

RESPONSE TO THE REVIEWER'S COMMENTS:

I would like to thank the authors for taking the time to address the comments and feedback from the reviewers. The manuscript has indeed improved, however, I feel that one fundamental notion related to the C dynamics needs to be considered, as I notice while reading the revised manuscript that the dissolved inorganic carbon (DIC) is likely to be confused with dissolved organic carbon (DOC). This did not occur to me, until I read the last paragraph of the discussion (L390), stating that “the majority of DIC that contributed to the speleothem CaCO₃ had its origin in aged soil OM”. I don't think this is correct, DIC= Dissolved Inorganic Carbon, and inorganic carbon is different from organic carbon (See for example Lechleitner et al., 2019). The distinction between DIC and DOC may be the main source of misconception, and potentially a misleading interpretation of the d¹³C variability. Here, I suggest a re-consideration of this conclusion, and may be a careful revision of the discussion.

We think that the reviewer misunderstood our methodology (LA-AMS): we can only detect the DIC in the stalagmite and not DOC with our method. The DOC is washed down into the karst system where it is decomposed likely by microbial activity, thus, contributing to the DIC in the seepage water. This signal then contributes to the C in the speleothem CaCO₃. This is explained in lines 79 – 81.

While re-reading the revised manuscript, I also feel that the three sections of the discussions are relatively disproportionate, with the middle section being the longest. I understand the reasons for the subdivisions (as it stands), however, the rationale for the division is not fully convincing, time-wise and data-wise (see the following paragraph). I'd suggest having a discussion section that pertains to Fig. 4 (before section 5.1). Any possible explanations of the climatic/environmental changes in either selected interval (i.e., before 8ka, after 3.8 ka, or the interval in between) could be grouped together in this section as a generalization. With all the assumptions and ideas grouped together here, it could be easier to later discuss the driving climatic changes for each selected interval, with more details given to the middle interval.

More about the subdivision of their records in 3 sections, the subtitle for the current sections 5.1, 5.2, and 5.3 is not parallel (hence again my suggestions above), and although the authors state that this subdivision is based on the dcf and d¹³C behavior, the timeframe rationale is not fully convincing. To be clearer with what I meant: the interval before 8ka lasts only ~0.5ka and the interval after 3.8 ka lasts also ~ 1.4ka, however, the mid- interval is quite long (almost 4ka), and there are some intervals (e.g. between 5-6 ka), with relatively stable dcf and d¹³c, but only the jumps in the record are put in highlight. If the interpretation of the pre-8ka (very short time period) is also applied here (i.e., stable), I started to wonder, why the interval between 5 and 6 ka is also not considered stable? In fact, the 5-6ka interval appears to show less variability than the pre-8ka. I think that if the authors re-group all the discussion as I mentioned above, and then reconsider the subdivisions (approx. a millennial subdivision), the data could definitely make a very nice climatic and environmental history about the Spanagel's cave region.

Here, we are answering both of the previous paragraphs combined. We think that a discussion on a millennial scale is not possible based on our data and still think that the three sections we chose are appropriate. We chose the d¹³C signature as the basis for the different sections as it exhibits two relatively stable intervals (i.e., > 8 ka and <3.5 ka). The 14C data assist in the interpretation of the relevant processes in each period. The longest section in the middle (3.5 – 8 ka) shows strong fluctuations in d¹³C.

To make our approach clearer, we added the following sentence to L 205/206: “ [..], i.e., two periods of comparably stable δ¹³C before 3.5 ka BP and after 8 ka BP and the interval in between with large and rapid fluctuations.”

