

Documentation of changes to our manuscript:

Climatic variations during the Holocene inferred from radiocarbon and stable carbon isotopes in a high-alpine cave

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Our answers are written in green. If not stated differently, we refer to line numbers of the original manuscript.

General changes in accordance with comments made by both referees regarding reorganization of the manuscript:

1. Everything related to the epoxy/¹⁴C anomalies is moved to the SI
2. Line 226 – 237, Fig. 2 and Fig. 3 moved to SI.
3. We created a new section (Section 2) in which we explain the use of ¹⁴C in speleothems, define the term dead carbon fraction and how it is linked to climate.
4. Throughout the manuscript, the radiocarbon concentration is now only expressed as dcf. Therefore, section 1 under results “radiocarbon” is moved to the SI. The new section 2 under Results is renamed to “Radiocarbon (dcf)”.

Referee#1: Comments on tables & figures:

Table 1 – Why is there a “*” after the “expected” in the table. I see no explanation of this. Also, under the “Expected* ¹³C” column of the “Old OM contribution to seepage water acidification” row, does the “shift to more negative values” mean <0 or <10? If it’s closer to <10, I’d label it like that since that’s what you have in the above two rows (for consistency).

Asterisk removed, “< -10%” added instead of “shift towards more negative values”

Figure 1: There is no ruler in this photo. It says the length in the caption, but it would be helpful to have a ruler for reference. This would especially be helpful when looking back at this figure during the discussion when you say the “old section of SPA 127 (>8,ka, >120mm)”, because then we’ll be able to go and see where in the sample this is.

1. ruler is now added and green box is removed (which indicated the region of ¹⁴C anomalies). The original Fig. 1 is now moved to the SI.
2. Caption of Fig. 1 is now shortened: “Picture of SPA 127 (top is left). Top and bottom piece with location of the stable isotope track marked in purple and LA-AMS test track in blue (the tracks corresponding to the data presented in this work were placed next to the test tracks). The total length of the slab is 14.6 cm.”

Figure 2: Are the d¹³C and d¹⁸O from other studies? If so, cite the studies in the figure caption after part (C).

1. The reference for the d¹⁸O measurements is added in the caption of Fig 4 (now Fig 2).

2. Line 120: added “measured in this study”
3. Line 121: added “previously published”

Figure 2: Also, it would be helpful to plot the age in this figure (since age is what you refer to in the discussion, not depth). You could add it by an additional x-axis on the top.

Fig. 2 is now moved to the SI.

Figure 3: You mention that “ $^{12}\text{C}_{\text{HE}}$ ” is the signal intensity in the figure caption, but nowhere in the text do you explain this further. Can you add a section somewhere that says this? It becomes confusing during the discussion section (e.g. lines 270-273), because in these sections you refer to it only as “ ^{12}C ”, and not $^{12}\text{C}_{\text{HE}}$ ”.

Fig. 2 is now moved to the SI and “HE” is removed.

Figure 4: What stands out to me is the jump at 6ka (increase in dcf, decrease in $\delta^{18}\text{O}$, and decrease in $\delta^{13}\text{C}$), but this isn’t mentioned in the text at all? How come?

See further down.

Figure 4, overall comment: “the yellow, white, and orange shaded areas represent phases with distinct stable isotope and dcf characteristics” but what about the two sections (1: $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ decreases; 2: $\delta^{13}\text{C}$ and dcf drops)? These transitions aren’t really talked about in the text, and I’m wondering why you chose not to select these areas as “phases” with distinct characteristics?

Actually, we chose these growth periods because of the stable C isotope characteristics. We removed “dcf” here. As we are not able to explain the dcf change at 6 and 5 ka (see above) we refrain from marking this interval as well.

Figure 4c: How do you know there are not hiatuses in between the U-Th ages? For example, it appears right before an age of 4 (the yellow/white boundary in panels 4a and 4b), there is a sharp decrease in dcf, increase in $\delta^{18}\text{O}$, and increase in $\delta^{13}\text{C}$, yet this is the part of your sample that has the longest gap of age control. How do you know there’s not a micro-hiatus here that’s undetectable? It may be worth mentioning you can’t rule this hypothesis out, just to cover your bases and to let readers know you thought of this (rather than just interpreting this as a purely real signal). Also, the U-Th ages need error plotted.

