

## ***Interactive comment on “ $\delta^{13}\text{C}$ values in stalagmites from tropical South America for the last two millennia” by Valdir Felipe Novello et al.***

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Response to reviewer 1. We wish to thank the reviewer for accepting to review our manuscript and provide such a careful and detailed review. There is a clear and genuine interest by the reviewer to help improve our manuscript, for which we are most grateful. We believe that all topics were addressed in the revision and clarified by the comments below.

reviewer comment (RC)

RC: Review of manuscript: ESSD-2020-184 by Novello et al., about a data set publication entitled ‘d13C values in stalagmites from tropical South America for the last two millennia’ The authors present a data set of d13C values for several speleothems from

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South America. Some records in the data set were already earlier published, some are new. But in all cases the according  $d_{18}O$  values were published elsewhere. The time interval of provided data in the new data set are strictly limited to the last 2ka, even when the stalagmites started their growth earlier.

Answer: We did actually provide the entire stalagmite records, including data prior to 2ka. Please see the ages and data of the stalagmites DV2, ANJOS1, TRA7, BTV21a and FN1 that have data older than 2ka.

RC: Furthermore, the authors provide some interpretation on the  $d_{13}C$  data sets, by applying correlation methods (with T and prcp) and principal component analysis. They conclude that the  $d_{13}C$  values in speleothems from South America reflect changes in hydrology, which is closely related to changes in monsoon variability. I like the approach about focusing on stable C isotopes in speleothem, as this is an underrepresented field, as this isotope system is usually much more difficult to interpret. Unfortunately, I think the paper is not ready to be accepted in ESSD. In fact, I suggest to make major edits to the data base and the manuscript text. Please find my suggestions below. I hope they are of some help to improve this interesting work.

Answer: Yes, thank you – the comments were most definitely helpful.

RC: First, I want to evaluate the data set. To my knowledge, this data set indeed seems to include all speleothems, which are published so far from this region and grew during the last 2ka. From this point of view, the data set seems to be complete. I, however, do not understand, why the authors decided to publish only the  $d_{13}C$  values for those parts of the speleothems covering the last 2 ka - even when some speleothems showed some growth in earlier periods. I understand that the interpretation is only focused on the last 2ka, but this shouldn't be a reason to not publish the older portions of the speleothem data (>2ka). In fact, my understanding of this journals scope is its interest in the data sets and their description and not so much on interpretation of data. For me it makes no sense to publish only parts of the available  $d_{13}C$  data for some

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speleothems. I agree, that the data base must not contain data from additional South American speleothems, which grew exclusively earlier than 2 ka BP, but I would expect to find at least the full data sets of those speleothems, which are already included. Another reason, why the d13C values of the older parts of the speleothems should be included, is the usefulness of the data set. I think, you intend to publish this data set with wanting the data to be used. But why should anyone use incomplete data and instead has to search for the full data set elsewhere?

Answer: Please see our answer above related to this issue. We did provide the entire stalagmite records, including data prior to 2ka, when available. However, most of the stalagmites grew within the last 2ka.

RC: In addition, I would also favor, if you could add the according d18O values to this data set. I know that they are already published in individual papers (but this is also true for some of the d13C records) and even in SISAL (see Comas-Bru et al., 2020, ESSD), but it would make things much easier, when trying to use the data, as most users are also interested in d18O. And it would be most inconvenient, to extract the d18O data from SISAL and d13C data from this data set (and maybe even to search for the older parts of the d13C data sets elsewhere) to obtain both time series.

Answer: As the reviewer correctly pointed out, the d18O datasets are already published and available elsewhere with their respective DOI. We prefer to avoid conflicts with previous publications and potential issues with archiving the same data in different formats in multiple repositories. We therefore provide only the unpublished data, in this case the d13C, which is the subject of this paper. However, it may also be worthwhile pointing out, that the first author of this paper (Dr. Novello) serves as the coordinator of SISAL for the South American domain, and in this role, he is committed to providing the d13C data from this paper to the next version of SISAL database. Thereby, the speleothem community will have easy access to both isotopes, as well to all information regarding these stalagmites and caves through the SISAL database.

