Answer to editor's report

Dear Jan Kaiser,

We thank you for the time and effort that was put in editing the manuscript. The comments raised by the reviewers are addressed in separate reports. The corrections that were proposed by you are addressed below, with the answer in *bold and italic*:

1) A lowercase theta is generally preferred over an uppercase one for the triple isotope fractionatio coefficient (Eq. 2).

The lowercase theta is used now throughout the manuscript.

2) You are free to use Δ(17O) symbol without a "prime" since there is no distinction necessary.

Thank you for this suggestion. We now use the symbol without a prime throughout the manuscript.

3) L. 129: Δ should be italics.

The symbol is changed to italics.

4) The amount fraction symbol y should be in italics.

The amount fraction symbol y is now in italics throughout the text.

5) Table 1, heading column 1 should be y(CO2) / (µmol mol–1). Column 2 should be δ(13C, VPDB) / ‰ [or δ_VPB(13C) / ‰, as in Eq. 5]. Similarly, for δ(17O), δ(18Ο).

The column headings are changed as suggested.

6) Εq. 4: The symbols on the left side of the equal sign should be u(δ(13C))/‰. On the right side, you should write Δy(CO2)/(µmol mol–1) and omit the symbol ‰ from the last term, to make the equations dimensionally correct.

We thank the editor for these corrections, which are now in the manuscript.

7) Figure A2 caption: spelling of triangles

The spelling is corrected.

Answer to referee #1:

We thank you very much for the comments and suggestions that improved the manuscript earlier, as well as the positive referee report after these revisions. The last comments of the referee are addresses in this report, with our answers in *bold and italic*.

In the conclusion, the authors state that:

"Our results show that the biosphere is not the dominant process for variations in $\Delta'(170)$. ∆'(17O) of atmospheric CO2 is therefore not suitable as a proxy for quantification for gross primary production at our study locations. The variation in the stratospheric source of high ∆'(17O) is possibly the cause for the high interannual variations we observe in the records." The data and model presented by the authors highlight the significance of the stratospheric signal. However, it's possible that more precise and higher-resolution data could reveal a bisphere signal. Therefore, I suggest that the authors focus on what can be resolved instead of dwelling on what cannot, and remove the sentence from their conclusions regarding the unsuitability of ∆'17O measurements for GPP estimations.

We thank the referee for this suggestion. We deleted the last sentences of our conclusion, that indeed, would fit better in the discussion.

In Line 207, the authors write: "Note that the scale described above for the ∆'(17O) values is indirectly linked to VSMOW, adding uncertainty to the compatibility of other ∆'(17O) scales." No internationally accepted reference material is yet available for ∆'17O in CO2. This means that the CO2 measurements are not directly linked to the VSMOW water reference, and there are many steps involved that can lead to uncertainty. For example, one could anchor their data to measurements of O2 gas from either fluorinating a carbonate reference material or to CO2 equilibrated with VSMOW. It would be useful if the authors could provide more information on the steps taken to link their values to the VSMOW scale, as this would help make the dataset more future-proof.

We realize that the scale that is used for our Δ^{17} O (and δ^{17} O) values is not ideal. *Measurement values from the BGC-IsoLab in Jena were used in combination with measurement values from the IMAU, described in lines 195-208. We chose for this approach as we are dealing with CO2 in air samples, and wish to keep the connection to the JRAS scale, which is realized by the BGC-IsoLab in Jena. The steps we take for connecting our measurements results to the VSMOW scale are, we think, described clearly in this manuscript in lines 192 to 212. The calibration method from the IMAU measurements can be found in Adnew et al. (2019), which is given in the manuscript.*

In the legend of Figure 3, I suggest specifying the location of the "Continuous monitoring".

We specified the location of the continuous monitoring points, and added "Flasks" to all other data categories to clarify the figure.

In Figure 4, there is an inconsistency in the line widths.

The inconsistency is removed in figure 4.

Answer to referee #2:

We want to thank the referee for his/her time and effort he/her put in commenting on the first manuscript, which did help to improve the manuscript, especially in more elaborately discussing the limitations of the dataset, as well as give an outlook for improving the measurements in the future. We are happy with the final report of the referee, and we answer the final comments and suggestions that were given in this report. Our answers are in *bold and italic:*

High D17O variability could be explained by enhanced D17O values from the stratosphere. I agree that due to the precision achieved at the moment, the difference/variation of γ 0.1-0.15 per mil cannot be resolved easily. I also agree that Figure C2 is a better way to show/demonstrate the spectroscopy data can be made useful and show that SICAS D17O pattern is very likely to be real. Please make this clearer, for example, by emphasizing this in the main text (not just in the appendix) and also the caption of Fig C1.

