

Review of the revised version of CP-2020-110 “Climatic variations during the Holocene inferred from radiocarbon and stable carbon isotopes in a high Alpine cave” by Welte et al.

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 CaCO_3 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 $\delta^{13}\text{C}$ variability. Here, I suggest a re-consideration of this conclusion, and may be a careful revision of the discussion.

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 $\delta^{13}\text{C}$ and $\delta^{14}\text{C}$ 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 $\delta^{13}\text{C}$ and $\delta^{14}\text{C}$, 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 re-consider the subdivisions (approx. a millennial subdivision), the data could definitely make a very nice climatic and environmental history about the Spanagel’s cave region.

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 CO_2 than the cave atmosphere). This is because the dripwater pCO_2 is in the process of equilibrating with the cave atmosphere pCO_2 . Otherwise, I am also confused with this gas exchange process. My other suggestions would be to discuss about CO_2 (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.

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

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

L30: variability in $\delta^{13}C$ with values ranging from -8 t +1 per mil and a generally lower dcf (the underlined texts are suggestion for rephrasing)

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)

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

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

L52: add coma before "decaying" and after "2013"

L55: the analyses

L61: add space Fohlmeister

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.

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.

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

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.

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

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?

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?

L102: please add a reference after "50%"

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

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)

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

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 micro-hiatus 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)

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

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

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

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

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?

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

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

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

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”)

L198-203: give the estimated age for each interval

L205-206: This discussion will be divided in three sections, which are based on the speleothems d13C and dcf characteristics (this is a suggestion for re-wording, however, please consider my general comments above)

L211: I think it is better to provide the carbonate d13C values under C3 vegetation rather than this -25

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

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)

L222: “falls coincides”—please revise

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.

L235: of (instead of ofs)

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

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

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.

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)

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)

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

L267: why DCF in capitals?

L272: Considering the potential offset between...

L273: use the section number..

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

L279-283: please rephrase and write clearer

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

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)

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

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

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

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.

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

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?)

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

L341: reference please after 5°C

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)

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

L393: With the novel LA-AMS...

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 dcf 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.

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.

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

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

L425: helped improving this manuscript.

Supplementary:

Figure 5: texts are too small, please revise

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.

References: Please update reference style to match Copernicus's

References:

- Cheng, H., Edwards, R.L., Shen, C.C., Polyak, V.J., Asmerom, Y., Woodhead, J., Hellstrom, J., Wang, Y., Kong, X., Spötl, C. and Wang, X., 2013. Improvements in ^{230}Th dating, ^{230}Th and ^{234}U half-life values, and U–Th isotopic measurements by multi-collector inductively coupled plasma mass spectrometry. *Earth and Planetary Science Letters*, 371, 82–91.
- Lechleitner, F., Lang, S., Haghipour, N., McIntyre, C., Baldini, J., Pruffer, K., & Eglinton, T. (2019). Towards Organic Carbon Isotope Records from Stalagmites: Coupled $\delta^{13}\text{C}$ and ^{14}C Analysis Using Wet Chemical Oxidation. *Radiocarbon*, 61(3), 749-764. doi:10.1017/RDC.2019.35
- McConnaughey T. (1989) ^{13}C and ^{18}O isotopic disequilibrium in biological carbonates: I. Patterns. *Geochim. Cosmochim. Acta* 53, 151–162.
- Urdowski E., Michaelis J., Botcher M. E. and Hoefs J. (1991) Factors for the oxygen isotope equilibrium fractionation between aqueous and gaseous CO_2 , carbonic-acid, bicarbonate, carbonate, and water (19 °C). *Z. Phys. Chem.* 170, 23 7–249.