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RC: Otherwise the data are well stored and easy to access in Pangea. I like this. However, as this journal is more about data sets instead of interpretation, it should at least contain a section in the manuscript, which describes how to access the data (even, when the section will be short, as it is very easy to access them). You might write about the file format, which program is suited to access the data. What other parameters are provided. What are the metadata? How were the age models constructed? (Describe in more detail how you constructed the composites if you decide to include them in your data base (see below).

Answer: As the reviewer stated, the data is stored and easily accessible via Pangaea. The direct link to access the data in Pangaea is provided in the paper. When this manuscript was submitted, the Pangaea webpage initially put restricted access to the data. However, we have contacted the editor of Pangaea and all restrictions to access the data have been removed. With a simple click on the link provided in the paper the reader will be taken to the dataset page. The data are available in the formats text and HTML (standard formats from PANGAEA) and no special software required to read them. The other parameters provided together with the d13C data are “depth” and “age”, which are both intuitive and discussed in the manuscript. We do not supply any kind of metadata and the geochronology provided was taken from the original papers (focused on the d18O), as discussed in the manuscript. When standardizing the records for our statistical analyses, we performed new data interpolations between isotopes and ages, which are described in the section “Methods”. Following the reviewer’s suggestion, we improved this section and the description of our age models.

RC: Furthermore, in the data set itself, you should: \* check if all the d13C values are really reported against the PDB standard as stated in the column header. (I think, they might be measured against the VPDB standard, aren’t they?) Answer: Thank you for pointing this out. In fact, they were measured against VPDB. However, this error was introduced when the editor of Pangaea provided the label of these columns as PDB. We will contact them to change the label back to VPDB.

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RC: \* remove the 'age (ka BP)' column. First this column is redundant and second it is not helpful at all, as you provide only two digits. This is very unfortunate for your subdecadal, high resolution records.

Answer: The editor of Pangaea provided this column. In the original spreadsheet that we sent to the website only the column "age (years AD) was included". It appears, however, as if the "age (BP)" column is a standard from Pangaea.

RC: \* provide more digits for the 'depth' column. Otherwise this column is also only of limited help. Answer: One tenth of a mm is the limit of the precision when sampling stalagmites the conventional way. All stalagmites presented in this study were sampled with a distance between the datapoints  $\geq 0.1$  mm. Thus, more digits in this column have no meaning.

RC: \* add information where to find the according d18O values (if you decide against including them here). \*add information, where to find the original U-Th data sets. (Those are important for researchers wanting to establish alternative age-depth models.)

Answer: This information is included in section 3.1 of our manuscript, where we list the original sources of each record in Table 1.

RC: \* add the stacked records as well, which you constructed here. You are free to follow this advice or not, but to my opinion this would save a lot time for others, which want to have such composite records and would make your data base much more attractive and unique. And you already have the composites at hand. So this shouldn't be much of an effort.

Answer: Thank you for this advice; we will provide these data as well.

RC: Second, I want to focus this review on the interpretation part of the data set. Line 60: Please be more precise and rephrase. Using a sentence like 'The initial source of C for speleothems is the soil CO<sub>2</sub> and tree roots' is very confusing. More correct would be that root respiration produces soil gas CO<sub>2</sub>. But it is also produced by microbial

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decomposition of SOM. So, please rephrase.

Answer: We agree. We have rephrased this sentence to “The initial source of carbon for speleothems is the CO<sub>2</sub> present in the soil, mainly provided by plant roots’ respiration and decomposition of organic matter”.

RC: Line 63: ‘ : : models were proposed to explain the dissolution : : ’ To my opinion, more correct would be ‘ : : models are the extreme cases of the dissolution : : ’, as there are a lot of intermediate conditions, under which carbonate can be dissolved and those are much more common.

Answer: We agree. We rephrased this sentence and now write: “Open and closed system models were proposed to explain two extreme cases of dissolution of calcium carbonate in the percolating solution”.

RC: Line 64: Please replace ‘Initially’ with ‘In an open system’.

Answer: Done.

RC: Line 66: You should finish the sentence by something like: ‘ : : fingerprint from the δ<sup>13</sup>C composition of soil gas CO<sub>2</sub>.’

Answer: Thank you for this suggestion. We rephrased the sentence to “. . .the bicarbonate in solution receives the δ<sup>13</sup>C fingerprint of this reservoir”.

RC: Line 69 to 71: You could be more precise, if you would add something like that the mixing ratio is expected to be approximately 1:1.