There are some remarks raised by Reviewer #1 for previous manuscript (L 362 and L 395-427), which I would also like the authors to provide more clarification because the argument “gas exchange process” is a bit ambiguous. Clearly at L322 of the revised manuscript (and the corresponding paragraph, two different technical terms are used: 1. Gas exchange process, and 2. C isotope exchange. These are completely different mechanisms, and if I am not mistaken, the most common process during stalagmite formation is degassing (given that the percolating water from the soil and the epikarst above the cave is more saturated in CO₂ than the cave atmosphere). This is because the dripwater pCO₂ is in the process of equilibrating with the cave atmosphere pCO₂. Otherwise, I am also confused with this gas exchange process. My other suggestions would be to discuss about CO₂ (de)hydration-(de)hydroxylation, if that is what the authors really meant by this gas exchange process (e.g., McConnaughey, 1989; Usdowski et al., 1991), but otherwise, I am confused.

We agree with the reviewer and are now more precise about the terminology. Instead of “gas exchange” we will use “C exchange between cave air CO₂ and DIC in drip water” throughout the manuscript.

We do discuss “degassing” as an important process in the manuscript, however, the combination of d13C, d18O and dcf indicates that this process is not enough to explain our observations as we would expect a similar behavior in d18O.

In text-comments

L19: instead of saying very high, just directly say the spatial resolution (in mm or in micrometer). It is better to be accurate

We added: “at spatial resolution down to 100 μm”

L21: please add microdrill for precision here. IRMS does not do the same job as LA-AMS. It needs more tools to extract the sample before it can analyze the samples

We added: “of micromilled samples”

L30: variability in d13C with values ranging from -8 ‰ to +1 ‰ and a generally lower dcf (the underlined texts are suggestion for rephrasing)

Changed accordingly.

L 30-32: something is not correct in this sentence why not only saying degassing (the ‘gas exchange process’ is perplexing and it sounds as if there are more gas in the cave. .but only cave air with CO₂)

As stated above, we changed the expression “gas exchange” to “C exchange between cave air CO₂ and dissolved inorganic carbon in drip water” throughout the manuscript.

L 36: the potential (please remove high, it’s just making the word ‘potential’ weak and redundant)

Done.

L44: may be add Cheng et al., 2013 for more recent techniques?

Done.

L52: add coma before “decaying” and after “2013)”

Done.

L55: the analyses

Done.

L61: add space Fohlmeister

Done.

L67: to be technically correct, you could say “is expected to vary considerably”. There is no expectation of a value to be complex, it is the process leading to the variations that is complex. Value and processes should not be confused.

Changed accordingly.

L75: I think you should also add in this paragraph (i.e., mention quickly) the pyrite oxidation so that readers are aware of the possible cause of acid dissolution of the host carbonate. The detailed explanation about pyrite oxidation can remain where they are below, but it is a good idea to list them all at once in this paragraph.

L81: added “In some karst systems, the oxidation of pyrite has shown to contribute to the acidification of the seepage water and hence to speleothem formation (e.g., Spötl et al., 2016)»

L76: I think this sentence is lacking some info about rainfall (soil carbonic acid does not exist without water from rainfall)

Added "In most karst systems, dissolution of the carbonate host rock is driven by soil-derived carbonic acid forming in meteoric precipitation seeping through the soil."

L78: add the ref. Bergel et al. 2017, Noronha et al., 2015 after "(SOM)", and add relevant reference for the second source, i.e., after "through the soil", and delete the following sentence "Recently, evidence was found for a potential additional C source stemming from CO₂ derived from the oxidation of "old" organic matter (OM) in the deep vadose zone (Bergel et al., 2017; Noronha et al., 2015)". Then, see my comment above (L:75) to be included here.

Reference added for root respiration (Cerling 1984) and (Trumbore 2000) for SOM degradation.

We did not change the structure of this paragraph, because we introduce three different processes that all lead to the contribution of organic C in speleothem carbonate: 1. CO₂ stemming from the degradation of soil organic matter through microbial activity, 2. root respiration and 3. degradation of old organic matter deep down in the karst.

L82: stable and radioactive instead of radiocarbon (otherwise, it is not parallel)

Done.