L136: we added in the text: “There is no macro- and microscopic evidence for the existence of hiatuses in this specimen. Further evidence for the absence of hiatuses is provided by two additional speleothems, SPA 12 and SPA 128, from the same cave are partly coeval with SPA 127. These additional speleothems have a higher dating density in parts, where SPA 127 has only a few radiometric U-Th dating points and also do not show evidence of hiatuses (Fohlmeister et al., 2013). In combination with the well replicated stable O isotope signals we are confident that the growth of SPA 127 was not affected by hiatuses.”

The sign size is larger than the U-Th errors. We added this information in the previous Fig. 4 (now Fig. 2) caption. We added StalAge-derived upper and lower limits of the age-depth model.

Comments by line:

Line 83-85: You mention the conditions in both an “open vs. closed” system can affect the dcf, but you only describe how the dcf typically is in an open system. I suggest adding a sentence describing what dcf would be in a closed system for clarity. Also detailing how there could be a change from an open to closed system may be helpful. I see you outline it in Figure 5, and also in Table 1, so perhaps just simply referencing these two figures/tables will help streamline this discussion.

Line 85: added "In a more closed system, this exchange is inhibited with the extreme case being a completely closed system, where for each mole of carbonic acid one mole of CaCO₃ is dissolved resulting in a dcf of up to 50%."

Line 88-92: "Several studies show more closed-system conditions under higher precipitation regimes" maybe reword this sentence because I'm not sure what exactly you mean by it. What constitutes "more closed-system" conditions? Once again, perhaps by referencing either Table 1 or Figure 5 this would help.

Line 91: added "(see Table 1)» and see point before."

Line 136: How do you know there aren't hiatuses present in your speleothem? Perhaps briefly state how you approached assessing hiatuses in your sample here. In addition, what is the error on each age? I don't see this stated anywhere, and it's not in Figure 4C. The error should be plotted.

See above.

Line 162-163: "offset between stable isotope and radiocarbon data of up to several hundred micrometers", how did you go about accounting for this? Perhaps briefly describe what you did to account for this offset so readers are aware of your methodology. *I see in line 350 that you are unable to apply a correction factor. Perhaps stating this earlier (such as at line 162-163) will help the reader better understand your process of approaching this.

We moved the description from L350 to L162 and rephrased it to: "LA-scans were placed as close as possible to the stable isotope tracks in order to facilitate matching between the two data sets (Fig. 2). However, the LA-AMS setup does not permit to place laser tracks close to the rim of samples causing an offset between the two sampling lanes of approximately 5 mm. Speleothem growth layers are often curved, resulting in a potential offset between stable isotope and radiocarbon data of up to several hundred micrometers, with the outer LA-scan appearing somewhat older than the stable isotope record. Since the curvature of the growth layers is most likely variable, a constant correction factor has not been applied."

L350: rephrased to "Taking a potential offset between the two records into account".

Line 193: It should be stated in the first sentence or two what you used this technique for. Example, directly stating: "FTIR was used for: : :.", because right now it is unclear why you used FTIR (it's not until later in the discussion when you explain identifying the contaminated epoxy area, and I think it'd be better to state up front in this section).

Line 194: we added "Fourier Transform Infrared spectroscopy (FTIR) was used to determine the specific composition of selected areas in our sample to clarify the causes of anomalies." (This part has been moved to SI now).

Line 276: "it's exact composition has been determined using FTIR". It is not until this sentence that I realize what you are using FTIR for. Perhaps add a sentence to the FTIR section (the section starting at line 193) that states, "we use FTIR to determine specific compositions of areas in our sample to clarify the causes of anomalies."

See above.

Line 243: "For the more than 1500 radiocarbon data points", I suggest just inserting the exact number of data analysis points that you have here, instead of saying "more than.."

Now stated the exact number: 1402

Line 259: Please add a reference at the end of this sentence to let readers know where the "previously published 18O values" can be found. Also, did you measure the 13C values in this study? Or did you pull data from another study? This is unclear and should be clarified.

