Author's response

P. A. Vignoni, F. E. Córdoba, R. Tjallingii, C. Santamans, L. C. Lupo, A. Brauer

We are very grateful to Referee #1, Referee #2, and the Associate Editor Irka Hajdas for carefully reviewing our manuscript and providing constructive suggestions and comments that significantly improved it. Here, we include the reviews received for this manuscript and provide detailed responses indicating the changes made to the text. All comments have been considered in the revised version of this manuscript.

Referee #1:

L16-17: You obviously go on to talk about TEMPORAL variation in reservoir effects too, but I felt that this should also be mentioned right at the start here, along with your noting of "spatial variations".

We have added "and possibly temporal variations" into that sentence. We have also added a new sentence on line 25 of the revised text to stress that temporal changes are probable: "Temporal changes of reservoir effects in sediment records are more difficult to quantify but ^{14}C ages from a short core from Laguna del Peinado may suggest temporal reservoir age variations of a few thousand years".

L24-25: You're absolutely right about the implications of this study affecting both precision and accuracy of 14C-derived chronologies, and they're obviously interwoven, but I wonder if these should be inverted to reflect the greater importance of accuracy over precision? (I.e., accuracy is fundamental – there's no point having inaccurate chronology is there?! – and, after that, increased precision then makes the data increasingly useful, no?)

We have modified the sentence and line 26-27 of the revised manuscript now reads "This study has implications for accurate ^{14}C-based chronologies for paleoclimate studies in the Altiplano-Puna Plateau and similar settings".

L31: You list "endorheic basins that host numerous saline lakes, playa-lakes and salars"; is there scope for these basins to episodically dry out completely, with consequent impacts (hiatuses!) upon age modelling/palaeoenvironmental reconstruction?

We address this point in line 291 of the revised version of the manuscript. We have added: "We do not observe lithological indications in the sediment core neither for a substantial sedimentation rate change nor for a hiatus in the record. However, since detection of a hiatus is not always straightforward, we cannot fully exclude this possibility".

L48-49: "Sometimes, even assumptions on temporal variations of the reservoir effect are included in the construction of age-depth models"; please could you include one or two references to support this statement.

We have added the references in line 54 of the revised manuscript: "(e.g. Grosjean et al., 2001; Moreno et al., 2007)".

L119-124: In order to interpret any radiocarbon data, it is essential to specify what chemical pre-treatment procedures have been applied. (I take on trust that this has been performed robustly, but this needs to be fully clarified, and is probably the most significant of my comments.)

We have now specified the chemical pre-treatment procedures that were applied. The new paragraph on radiocarbon analysis (lines 122-136) in the Materials and Methods section reads as follows:

"Samples preparation, chemical pre-treatments, and accelerator mass spectrometry (AMS) ^{14}C measurements were carried out in the Poznan Radiocarbon Laboratory (Poland). A full description of the procedures can be accessed at: https://radiocarbon.pl/en/sample-preparation/. After mechanical removal of macroscopic contamination under binoculars, the samples underwent a sequential acid-base-acid (ABA) treatment following the protocols established for each material (UW protocol for the wood sample PEI19-P-3 and UV protocol for all other plant samples). Samples were first treated with 1 M HCl at 80°C for 20 min or longer if needed until gas bubbles emanations finished (UV, UW), followed by 0.1M NaOH treatment at room temperature for fragile plant remains (UV) and 80°C for wood (UW); and then 0.25M HCl at 80°C for 1 hr. After each treatment, samples were rinsed with deionised water (Millipore) to pH=7. The NaOH treatment step is repeated a few times until no more colouring of the solution caused by humic acids is observed. For the wood sample PEI19-P-3 (UW), an additional treatment with 5% NaClO2 at room temperature was applied for 30 min. The resulting ^{14}C ages are listed in Table 1 with one standard deviation (σ). Samples with percentage of modern carbon (pMC) and radiocarbon ages were converted to fraction modern ^{14}C ($F^{14}C$) values (Stuiver and Polach, 1977; Reimer et al., 2004; Stenström et al., 2011) using the R package 'rintcal' (Blaauw, 2003). Two post-bomb dates from terrestrial plant samples collected at the shore without contact to lake water were calibrated with CALIBomb (Reimer et al., 2004; Reimer and Reimer, 2023) using the Southern Hemisphere Zone 1-2 calibration data set (Hua et al., 2022)."

L123-124 (and also for Table 1): Why only calibrate the post-bomb 14C measurements, but not the pre-bomb?

We did not calibrate the pre-bomb ages because the reservoir effect of these dates is not known. We now clarify on line 134 that we only calibrate the two post-bomb ages (see answer above). We consider the calibration as not relevant for our main argumentation and, therefore, deleted the column with calibrated ages from Table 1 but kept it in the text as an additional information (lines 154-155, 188-191 of the revised text).

L125-127: Again, what chemical pre-treatment procedures were applied to these samples prior to d13C analysis?