Answer: Between lines 69 and 71 we write; “The rock dissolution is limited by the initial amount of CO<sub>2</sub> and, consequently, through this process the δ<sup>13</sup>C from the bedrock influences the isotopic composition of the remaining solution”. We believe that this statement clearly indicates that the bedrock only has an influence on the isotopic composition. It does not suggest that the mixing ratio is 1:1.

RC: Line 72: You might should replace ‘partially open system’ by ‘intermediate disso-

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lution system’.

Answer: We rephased the sentence to: “the interaction between the percolation solution and the host-rock occurs as an intermediate way between open and closed systems”.

RC: Line 74: I would not agree with using the <10% value anymore. In 2001, it was indeed the case that investigated speleothems showed ‘usually’ values like that. But, I wouldn’t use this finding anymore, as since then, there were a lot of additional studies, showing that quite often the host rock contributes a lot more carbon to the speleothem than 10% (e.g. Jackson and McDermott, 2008; Hoffmann et al., 2010; Griffiths et al., 2012; Lechleitner et al., 2016a;2016b; Spötl et al., 2016; Bajo et al., 2017; some examples in Markowska et al., 2019; Therre et al., 2020).

Answer: We agree. Thank you for indicating these references. We removed this last sentence from our manuscript, which in our opinion, does not impact the text.

RC: Line 93 and 94: I have to admit, that I haven’t read the paper by Mickler et al. (2019), but I wonder, if they really say that PCP removes preferentially  $^{12}\text{C}$  from the solution. Usually and according to published fractionation factors for C, during carbonate precipitation  $^{13}\text{C}$  is preferentially removed. Nevertheless, the  $\delta^{13}\text{C}$  values will increase in the solution during PCP. That part of the sentence is correct. But this effect would be due to the simultaneous degassing of  $\text{CO}_2$ . This process is responsible for an increase in  $^{13}\text{C}$  in DIC as the light C isotope is preferentially removed during degassing.

Answer: Maybe this sentence was not clear. Indeed, the PCP does not preferentially remove the  $^{12}\text{C}$  and it was never our intention to imply that. We rephased this sentence to avoid any misunderstanding. Now the sentence reads: “ $\delta^{13}\text{C}$  values from dissolved inorganic carbon (DIC) can undergo fractionation through prior calcite precipitation (PCP), which is forced by  $\text{CO}_2$  degassing that preferentially removes  $^{12}\text{C}$  from the solution to the cave atmosphere (Mickler et al., 2019), depleting  $^{12}\text{C}$  from the

final isotopic product recorded in stalagmites (Baker et al., 1997”).

RC: Line 95-96: ‘: : : exposure of seepage solution to air pockets along the epikarst flow routes : : :’ Maybe that is also a reason, but usually, I thought, it is assumed that PCP is increased, due to larger drip intervals – leading to longer time intervals, where water is in contact with the cave atmosphere leading to more CO<sub>2</sub> degassing and CaCO<sub>3</sub> precipitation before the water reaches the speleothem top.

Answer: Both are the same process. Since the solution is in contact with air, the CO<sub>2</sub> degassing occurs, which increases the PCP. But the reviewer is correct to point out the importance of longer drip intervals. We rephrased the sentence to incorporate this suggestion. Now the sentence reads: “PCP increases during drier periods due to the increased exposure of seepage solution to air. This can occur during the contact of the solution with air pockets along the epikarst flow routes and/or with the increase of dripping interval, where the solution is exposed to the cave air for longer time at the stalactites; both results in CO<sub>2</sub> degassing from the solution, promoting the carbonate precipitation in the epikarst and/or stalactites. . .”

RC: Line 98: Maybe it is more appropriate to talk about ‘variations in PCP are climate related’. To my opinion, the occurrence of PCP itself is independent of climate, but its variations depend very well on climate conditions.

Answer: We agree. We rephrased this sentence to: “the variations in the PCP rate are climate related”.

RC: Line 174-175: I think it is appropriate to add ‘seasonal’ in this sentence as you refer to T-variations ‘throughout the year’ in the lines before.

Answer: Done. Now the sentence reads: “These conditions minimize the seasonal effects of ventilation and temperature, degassing and overall kinetic effects on the isotopic composition of the stalagmites.”