See above

Line 265: I think this paragraph could be reworded so it's clearer. I'm a bit confused about how the different sections of the discussion are broken up the way they are. A suggestion: section "1. LA-AMS anomalies in the old section of:" should be an entirely separate section than the ones below, because it just details how the presence of epoxy caused contamination, and there are no other interpretations of the data associated with it. This entire section be a brief paragraph at the start of the discussion section, and then you could transition into a "part II" of the discussion that's exclusively about the interpretation of the isotope systems across different parts of the sample.

We moved the epoxy discussion to the SI as suggested by Referee #2 and changed the introductory sentences of the Discussion (L265-268) accordingly.

Line 266: A suggestion to clarify this sentence: "...in the bottom part of the sample"

Manuscript changed accordingly.

Line 270: Please add the reference to Figure 3 after this sentence: "The five ^{12}C current peaks correlating with: (Figure 3)" so the readers know this is what you're referencing. Also change "indicating" to "indicate".

This part is now moved to the SI.

Lines 300, 328, 335, 337, 339, 396, 397: Change "C" to "C-isotope". I probably missed additional places you refer to just "C", so please change everywhere this occurs.

Manuscript changed accordingly.

Comment on section "2. Old section of SPA (>8 ka BP) (Line 293-315): The first paragraph of this section (line 294-309) walks readers through interpretations of high δC values and low ^{13}C values from >8ka. A transitional sentence is needed in the beginning part of the second paragraph (lines 310-315), to set up the connection of the two (i.e. the warmer temperatures for the Holocene could have caused the mobilization of the older OM), because right now it feels out of place a bit. Also adding a concluding sentence would be beneficial to wrap up this section of the discussion.

We changed the manuscript accordingly.

Line 319: Perhaps state at what depth/age you are referring to here in this sentence. For example: "As indicated by the reduced growth rate in SPA 127 (Fig. 4, 3.8 ka).."

Manuscript changed accordingly.

Line 320: Misspell of "precipitation" (it says "recipitatin").

Manuscript changed accordingly.

Line 321: Please add this to the sentence for clarity: "The low ^{13}C -values of the first growth period are superseded by rapid and very large variations of ^{13}C ."

Manuscript changed accordingly.

Line 325-328: You state here "the δC between 3.8 and 8 ka is generally lower than the older section: but at 5.8-6ka δC jumps relatively high and stays high until 5.2 ka. I think this should be addressed somewhere in your discussion.

This is now added to the discussion (L249-254 in the revised manuscript).

Line 343, 392: what is (Fig A7?) Do you mean supplemental Figure 7 (Fig S7)?

Manuscript changed accordingly.

Line 330: Hypothesis 1, general comments –

Line 351: “a positive correlation between main features of $\delta^{13}\text{C}$ and dcf are observable for the middle period, especially between 3.8-5 ka and 6-8 ka BP.” I would argue @ 4ka they appear reversed, but 6-8ka I believe I see this correlation. A suggestion: zoom in on these two time slices for what’s plotted in Figure 4B, especially the 6-8 ka one, so the correlation is clearer (the blue and orange lines are kind of on top of each other in the figure now so it’s hard to see). The argument for this could be stronger if you could demonstrate the relationship more clearly.

Enlargements are now provided. We also differentiated between both periods, as the 6-8 ka period shows a better correlation than in the 3.8 – 5 ka period.

Line 354: A bit more explanation for why “an increase of the dcf to 100%” is needed for this mechanism to work would help the flow of this argument better.

Added: “because of the diminishing ^{14}C -rich soil signature”

AND

Line 355: “Generally, the dcf is even smaller than in the youngest and oldest section of the stalagmite.” What about at from 6-5ka? This need addressed.

See above.

Line 354: “..this is expected to be accompanied by an increase of the dcf to 100%.” Why? Some elaboration on this would strengthen your argument.

See above.

Line 362: Hypothesis 2, general comments – Overall the text in the discussion of this hypothesis is clear. But I disagree with your growth rate argument. As stated in previous comments, I’m not sure if a change in the growth rate (19m/11m to 30m) is “significant” – I consider this just a “lower” growth rate. I therefore don’t think you can use this piece of information to suggest it was caused by an overall reduction in precipitation amount. I think you should either try to bring in literature that demonstrate regional drier conditions, or some other support for this argument other than growth rate.