L83: Figure 1: In this figure, you show soil as a source of OM, do you refer here as the root respiration? If yes, please update the figure to accurately reflect the text, or update the text + figure to fully incorporate all the sources. Some other instances about soil source are also discussed in the remaining parts of the manuscript (e.g. the sentences after L97), and could be helpful if presented here?

Done.

L88: about this 'reservoir effect', is there a difference between the source/factor as you discussed above (OM decay, root respiration, and pyrite oxidation), or does this reservoir effect only apply to OM?

The dcf includes everything that causes a deviation of the 14C content in the stalagmite CaCO₃ relative to the atmosphere at the time of stalagmite formation. This is explained in section 2 of the manuscript.

L102: please add a reference after "50%"

We added «Hendy 1971»

Table 1: some key ref. should be added for Table 1

We added to the caption: "This table is a compilation of data from Fohlmeister 2011b, Spötl et al. 2016, and Therre et al. 2020."

L127: my knowledge about the mineralogy of gneiss is as follow: quartz, Feldspar, hornblende, and biotite. Here the authors do not really specify if this is a pyrite-bearing gneiss or if the pyrite exists as a vein somewhere in the outcrops. Some explanations are needed here. Otherwise, a relevant reference after "gneiss" (e.g., some papers reporting on the geology of the study region)

Pyrite occurs as accessory mineral in this gneiss. We added a reference (Spötl et al., 2004) in L 62.

L133: with an average growth rate of 25µm/a based on 9 U/Th ages

Changed accordingly.

L134: About hiatus: did the authors have any microscopic evidence for this (any figure for reference?) Also coeval existence of other stalagmites from the same cave does not explain the absence of microhiatus in a sample. I suspect that Rev. 1 is correct (if the authors do not have any petrographic evidence to support their claim, I'd suggest responding carefully to Rev. 1 and considering this in the manuscript)

May be the reviewer misunderstood something: The depth-age model shows a gradual age progression (Fig. 3C) and no evidence of growth rate changes. And we added in the previous manuscript version that also the macro- and microscopic inspection of the polished slab shows no petrographic evidence of a hiatus (previous version I 134, now in I 141-145).

L139: may be "interrupted" instead of "affected" is more grammatically correct here

Changed accordingly.

L155: better to indicate the lab where the analyses were performed as in L147

Added: "at the Laboratory of Ion Beam Physics, ETH Zurich, Switzerland,"

L172: indicate the section number (as the subsections in your manuscript are numbered)

Changed accordingly.

Figure 2: is it possible to add in this figure the location of the age trench, please.

Done. Caption added: “Roman numbers and dashed black lines mark the three sections discussed separately.”

The handwriting is a bit confusing, and if looking at Figure 3, the dft goes beyond 1400, where exactly are these trenches located?

Also, it seems that the figure for the bottom pieces have been removed? Should the word “top and bottom piece” be updated in the caption?

“Top and bottom piece” is removed.

L191: Please rephrase as scan ref. T1, T3, and B3 so that readers don’t get confused with two pieces and the scanning method.

Done.

L192: “From the 14C profile” -- I think you mean “Using the dft from the 14C scans”?

Rephrased to: “Using the 14C profile of the speleothem, the StalAge (Scholz and Hoffmann, 2011) age-depth model applied to previously published U-Th data (Fohlmeister et al., 2013) and the known 14C content of the atmosphere during the Holocene, ...”

L194: do you mean 1502 instead of 1402? (see previous comment by Reviewer 1, L243)

No, this is the correct and final number. It was a mistake.

L195: some inconsistent labelling, please correct (there are many instances in the manuscript that the authors need to check and correct. If in the figure they use “a”, they should keep that labelling in the text and not change it to “A”)

Changed accordingly.

L198-203: give the estimated age for each interval

Changed to: “between 30 and 130 mm (ca. 4.0 - 8.1 ka BP). Layers exhibiting a comparably stable $\delta^{13}\text{C}$ occur at the top and bottom of SPA 127, specifically ranging from 4 to 25 mm (ca. 2.6 and 3.7 ka BP) and from 130 to 144 mm (ca. 8.1 – 8.4 ka BP).”