Thank you very much for these suggestions. We agree that figure C2 is convincing in the observed pattern of the SICAS ¹⁷O measurements. In the main text, this is stated in lines 240-242. We added the specific reference to figure C2 in line 241 to make it clearer, as suggested.

Then we state the following in appendix C (lines 511-513): "When considering, however, all datapoints of the Lutjewad flasks from the SICAS and from the IMAU, significant interannual variability is reflected in both datasets. Both the IMAU and the SICAS measurements show lower values in 2020 than in the period before, as observed in figure C2."

We therefore did not add this information in the caption of figure C1, to avoid repetition.

0.08 per mil elevation in the model with dT term included: How much contribution is the newly added term compared to the previous one from N2O? I did a quick estimate from, for example, Koren et al. (2019, JGR) Figure 1, the newly added term is comparable to the existing term, obtained by taking D17O=0.7 per mil from the stratosphere and a factor of ~10 dilution from the land and ocean and the 0.08 per mil increase. That is to say that to explain the data with the newly added term, one needs a lot more CO2 from the ocean (as the site is near the coast and the authors also claimed from absence of correlation between, for example, d13C and conc in CO2; this statement is confusing, see more below), especially to explain the changes from mid-2019 to late 2020, to bring the model values down to match the data. An alternative is to reduce the contribution from the first term, the N2O (eq. 8), which also does not show significant interannual changes. But to do that may change the correlation of CO2 D17O and N2O in the stratosphere. This has to be checked and discussed, at least briefly if not possible to be made thoroughly.

We thank the referee for these comments and suggestions. We agree that there is an offset between the revised model run and the measured values. However, we think the revised model run should be considered as a very first, conceptual model to show that stratospheric input of 17O is more variable than previously described. The mechanism

that is now used in both the original and revised model, is to impose empirical relations for the value of ¹⁷O in the simulated stratosphere following simulated N2O and temperature. In reality, there will be a combination of change of both the \varDelta^{17} O value and the *stratosphere-troposphere transport. We now state this more clearly in the text, line 416: "Note that both the N2O and the temperature anomaly are used as proxy values for the ∆(¹⁷O) in the stratosphere. Thereby, the temperature relation represents both temperature dependence of the actual ∆(¹⁷O) as suggested in Wiegel et al. (2013) and the temperature dependence in stratospheric exchange, which might not be sufficiently represented with only 25 vertical layers in the current model (see e.g. Bândă et al., 2015, for the influence of vertical resolution on stratosphere-troposphere exchange)."*

Biological uptake

Line 327-328: It says "We did not observe any of this, indicating there is no significant biosphere signal in our Lutjewad ∆'(17O) record." This is inconsistent with that described in the paragraph starting at Line 350, that evapotranspiration is taken to explain the reduced D17O during summer time; usually high evapotranspiration is associated with high biological uptake/cycling. How's wind direction during CO2 sampling time? Does it support terrestrial origins of CO2 (which could be affected more easily by evapotranspiration processes)?

Thank you for this comment. We agree that the statement is inconsistent, and we should formulate it differently. It is now formulated as follows:

Line 327: "We did not observe any of this, indicating there is no significant seasonality caused by the biosphere signal in our Lutjewad ∆'(17O) record."

Also super low values less than -0.3 per mil, due to anthropogenic? d13C and [CO2] are better measured. How much reduction is needed in d13C and [CO2], if it's from combustion? Is it supported by CO?

The answer to this question is given below.

For better understanding the CO2 isotope data and variability, please show Keeling analysis results for d13C and [CO2] and briefly discuss the endmember obtained. As claimed in the text, very low values of D17O are likely due to combustion; please highlight the data points having low D17O values.

CO² and d13C values are discussed in the text in relation to the low D17O values, please see line 359: "All points that have lower ∆(17O) than -0.3 ‰, and are sampled during winter/spring, have more depleted δ(13C) values and more enriched CO2 values than would be expected from the seasonal trends. This indicates that local CO2 emission sources are the reason for the more depleted ∆(17O) values in winter. Samples that are very enriched in CO2 amount fractions are not shown here, as these results have very high measurement uncertainties. This could be the reason that a correlation of ∆(17O) and CO2 amount fractions does not appear in figure D1."

We don't want to overanalyse the data, as we know there is a bias towards the high CO2 values. SICAS measurements with high CO2 values (far outside the range that is covered by the calibration cylinders) are not included in the results due to their high uncertainties. We therefore chose to not make any Keeling plots, as our dataset will be a limited representation of the different endmembers.

Line 73: peninsula

Line 73 was corrected as suggested.

Line 322: Suggest to replace Hoag et al. by Koren et al.; Hoag et al. did not study seasonal cycle explicitly, even though it can be inferred.

We changed the reference as suggested.