We did not apply chemical pre-treatment prior to analysis. However, we have added further information on the samples and their processing. The new paragraph on carbon isotopes analysis (lines 143-150) in the Materials and Methods section reads as follows:

"Additionally, $\delta^{13}C_{carb}$ was analysed in four samples from the carbonatic matrix sediments at 0-2, 24-26, 46-48, and 71-72 cm depth from the core where the plant macrofossils have been taken, and in one sample from the microbial mats in the southern hot spring. Samples were frozen for 24 to 48 hours, freeze-dried for 72 hours, and ground to powder. Carbon isotopes analysis of carbonate powders ($\delta^{13}C_{carb}$) were carried out on an automated carbonate extraction device (KIEL IV) coupled to a Finnigan MAT 253 IRMS (Thermo Fisher Scientific) at the GFZ Potsdam. In brief, acid digestion of carbonates with phosphoric acid takes place in the KIEL IV to produce CO_2 that is ultimately analysed for $\delta^{13}C_{carb}$ in the coupled MAT 253 IRMS. Results are expressed in the conventional δ-notation in per mille (‰) relative to VPDB (Vienna Pee Dee Belemnite; Table 1). Repeated measurements of the reference material NBS 19 ensured an analytical precision better than ± 0.07 ‰ (σ)."

L129: Surely this is "precision" rather than "accuracy"?

We have replaced "accuracy" with "precision" (now line 150).

L143: Your samples were collected in 2019… and so the latter age (2018-2019 cal CE) makes sense. But how do you explain the former age (1994-1996 cal CE)? A freshwater reservoir effect wouldn't ENHANCE the 14C (112.39 pMC c.f. 101.61 pMC). Precisely what was the material sampled (for both of these samples)? Is the former sample more woody material (with an associated inbuilt "storage age")? Please give more information around these samples, and suggest what has led to this.

We believe the most likely explanation for the 1994-1996 cal CE age obtained for sample PEI19-P-3 is that the plant was not alive at the time of sampling as it had no new sprouts. We now explain it in the text in lines 187-191:

"The only two terrestrial plant samples in our study that yielded recent ages and were free of any reservoir effects were found 15 and 5 m away from the lake shore line (Fig. 1 and 4). PEI19-P-3, a woody plant of the genus *Adesmia,* was most likely dead at the time of sampling as it had no sprouts explaining the 1994-1996 cal CE age (Table 1, Fig. 2a, the front plant was sampled) while PEI19-P-4, a Poaceae possibly *Festuca ortophylla*, dated to 2018-2019 cal CE in agreement with the sampling year (Table 1, Fig. 2b)."

However, as noted in our previous response, the relevant point for this study is that both samples show modern ages and neither is affected by reservoir effects.

L166-167: I would say that this wording is misleading; Yes, terrestrial plants are "expected to provide modern radiocarbon ages without any reservoir effect involved" (generally speaking! Although there could be rare examples where the expectation may differ…) BUT aquatic plants obviously take on their carbon from the water, and so they wouldn't be "expected to" provide modern radiocarbon ages, surely? Isn't that a fundamental premise of the present paper? I just find the wording of this sentence unnecessarily misleading, taken in isolation.

We agree and have modified it to "Present-day terrestrial plants are commonly expected to provide modern radiocarbon ages, while aquatic plants potentially take up old carbon" (now line 186).

L168-169: This is really interesting. I am not a biologist – is the aged C being taken in by the grass from the air (localised atmospheric depletion from C release from the hydrothermal spring), or is the aged C being taken in through the roots (in the water taken up by the plant)?

In order to clarify we have added a sentence and lines 263-268 of the revised manuscript read as follows:

"It has been reported that diffuse emanations of magmatic CO_2 through soils lead to a substantial ^{14}C depletion in terrestrial plants when ^{14}C-free CO_2 is assimilated during photosynthesis (Pasquier-Cardin et al., 1999). This might explain the old age of the terrestrial plant sample since it grew at a distance of only ~15 cm from the local hot spring and was not in direct contact with water. Potential uptake of soil DIC through the roots might additionally contribute but only to a very minor degree since it usually represents less than 1% of the total CO_2 fixed by plants (Loczy et al., 1983; Brix, 1990; Enoch and Olesen, 1993; Ford et al., 2007)."

L169: Clarify again that here you are referring to aquatic species(?).

We have added "aquatic" for clarification (now line 193).

L182-184: Give an approximate representation of the values given for the cited study.

We now give the values for the cited study. Lines 206-209 of the revised text read as follows:

"A similar pattern of spatial variability has been observed in lacustrine systems in the Tibetan Plateau, with high reservoir effect in tributaries and spring waters and lower reservoir effect in the central regions of lakes with differences of up to 19,000 ^{14}C years between different locations within individual systems (Mischke et al., 2013)."

L191-194: "The dissolution of carbonate-rich sediments or rocks in the catchment area is usually considered a main source of 14C-dead carbon influx into a lake (Macdonald et al., 1991; Ascough et al., 2010). However, the dissolution of catchment carbonates can only be a minor source of 14C-dead carbon into Laguna del Peinado because the lithology of the basin is dominated by volcanic rocks". Does this contradict what was written earlier on ("Abundant carbonate precipitation takes place in the El Peinado basin…", L81), or do I misunderstand? (Even if the latter, perhaps clarification is still needed?)