We agree that a reduction in growth rate is not the only potential reason and there are multiple ways to interpret this signal: 1) less precipitation and 2) an exhausted OM reservoir. We discuss both options in the beginning of section 2 in the discussion, because we think, a reduction in precipitation could be very well responsible for the decrease in growth rate. Alternatively, there was reduced contribution of the decreased OM to the acidification of the solution in the karst, which resulted in lower Ca^{2+} concentrations. This has the same effect as a lower growth rate. We will modified this in the current manuscript.

Lines 395-427: I’m a bit confused with this paragraph. I follow your discussion, but are you saying this is your main interpretation for what is happening during this interval and causing all the fluctuations? (i.e. is this what you mean by “in-cave” processes). You state in the conclusion that it’s not bedrock dissolution or fractionation processes, so are you interpreting the strong variability in $\delta^{13}\text{C}$ as a change of gas exchange processes? If so, this needs to be stated clearer, because right now it’s a bit ambiguous whether you mean this or not.

L396-399 We rephrased this and attempt to be clearer: “Another process that is a potential candidate for causing the behavior observed in SPA 127 in this interval is the C isotope exchange between CO_2 of the cave air and C dissolved in the drip water. The gas exchange process may be dominant if the stalagmite growth rate is sufficiently low and when drip intervals are long and/or the differences between the pCO_2 of the water and cave air is small”.

Referee #2

The strength of the paper is the combination of radiocarbon concentration and $\delta^{13}\text{C}$ in stalagmites samples at a continuous and high spatial resolution to understand what happens above the Spannagel cave during some period of the Holocene. However, I found that the manuscript is moderately written (please don't take this personal, and as a reviewer I am not qualified to comment in detail about the writing, but I strongly believe there is a room for improvement and writing is a learning process).

Also, in my opinion, the manuscript requires a lot of reorganization of ideas to make it clearer. For example, the first section of the discussion section, the anomalies in the old section of SPA 127 do not highlight the importance of the proxies being studied. Instead, it discusses the result based on methodological approach. Please note that the journal "Climate of the Past" is not a methodology journal (if this paper was submitted to a method paper, then I would not argue about having this part in the discussion). In fact, I see that this whole section either belongs to the method or some part of it could go as a quick interpretation of the data in the result section, but it should not belong to the discussion section.

We moved this part from the discussion section to the SI and only shortly describe it in the results. Additionally, we moved Figure 2 and 3 in the SI and instead add a new zoom-in Fig. of current Fig.4 as suggested by Referee 1.

Another example, a separate generality section about dcf can be helpful here (see detailed comments) where the authors could explain the difference between radioactive ^{14}C and dead carbon, and what are the potential sources of them. With this said, the sections at L 67-105 could belong to that **independent section** right between the introduction and the Materials & methods. If that general&fundamental notion is separated from the introduction, I am certain that the introduction could become concise and clearer, with a clear statement of the problematic, and a proposition of the new method and its potential relevance in future paleo– reconstruction.

We now only use the dcf and not the initial ^{14}C . This has also been changed in Fig. 6 (including the caption). L 67-105 are now moved to a new section right after the introduction named "radiocarbon and dead carbon fraction" that aims at clarifying the difference between the two and discusses the sources and processes affecting the dcf. Here, we added a new Figure (Figure 1) that shows the typical C signatures in karst environments.

I also feel the title does not fully capture the content of the manuscript. While the authors entitled their manuscript "Climatic variations during the Holocene inferred from radiocarbon and stable carbon isotopes in a high Alpine cave", I found that the manuscript mainly use radiocarbon and $\delta^{13}\text{C}$ as a proxy for local changes and specifically what happens right above the cave in the epikarst, and not directly to climate. In their conclusion, it was made clear that these two are good proxies to understand carbon dynamic. **Hence, I think the authors should emphasize the importance of $\delta^{13}\text{C}$ and $F^{14}\text{C}$ in the use of stalagmites in paleoenvironmental reconstruction and build their discussion based on that, rather than jumping directly to climate, which at this stage seems more speculative.**

- There are several points by the authors in manuscript that support my comments. For example, in the abstract, the authors used the variation in ^{14}C and $\delta^{13}\text{C}$ as an evidence of host bedrock dissolution or organic matter reservoir contribution from the epikarst to the cave. And in fact, this has been one of the focuses of the interpretation/discussion. **The authors should make that clear that from using such inferences**, information from the local place can later be applied to climatic context.