L205-206: This discussion will be divided in three sections, which are based on the speleothems $\delta^{13}\text{C}$ and dcf characteristics (this is a suggestion for re-wording, however, please consider my general comments above)

Done and we also considered the other comments.

L211: I think it is better to provide the carbonate $\delta^{13}\text{C}$ values under C3 vegetation rather than this -25

We rewrote this sentence to make it clearer: “The relatively low $\delta^{13}\text{C}$ value of -5‰ actually contradicts this [..]”

L215: greater instead of larger (dcf values do not represent size, thus, they can’t be large or small)

Done.

L216-217: the presence of two “which” complicates this sentence, please simplify (either by removing the second explanation with “which” or by splitting in two sentences

Rephrased to: Thus, in addition to the atmospheric radiocarbon contribution from living vegetation, an “old” OM source, which respire radiocarbon-depleted CO_2 , is required to explain the depleted $\delta^{13}\text{C}$ values and elevated dcf.

L222: “falls coincides”—please revise

Removed “falls”

L229: Figure 3: Please indicate in this figure the three subdivision you mention in the discussion (may be with a horizontal bar, and annotation I, II, and III)—also, please revise the title of the subsection of your discussion. And again, with the figure labelling and the use of capital or lowercase character (a vs A, b vs B), please be consistent with both the figures and the text.

Done.

L235: of (instead of ofs)

Done.

L236: to be specific, may be better with 3c?

Done.

L241: too wordy, “...processes must be considered”

Done.

Additional general comment about this paragraph: In some part, I may agree with Referee 1 with some of the comments. The high dcf values at 5-6ka (which is closer in amplitude to the interval before 8ka. In fact, the section before 8 and after 3.8 ka are quite short compared to the mid-interval. I think the rationale in the subdivision is not fully convincing. The authors just left the middle interval as is and may seem to overgeneralize it rather than classifying it more appropriately based on the degree of correlation between d13C and 14C.

As stated above, our choice of division is based on the d13C signature. The 14C signal assists in the interpretation of the d13C signal, but a large proportion of the dcf remains elusive. This includes the prominent dcf feature around 5 – 6 ka.

L249: Now, as I re-read the discussion, this section about the 5-6ka should be treated separately, and not shadowed under section II (see again my general comments at the beginning of this review)

We are not shadowing the section between 5 – 6 ka but discuss it within the according subdivision (Section 5.2; L 249 – 254).

L253: Do you mean proposing two hypotheses? (I don't think you are testing a hypothesis here, you're proposing xx and based on your data, you make assumptions on which factors/drivers could best explain your case)

Changed accordingly.

L257: the use of soil-derived CO₂ needs to be specified because it is unclear if this is soil-respired from plant roots or from decaying om in the soil

We added "(from root respiration and microbial decomposition of SOM)"

L267: why DCF in capitals?

Changed accordingly

L272: Considering the potential offset between...

Changed accordingly

L273: use the section number..

Changed accordingly

For the positive correlation, can you please provide the r² and the p-values?

We changed this to «a high degree of similarity», because of the difficulty in aligning the two data sets, the correlation cannot be mathematically calculated, but only qualitatively observed (L273 and 280);

L279-283: please rephrase and write clearer

Rephrased to: "Changes between open and closed carbonate dissolution regimes are expected to result in a correlation between the dcf and d13C, which is observed in SPA 127. However, the magnitude of d13C variations is too large to be explained by a change of dissolution regime even when considering the extreme switch from completely open to completely closed."

L294: Please indicate at the end of the sentence the corresponding growth rate for the older section (a good reminder for the readers)

Done.

L297-300: if the drip water pCO₂ is low, as hypothesized by the authors, the CO₂ gradient between the drip and cave is small, hence, degassing is less, and thus should not lead to a significant increase in d13C (via degassing)

We agree with the reviewer (see Guo 2020 GCA) and removed the term "fractionation" from line 299.