RC: Line 179-181: Here, you are describing how you calculate the average d<sup>13</sup>C data,

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which you use later (Fig. 2) to analyse them with respect to T and precipitation. I think you need to be a bit more precise here. To my understanding you calculate the mean  $\delta^{13}\text{C}$  of the full record (for the last 2ka) and compare this with recent T. I doubt that this should be done. You might want to use the  $\delta^{13}\text{C}$  values of the last 50 or 100 years or something like that. But using the mean  $\delta^{13}\text{C}$  values of the last 2ka, shouldn't be compared with present-day T and precipitation - especially, after you mention a global T increase of 1.44 °C after 1850 CE (see line 145 of your manuscript).

Answer: The reviewer understood correctly. To clarify this issue we improved this sentence that now reads: "For the correlation between the  $\delta^{13}\text{C}$  values and local temperature and precipitation we use a single average  $\delta^{13}\text{C}$  value for each stalagmite calculated over the last two millennia". However, we emphasize in the text that not all stalagmites cover the entire 2ka, and that we use the full time period that is common to most records. We understand that the comparison of the mean  $\delta^{13}\text{C}$  values over the last 2ka with current temperature and precipitation may seem inappropriate. However, only few records have data available for the last 50 years, and some of the others have only few data points during this time interval. If we consider the last 100 years, most of the records will have data only for the period at the beginning of the XX century, which is not a representative time period. Thus, if we use only data from the recent period, we are limiting the use of our dataset and introducing a regional bias by excluding most of the records. The comparison over the full record allow us to use all stalagmites to provide a spatially more complete picture of the records from South America. Furthermore, with this approach, we minimize possible inconsistencies in the data related to geochronology and resolution. In theory, the recent  $\delta^{13}\text{C}$  values should be more closely related to current temperature and precipitation. However, as we show in Figure 2, using the  $\delta^{13}\text{C}$  average of the full record we still obtain a high R-square (0.67 for precipitation and 0.45 for temperature) with high statistical significance ( $p < 0.01$ ), which indicates that average  $\delta^{13}\text{C}$  values over the last 2 ka are indeed representative of the current climate situation.

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RC: Line 190: You describe here, how you constructed composite records. I understand from those lines, that you normalize the data before the stacking procedure starts. But the stacking procedure itself is not described. Please elaborate more on this. Maybe it would also be a good idea to also present the stacked records in your data base (see above). This would be really a step forward.

Answer: This methodology is not new; thus, we had provided the references of Deininger et al. (2017) and Campos et al. (2019) for this data treatment. But following the reviewer's suggestion we have improved our description in the new version of the manuscript. Please see our new description in the Methods section. We will also adopt the recommendation and provide the stacked records. Thank you for this suggestion.

RC: Line 191: What do you mean by 'the inverse operation'?

Answer: As stated above, we have improved the description of our methods. In the new version, this statement is no longer included.

RC: Line 193: 'do not cover the entire period of the last 2000 years'. But you are calculating the PCA only from the period between 650-1950 CE (see line 197)? So, it shouldn't be a problem that those speleothems do not cover the full period of the last 2 ka. Or are they even shorter than this more limited period?

Answer: They are indeed even shorter than this time period. See Table A1. We have rephrased this sentence to make this information clearer. Now the sentence reads: "Four stalagmites presented in Fig. 1 (MV3, FN1, TRA7, BTV21a) were not included in the PCA because these records cover only a limited time period and no other stalagmites from the same karst systems exist that could be merged..."

RC: Line 197 to 199: I am not very familiar with the PCA, but my understanding of a PCA was, that it compares only relative changes in data, not absolute values. Thus, I do not understand your argument to exclude high-altitude sites due to large offsets. In

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addition, I do not understand, why you need a linear regression before you perform a PCA. Please explain.

Answer: The reviewer misunderstood. Nothing that we state in this sentence is related to the PCA. This confusion occurred because this sentence was included in the same paragraph where we are describing the PCA. To clarify this issue, we rephrased this sentence and moved it to a new paragraph in Methods section.

RC: Fig. 2: Indeed that are interesting relationships, however, I still wonder if you compare the mean  $\delta^{13}\text{C}$  values calculated from the speleothems grown over the last 2ka with recent T and precipitation? Please clarify and recalculate the average of  $\delta^{13}\text{C}$  over a more appropriate period (if not already done so).

Answer: See our comment above.