We explain how dcf and $\delta^{13}\text{C}$ can be linked to climate in section 2: Radiocarbon and dead carbon fraction

- In my reading of the manuscript, the bridge 'local response–climate' is quite obscure (possibly by the current way how the manuscript has been organized, or because this aspect is still difficult to fully link with confidence). Realistic suggestion: reorganizing the ideas would significantly improve the manuscript.

Yes, it is still difficult to fully link changes in C-isotopes to climate variations. But in our work we were able to provide evidence for climate related changes in the various proxies of SPA127. We also follow the reviewers ideas for restructuring the ms in order to make the points clearer.

In addition, interpretation of d13C is very complex compared with d18O, although the water-rock interaction may also complicate its interpretation. Among the factors that complicate the interpretation of the C records in speleothems is the-so called PCP (or prior calcite precipitation, or to be general Prior Carbonate Precipitation, to avoid discrimination between the two common CaCO₃ polymorphs, calcite and aragonite). Could this factor influence the proxies being investigated in this study? E.g., for the large range (-8 to +1 per mil)?

We agree that this might be an option for the rapid changes in d13C. Therefore we added a third paragraph under hypothesis 2 and named it PCP and added an explanation, why this cannot be responsible for the observed signal. PCP can have an effect on d13C, even as large as observed for our stalagmite. While this would not have an effect on 14C, we would expect, that d18O should show a similar behavior, which is not the case. Thus, we can safely assume, that PCP is not responsible.

Minor but crucial: There are some confusing technical terms used in the manuscript that need to be specified. For example, the word 'precipitation'. The authors should specify if the precipitation reflects rainfall which is climate or if it represents the carbonate precipitation leading to the formation of speleothems.

We clarified this throughout the manuscript.

To summarize my general comment, I see that the dominant aspects of the hypotheses are focused on the local processes that may affect the carbon stable composition. The paper and the research are interesting, but there is plenty of room for improvement. I hope my general comment and the detailed comments would help improving the paper.

Detailed comments:

L28- Please rephrase first sentence as: "Rapid and continuous analysis of radiocarbon concentration in carbonate samples at high resolution has been possible with the new LAAMS technique."

Done and we added "very"

L30: time

removed

L42-43: I do not fully understand this mechanism, and it needs to be elaborated more

This is now explained in detail in the discussion section (Hypothesis II, part III, line 326 of revised manuscript).

L47: I think there are some missing perspective remarks at the end of the abstract

Added: This study reveals the high potential of combining high-resolution 14C profiles in speleothems with $\delta^{13}\text{C}$ records in order to disentangle climate-related C dynamics in karst systems.

L52: Fairchild et al. 2006: are you sure this is the best representative reference here?

We added other, more appropriate references.

L53: "except" Antarctica

Done

L57: add a relevant literature reference after signal

Done

L59: extra brackets (please remove, and anywhere in the manuscript)

Done

L57-60: There is a recent paper by Fohlmeister et al. 2020, GCA that could be of relevance to this paragraph

Added

L67: Some basics about dcf (as noted in the general comments), that could be an independent general context apart from the introduction

This has its own section now and is described in more detail.

L72: "if a radiocarbon independent..." : why if?

Because otherwise the dcf cannot be calculated. Either because there is no age-depth model at all or if the chronology was established by radiocarbon dates, an assumption for the dcf had to be made to establish an age-depth model.

L77: Values of dcf? (please specify)

done

L78: "commonly vary within a single speleothem with time" : reference please? Earlier you said no data still availabilities statement is therefore not proven nor supported by data?

We added references here.

L85: reference please?

We added a reference.

L88: several studies? May be better to say two studies (as only two references are provided). Otherwise, please be accurate.

There are more studies available than only two, which show such a behavior. We added three more references.

L89: dcf in what?

Added: in the stalagmite

L90-92: I do not fully understand this statement, how can rainfall accelerate SOM decomposition? how does rainfall increase the mean age of soil gas CO₂?