L303: do you mean "isotope fractionation"? or something else?

Yes, changed accordingly.

L318 (about PCP): I think it would be nice if in Figure 3, d13C and d18O are plotted as a time series together (such as the d13C-dcf pair), it is currently difficult to see that in the current figure, and then to assess the PCP

Changed accordingly

L320: "While this would not have an effect on 14C." Why not? Any reference to support this claim?

When measuring 14C through AMS, a fractionation correction is applied conventionally for in the radiocarbon community. Thus, all samples are reported as if they had a d13C value of -25 per mill and all fractionation, which occur to 14C, would have been corrected for. We added a sentence to the materials & methods section (L 175): "and conventionally a fractionation correction to a δ13C of -25‰ was applied (Stuiver and Polach, 1977)."

L322: I think the term “gas exchange” is relatively incorrect, because the drip water is mainly composed of DIC, so it could be more technically correct to say DIC exchange with cave atmosphere. Gas exchange is more likely believed to occur in open system (e.g., soil pore CO₂ exchange with atmospheric CO₂). Please be careful with inaccurate terminology.

Changed throughout the manuscript (see above).

L324: I think this terminology about C-isotope exchange is more accurate

Changed throughout the manuscript (see above).

L325: this term “are long” is vague (and one of the reasons for my comment in the previous version). Please specify the time for the drip intervals (how many drips per second?)

Rewritten as: “meaning that 95% of the CaCO₃ precipitated either via PCP or on the stalagmite (Guo, 2020), “

L332: why two biogenic sources and providing only one value, and what are they?

Because the three biogenic sources have the same d13C value but potentially different 14C values (see Fig. 1 and section 2); see also above.

L341: reference please after 5°C

We added Mook et al. (1974).

L352-355: this does not belong here and does not provide any conclusions about the large changes between 5-6ka. Please revise (following the guidance above)

We agree with the reviewer and removed this section.

L376-...: although this is a great story about the landscape change in the region, the d13C seems to contradict with the statement (look that the d13C values are very high, and these are not typical for trees nor C₃ plants

The d13C is the stalagmite is the result of mixing of different sources and numerous processes. This has been discussed in detail throughout the manuscript (see Table 1, Fig. 1 and section 2).

L393: With the novel LA-AMS...

Done.

L397: This interval before 8ka is only covering a period of <1ka? (which is comparable to the interval of 5–6k where both d13C and d14C also appears stable, but higher). As noted above, I think the choice of time interval subdivision is not fully convincing, leaving the longest part of the record (between 8 and 3.8ka) under debate. Please consider the general comments above.

See above.

L401: I think some of the arguments presented in this paragraph still support PCP. Because of the reduced meteoric precipitation, the water percolating down to the cave is not sufficient to reach the cave itself. Instead, it precipitated CaCO₃ in the epikarst prior to reaching the top of the stalagmite, hence PCP.

We don't understand this comment, because the stalagmite will not grow without water.

L405: this is the reason why pyrite oxidation should be shown in Figure 1

Ok.

L424: please revise (FTIR no longer belongs to the manuscript)

FTIR is still part of the manuscript, even if it is now only in the supplement. Therefore, we prefer to leave this acknowledgement sentence as is.

L425: helped improving this manuscript.

Done

Supplementary:

Figure 5: texts are too small, please revise

We agree that this would be nicer but it is due to logistical reasons not possible.

Figure S10: It would be better if the width of this figure is expanded to fill the entire width of the page and reduce the height by 2/3.

We prefer not to change the Fig.

References: Please update reference style to match Copernicus's

Done.

Additional changes:

Title: we added "in speleothems" to be more precise

L21: removed "(IRMS)

Caption Figure 1: spelt out OC (organic carbon)

Caption Figure 2: removed the explanation in the parenthesis (it is confusing and doesn't add relevant information)

L194: we added what the abbreviation "SG" means (Savitzky-Golay)