RC: Furthermore, I do not fully understand why there are 9 dots in a) ( $\delta^{13}\text{C}$  vs T) and 11 dots in b) ( $\delta^{13}\text{C}$  vs prcp). Why both plots do not have the same number of data? In addition, according to Tab. 1 there exist data from 18 different caves. Please explain, why there are much less points in Fig. 2.

Answer: These graphs were made using the  $\delta^{13}\text{C}$  values listed in Table A1 e and precipitation and temperature values listed in Table 1. As can be seen in these tables, some of the stalagmites (from the same or different caves) are from regions with the same amount of precipitation and/or temperature, however, they have slightly different isotopic values. When this happens, we calculated the average between all isotopic values that are represented by the same amount of precipitation (or temperature), which resulted in the lower number of the data points. We use this approach to avoid a spatial bias, since some climatic regions have many more stalagmites (and caves) than others. However, we noted that we did not adequately explain this aspect in the text. Now we provide this information in the methods section.

RC: Line 282-284: I am not sure, if it is correct to argue that both regions experienced

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a larger isotopic contribution from host rocks to the  $d_{13}C$  values. I think, there is much more what influences  $d_{13}C$  than the host rock contribution. Such a claim could only be proved with additional  $^{14}C$  measurements. But usually other processes are more important for a change in  $d_{13}C$ , as e.g., soil gas  $CO_2$  production rate, fractionation processes in the cave. Both of those processes lead to elevated  $d_{13}C$  values during drier conditions and should have a higher impact on  $d_{13}C$  as changes in carbonate dissolution processes.

Answer: We agree. We rephrased this sentence to: "... both regions experienced more positive  $\delta_{13}C$  values in their stalagmites during periods of sparse vegetation and thin soil layers above the caves".

RC: Line 310-311: Maybe it is better to be more specific and add 'derived soil gas  $CO_2$ ' after 'soil organic matter', as this is more interesting for speleothem  $d_{13}C$  than the amount of SOM alone.

Answer: Done.

RC: Line 336-337: You argue about T and atmospheric  $CO_2$  to be responsible for the missing coupling between  $d_{13}C$  and  $d_{18}O$  after \_1750 CE. Around 1200 CE, there is a similar decrease in the PC1 of  $d_{18}O$  and  $d_{13}C$ . While  $d_{18}O$  goes back to values similar to before this events,  $d_{13}C$  does not. So there appears to be a similar decoupling of both isotope system as well. However, there is nothing comparable like T or  $CO_2$  increase. Thus, I wonder, how the decoupling at this period can be explained and if T and  $CO_2$  are indeed the most likely explanation for the decoupling after 1750CE. I do not have another explanation myself, but this observation leave me back in some doubt about the T and  $CO_2$  argument. That is really puzzling.

Answer: We do not agree that the decoupling between the PC1s during these two time periods are similar. Around 1200 CE,  $d_{18}O$  goes back to values similar to before and  $d_{13}C$  does not, but both PCAs present the same behavior (although with different range), however, after 1750 the behavior of both PCAs are clear different. The main

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control of the d13C values in the stalagmites is the local hydrology that drives PCP and the CO<sub>2</sub> production in soil, while the main control of the d18O values is the amount effect (an atmospheric process of the convection over the entire continent). We explain this on lines 91-109. Thus, the processes driving d13C and d18O variability are different, although related. Because that, differences between both PC1s are expected. We believe that this was case for the period around 1200.

RC: Line 340-341: You argue, that d13C is influenced by temperature here and refer to Fig. 2. But to my opinion, this plot shows only that the level of d13C values appear to depend on T (but still, you have to answer for which period you have calculated the average d13C values). I think, this is not a proof that the time series for d13C will also react on temperature in the same way. Therefore, please rephrase this sentence or leave it away, as you do not really need this.

Answer: We agree. We have removed this sentence.

RC: Technical corrections: Line 90: Please pay attentions to the spelling of 'Schubert and Jahren'.

Answer: We have corrected this error.

RC: Line 311: Replace 'low' by 'decreasing'

Answer: Done.

RC: Line 330: You are using 'fraction' of explained variance, which calls for numbers between 0 and 1, but in Fig. 4 you provide numbers larger than 1. Please fix this.

Answer: We replaced "fraction" with "percentage".

RC: Line 362: 'much larger'

Answer: We have corrected this error.

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