We removed this.

L93: from some of the points in this paragraph, and looking back at the title, wouldn't it be more appropriate to use these two proxies for dissolution rather than climate?

Yes, indeed it is possible to derive dissolution conditions from both proxies. However, dissolution conditions have been shown to depend on climate (e.g., Griffiths et al. 2012; Noronha et al., 2014; Bajo et al., 2017; Therre et al., 2020)

Table 1: The context of these given values are not fully clear. Please explain in the table caption, and indicate under what type of vegetation cover?

We improved the table according to suggestions by Reviewer 1 and added that the δ¹³C values are expected under a C3 vegetation cover.

L106: Please rewrite as "the studied stalagmite grew..." And please add the Lat/Long of the cave

Done in Line 127 (mat & Method section)

L109: add a relevant reference for the geological aspect of the region

Added: Spötl et al. 2004

L134: what does "in situ" imply? , and please add a reference supporting that it was not ice covered during the Holocene (L135)

We removed in-situ and added Fohlmeister et al., 2013 as a reference.

L160-163: aren't there be any markers in the stalagmite layers to match the wiggles?

(please also see more comments further below)

We provided now more details, why it is not possible to correct the measurement depths offsets from the stable isotope and ¹⁴C sampling tracks (Lines 160-165 in the revised manuscript)

L218, 240: is it ^{14}C or was there a typo?

Yes, it the ^{14}C measurements we are referring to in both lines. It is no typo.

L246: are displayed

Done

Figure 4c: would be informative to see the age uncertainty by analysis, and the StalAge model with corresponding confidence interval. Where there is a discontinuity at 7.5ka?

Changed accordingly. No at 7.5ka is not discontinuity. Isotope values are missing there. Therefore, there is a gap. We improved this now by closing the gap in the updated figure.

L259: please add relevant reference as a guide for readers

Done

L265-268: Please remove, if the structure of the manuscript is clear enough, this speechkind of text can be easily removed. It brings no relevant information to understanding the significance of research.

Shortened to: The interpretation of the results on C isotopes in SPA 127 will be divided in three main parts that correspond to three sections identified on the speleothem through distinct $\delta^{13}\text{C}$ characteristics.

L271: please refer to corresponding figures

Moved to SI.

L272: What bulk are you referring to?

Moved to SI.

L270–277: should belong to a short interpretation of the results. It does not give value to the title of the manuscript, which the authors should highlight the importance of F^{14}C in paleoenvironmental reconstruction

General: This whole section 1 of the discussion can either belong to the method or part as a quick interpretation of the data, but it should not belong to the discussion section. The authors should emphasize the importance of $\delta^{13}\text{C}$ and F^{14}C in the use stalagmites in paleoenvironmental reconstruction

We transferred the whole section of the LA-AMS anomalies to the SI.

L310: epikarst (instead of karst), and please add a relevant reference at the end of the sentence at L311

Changed to epikarst. We added “likely” here.

L311-312: There are some contradictions here. More CaCO_3 dissolution does not mean nor imply higher growth rates. More dissolution may suggest more acidic solution, and thus supersaturation in CaCO_3 was not achieved to allow calcite or aragonite to precipitate.

Here, we disagree with the reviewer. Usually carbonate dissolution is a fast process, which happens in the range of hours (Dreybrodt and Scholz, 2011) until saturation is achieved. But even if saturation with respect to Ca^{2+} should not be achieved, a higher soil air pCO_2 leads to a faster dissolution, resulting in a larger degree of CaCO_3 dissolution and in a higher Ca^{2+} concentration. Once such Ca^{2+} - undersaturated solutions enter the cave, they become pretty fast saturated and supersaturated with respect to Ca^{2+} , as CO_2 will degas immediately, as cave air CO_2 values are smaller than that of the soil gas. Then the water, which had the change to dissolve more CaCO_3 , will potentially be able to precipitate more CaCO_3 .

L313: please remove indeed

Done

L314: Mangini et al. 2005: this reference is missing

Done

L320: please correct for the typo. Also, do you mean here rainfall or CaCO_3 precipitation?

