and it is not clear to me what the 0.29 PgC/yr correction is (e.g. from land to ocean, or the other way around?). Table 5: it would be good to add the numbers from Friedlingstein et al. 2020 in the table for reference.

Response: We updated our data using the numbers from Friedlingstein et al. 2020 and the sentence about the 0.29 PgC/yr correction was revised. (P13, L7-L8)

Page 11, line 30: "first time in the world": what is the first time in the world? The GOLLUM program is also an intercomparison program between laboratories.

Response: Because the span sensitivities of the respective laboratories are not compared in the GOLLUM program, quantifying respective span sensitivities is performed the first time in the world in this study.

Page 11, line 31: rewrite the 0.29 PgC/yr, to be more specific what the number means. See comment above.

Response: We revised the sentence in overall. (P13, L24-L25)

Page 12, line 3: what does "other four" mean in comparison to the GOLLUM program?

Response: We used five round-robin cylinders and "other four" mean four cylinders in the five cylinders. (P13, L27-L28)

Page 12, lines 4-6: it would be good to be more specific here on the implications, rather than repeating the causes for the decrease.

Response: We described the implications in the sentence according to your comment. (P13, L29-p14,L1)

Page 12, line 11: can you quantify the bias?

Response: We add the value of the bias. (P14, L6)

Page 12, lines 11-12: how do the results improve the carbon budget and OHC increase?

Response: We added the sentence about improvement of the carbon budget and OHC increase. (P14, L7-L8)

Conclusions: it would be useful to include an outlook. Will this intercomparison continue in the future? Will other laboratories be invited to participate?

Response: We added an outlook in end of the conclusions. (P14, L8-L11)

Table 1: can the authors add more information about the expansion rates? What is meant with the standard uncertainty?

Response: We add more information about the expansion rate. We explained that the standard uncertainty was calculated according to the law of propagation of uncertainties.

Figure 2: what is on the x axis?

Response: We revised the x axis which represent gravimetric values.

Table 3: how is the standard uncertainty determined?

Response: We added how the standard uncertainty is determined.

Figure 3: should the y-axis in panel a read NMJI/AIST instead of grav? Maybe also include the average residuals to compare lab precisions?

Response: This figure represents the relation of the gravimetric values and the measured values. We revised the caption.

Figure 4: maybe add a panel with the differences/bias? Why not on the Scripps scale? It would be good to include in the caption that these are duplicate samples, not measurements of the same flasks.

Response: The difference values were added in figure 4. We described that these are duplicate samples. We explained why not on the Scripps scale in previous part.

Table 5: change to Friedlingstein et al. 2020 (see comment above), and add numbers for comparison. Which numbers are Tohjima et al. 2019? Rewrite "changing rate".

Response: We changed values of fossil fuel and atmospheric CO_2 to Friedlingstein et al. 2020 and revised the footnotes.

Detailed comments:
Page 2
Line 2: "molar fraction" could be changed to "mole fraction", which is more commonly used in the text.

Response: We understand that "mole fraction" is more commonly used in the field. But we should use "molar fraction" because derived quantities should be defined by quantities and not by units (mole is a unit). Angles can be defined as 'length ratios' and not as 'meter ratios'. A mass fraction is not called gram fraction either.

1

2 Line 3: explain "some" stoichiometric ratios.

3 Response: We revised from "some" to "distinct". (P2, L7)

4

5 Lines 2-5: references to earlier studies would be appropriate here.

6 Response: We added references of earlier studies. (P2, L6-L8)

7

8 Line 6: "changes" should be "measurements".

9 Response: "change in" was revised to "measurements of ". (P2, L10)

10

11 Line 11: rewrite "the ocean uptakes"

12 Response: We revised to "the ocean uptakes takes in more than 90% of the Earth's excess

13 energy evaluated based on ocean temperature measurements using Argo floats" (P2, L15-17)

14

15 Line 14: "precision" -> "precise" and rewrite micro-mole-per-mole

16 Response: "precision" -> "precise" (P2, L18)

17

18 Line 27: "per meg" instead of "per meg units"

19 Response: we revised from "per meg" to "per meg units" (P3, L2)

20

21 Page 3

22 Line 6: "remain"?

23 Response: We revised the sentence. (P3, L16)

24

25 Line 22: explain "round-robin"

26 Response: We add the explanation of round-robin in introduction. (P3, L25-26)

27

28 Page 4

29 Line 6: "hereafter the" -> "hereafter called the"

30 Response: we revised from "hereafter the" to "hereafter called the" (P4, L20-L21)

31

32 Line 24: why are the Ar values much more variable compared to tropospheric air?

33 Response: We revised the sentence because the "variable" is wrong. (P5, L10)

34

35

36 Page 7

1   Line 10: what is meant with "unequal to"?

2   **Response:** We revised it to "not equivalent to". (P8, L9)

3

4   Page 8

5   Lines 5-8: the sensitivity factor and interference factors could be further explained to be

6   clearer.

7   **Response:** We explained the sensitivity factor and interference. (P9, L11-12)

8

9   Page 9

10   Line 1: what are "changing rates"?

11   **Response:** The changing rates represent change speed of δ (O$_2$/N$_2$). We revised the sentence

12   a little. (P10, L7-8)

13

14   Lines 29-30: what is meant here by selecting mixtures from the round-robins?

15   **Response:** We removed the sentence. (P11, L5)

16

17   Page 10:

18   Line 5: rewrite "Figure 3a plots"

19   **Response:** We revised to "Figure 3a represents". (P11, L14)

20

21   Page 11

22   Line 10: "corrected" -> "improved"?

23   **Response:** We revised from "corrected" to "improved". (P12, L29)

24

25   Line 13: rewrite "secular changing rate"

26   **Response:** We revised to the average changing rate of atmospheric O$_2$/N$_2$ ratio and CO$_2$ molar

27   fraction reported on the NIES scale. (P13, L3)