Done

L327: precipitation: please specify

Done

L332: few re-writings suggestions: End the first sentence at “period.” Then start with “The first is from soil CO₂..... The second is from sulfuric acid....

Done

L335: is it known how fast changes in soil d13C is transferred to speleothems?

That is a difficult question. We would think that it is not possible to see any change in soil gas d13C earlier than the mean water residence time, which is typically in range of years for most caves. But within a few decades such soil gas d13C change should be visible. But there is no study around, which investigates such a change systematically. Therefore, we do not change the text with respect to this question.

L337-339: any references for this?

We added Bajo et al. (2017) and Spötl et al. (2016)

L340: why modern?

Added “comparably”

L344-345: this could be corrected prior to making comparison (see comment below)

See below.

L350: This is an instructive remark; however, it may be worth comparing (see comment below)

Comment about this paragraph: My problem with this part is that there is no correction done on the dft for d13C and 14C, but then the author commented on the correlation between the two variables. This aspect weakens the paper, because the authors could have done it better. For example, if the scanning speed of the laser is known (plus, they provide the spot size of the laser beam), it is possible to define the dft of the high/low peaks in the LA scans with the dft of the d13C. By selecting about 15–20 key points in the 14C data, I think it is possible to connect the 14C profile with the d13C profile with better accuracy.

We agree with the reviewer. Usually, multi proxy studies with speleothems often suffer from missing alignment between individual measurement tracks. However, often this problem is not mentioned in manuscripts. We appreciate and agree with the suggestion of the reviewer for any improvement. Unfortunately, it is not a problem of not knowing the distance from top of the LA-track. The problem lies in the curvature of individual growth layers, which might be different for different parts of the stalagmite. Thus, even a spatial distance of only 0.5 cm between the stable isotope and LA-tracks can cause some problems in alignment. In order to make this problem more understandable, we moved the description from L350 to L162 and rephrased it to a more detailed description of the problem: “LA-scans were placed as close as possible to the stable isotope tracks in order to facilitate matching between the two data sets (Fig. 1). However, the LA-AMS setup does not permit to place laser tracks close to the rim of samples causing a distance between the two sampling lanes of approximately 5 mm. Speleothem layers are often curved, resulting in a potential offset between stable isotope and radiocarbon data of up to several hundred micrometers, with the outer LA-scan appearing somewhat older than the stable isotope record. Since the curvature of the growth layers is most likely variable, a constant correction factor has not been applied.”

In line 350 we rephrased the statement to “Taking a potential offset between the two records into account”.

L352-353: how about PCP?

Section added.

L365: fast dripping =>fast growth: not necessarily. Fast drip sites in cave may be characterized by undersaturated solution with respect to CaCO₃, and may not lead to CaCO₃ precipitation (but instead, corrosion of the earlier deposited carbonates)

See above. We have no idea about the original drip rate of the speleothem as the speleothem stopped growing about 2ka ago. However as we do not see evidence for corrosion on the

speleothem, we can safely assume that the the drip water always reached the top of the speleothem in a state of supersaturation with respect to the Ca²⁺ concentration. And as long as the drip water arrives at the drip site in a supersaturated condition, drip rate is a major driver for growth rate (see speleothem modelling studies; e.g., Dreybrodt and Scholz, 2011; Scholz et al., 2009)

L370: low drip rate may in some cases increases the CaCO₃ precipitation rates, unless the number of drops/sec is very slow, and hence could lead in hiatus in deposition

We are sorry, but we cannot follow the argument provided here. Following cave modelling studies about growth rate systematics of speleothems it is always the case, that longer drip intervals lead to reduced speleothem growth rates (e.g., Dreybrodt and Scholz, 2011; Scholz et al., 2009) as long as a Ca²⁺ supersaturated solution impinges on the stalagmite top. We are not aware of the contrary from any study.

L378: Fractionation effects: please be precise, this title is quite vague

We are now more precise and termed the title „changes in fractionation effects“

L381: lighter molecules change.... Please rephrase, it reads awkward
changed to: “are preferentially transferred”

L382: Please delete : “this is valid for 13C and 14C isotopes”. The statement does not bring any novelty

Done

L386: any figures for reference here?

No. This statement follows from the conventionally and generally applied concept of fractionation correction of radiocarbon measurements by d13C values (Stuiver and Pollach, 1977).

L388–390: the authors should elaborate on this with one or two sentences

If changes in fractionation occur, then this has some imprint on d13C and d18O. Both proxies should correlate over a short time interval. This was highlighted and explained in the sentence before. In the sentence in line 388-290 we only mention the method, with which we tested for this in a section wise and time resolving approach. We think that is already well explained.

L392: FigA7: this figure does not exist? Or did I miss it?

We are sorry. It should read Fig. S7. We improved this.

L398: long drip interval: I think if drip interval is long, this could enhance degassing/evaporation

We are not sure if we understand the reviewer with respect to the degassing argument. CO₂ degassing is always the first process what happens (see. e.g., Dreybrodt and Scholz, 2011; Guo and Zhou, 2019). The degassing provides the means for the solution becoming supersaturated with respect to Ca²⁺, which finally allows CaCO₃ precipitation. After the initial degassing, further CO₂ degassing is only triggered by CaCO₃ precipitation as for each CaCO₃ molecule one CO₂ molecule will be released.

Evaporation processes should be minimal as the cave has a high relative humidity close to 100%.

L403: which is approximately -8permil (and please add reference)

Added reference.

L408: ref. please?

Added reference.

L428-431: here the authors clearly state that their approach could bring light on understanding processes affecting C isotopes in the subsurface (one of the reasons I made a comment about the mismatch between the title and the content of the manuscript)

See above.

L442: contributed CO₂ to CaCO₃: this is awkward, please rephrase

Rephrased to “We propose that climatic conditions have changed such that this old OM pool in the karst, which is decoupled from the atmosphere, is being increasingly decomposed resulting in a

successively increasing CO₂ concentration in the karst. This CO₂ results in acidification in a comparable way as soil CO₂ enhancing carbonate dissolution and ultimately contributes to stalagmite CaCO₃. The isotopic C imprint, however, is significantly different to soil CO₂ causing the observed increase in dcf.”

L463: why only CO₂ (shouldn't be DIC that is more appropriate?)

Done

Figure 6 is very confusing to me; I am not fully sure what information to take from here?

What is the importance of elevation in F14C?

We improved the figure outline and use dcf now instead of F14C. In the text we describe, that it was very likely, that there was more vegetation than today during the early Holocene, as the timberline was higher, this allowed to produce more SOM, which become uncoupled from the atmosphere later on. As soon as the timberline decreases only a few grasses are expected to survive. Those contribute little CO₂. We expect the majority of soil/karst CO to come from old organic material established during the early Holocene. Under such conditions we would expect to see an ageing component in the F14C or dcf. This observation is indicated by the dashed line.

L477: Re-writing suggestion:

“Results from this study allow to distinguish three intervals with different carbon dynamics.

(1) The interval before 8ka BP....

(2) The interval between 8–3.8ka BP (switch the number to follow the temporal logic)

(3)”

Done

We changed the numbers the older being mentioned first and the younger date second throughout the manuscript.

L479: potentially enhancing bedrock dissolution

Done

Additional changes:

We corrected the use of English language throughout the manuscript and corrected misspelt words.

L30: rephrased to: This novel approach can provide radiocarbon data at a spatial resolution similar to that of stable carbon [...]

L33: comma added

Line 44: “contributed” instead of “is contributing”

Line 45: “was” instead of “is”

Line 47: “hint” instead of “are hinting”

L98: added “(marine-derived)»

L106: rephrased to: “We investigate a stalagmite that grew in the high-alpine Spannagel cave system(47.080278°N, 11.671667°E, Tyrol, Austria; Spötl et al., 2004) by means of C isotope systematics.”

L130: parenthesis removed

L137: “carbon” added

L140: “(compare Fig. 1)” removed

Section Results: “1. Radiocarbon” moved to SI (as suggested by reviewers) and content changed accordingly.

L301: “F14C of 0” replaced with “devoid of 14C”.

L328-329: sentence rephrased by adding “speleothem” and “consequently”.

L406: we added a more accurate value for the d13C of the bedrock: +2.5‰ instead of 0‰

L409-410: we adjusted our calculated values from “-12 and -11‰” to “-11.5 and -9‰”