We agree with the reviewer that clarification is needed and therefore we have made some changes to the text. We have modified line 85 (former line 81) and now reads "Carbonate precipitation takes place within both lakes and the hydrothermal springs environments as a result of CO_2 degassing, evaporation, and biological processes…". At the end of line 220 we have also added "…and extreme arid conditions prevail". Finally, we have included a statement in line 237 that reads as follow: "Furthermore, calcium available for carbonate formation in this lacustrine system is interpreted to derive from the alteration of the volcanic bedrock by fluids at high temperatures as described in other lake systems in the Altiplano-Puna Plateau (e.g. Laguna Pastos Grandes; Muller et al., 2020)".

L213 and 216: Can you clarify what you mean by the terms "old" and "ancient" groundwater? (Is it the "100-10,000 years or longer" noted below, L219?)

To avoid confusion, in the revised manuscript we have changed the word "ancient" to "old" in lines 224 and 242. We have also added a statement to clarify and lines 238-240 now read:

"The influence of old groundwater in the lake is consistent with ^{3}H analysis in wetland systems in the Southern Puna Plateau proving that these environments are mainly sustained by old waters that can be centuries to several millenia old, with only minor contribution of modern water (<60 years old) not exceeding 10% (Moran et al., 2019, 2021; Frau et al., 2021)".

L279: Is it possible to measure 14C on (the DIC/dissolved gasses of) the water itself? And would/could this, in combination with other isotope measures (including d13C and d3H, mentioned earlier) help to understand the "dominant process" question?

This is a very interesting question, however, this kind of measurements would not help much in our study area because of the dilution effect from the input of ^{14}C-free volcanic CO_2. We mention an example of ^{14}C measurements on the DIC in both the original and revised text (lines 231 and 258, respectively). We have added in line 244 the following: "Although ^{3}H data of waters is lacking for the El Peinado basin, this system most likely is almost entirely supported by old groundwater as reported also from other sites in the region (e.g. Frau et al., 2021; Moran et al., 2019, 2021; Godfrey et al., 2021)."

L287: I would actually say that "corrections of 14C chronologies based on a single reservoir age for an entire lake…" would result in INACCURATE results, rather than just "large uncertainties" (which, as I noted earlier would be a bigger problem). You would only end up with "large uncertainties" if these uncertainties were ACTUALLY accounted for and, the point that I think you're making (which I totally agree with!) is that often these "large uncertainties" are NOT properly accounted for (…producing small uncertainties, but inaccurate chronologies).

We have changed "large uncertainties" to "inaccurate chronological models" in line 317 of the revised manuscript.

Finally, a more general question relating to your Discussion: If the C assimilated by the species in the hydrothermal pool were solely sourced from magmatic C (rather than "old groundwater"), this would yield "infinitely old" 14C ages… And so, in that scenario, even the older 14C sample would still include some proportion of "modern" C input? (Is that reasonable to assume?) Why not perform a quick endmember "mixing model" to estimate the proportion of C (for each sample) that is from a modern (2019 CE atmospheric) source and what proportion from geologically old (14C dead) C? (N.B. this is a simple "back of an envelope" calculation, rather than requiring "proper" modelling!) I suggest that this will give a "better" impression of the differing contributions (of old vs modern C), which can be skewed by the exponential nature of the 14C decay curve, which can then carry through to all of your samples through the lake. (I.e., for each sample, what proportion of C is sourced from "modern" vs geologically "dead" sources?)

As suggested, we have included the simple "mixing model" in the revised manuscript. This is now included in the methods section in lines 137-142:

"We conducted a simple end-member mixing model to calculate the approximate proportion of dead (^{14}C-free) versus modern (i.e. atmospheric) carbon in each sample following Pasquier-Cardin et al. (1999) as:

$$Dead\ carbon\ (\%) = [1 - (\mathrm{F^{14}C\ in\ sample / F^{14}C\ in\ reference\ plant})] \times 100 \quad (1)$$

We considered sample PEI19-P-4 as the reference plant that best represents local atmospheric $F^{14}C$ (Table 1) at the time of sampling (2019) compared to the average value for Southern Hemisphere Zone 1-2 (1.019; Hua et al., 2022). We assumed that the ^{14}C content in this sample was in equilibrium with the local atmospheric carbon."

The conversion of radiocarbon ages to $F^{14}C$ is detailed in line 132 of the modified paragraph about radiocarbon analysis described in a previous answer.

We have modified Table 1 and included two columns with the "$F^{14}C$" values and the "Proportion of dead carbon". To show these results more graphically, we have also added pie charts in Figure 2 showing the estimated proportion of modern and dead carbon for each sample. For Figure 2 and Table 1 please refer to the revised version of the manuscript.

The results and discussion sections have been also revised accordingly. We have added the following:

Lines 163-165: “A simple mixing model revealed highest proportion of dead carbon in microbial mats from the southern and western hot springs (96.4% and 90.2%, respectively), while values for the lake modern aquatic macrophytes ranged between ~78 and 82% (Table 1, Fig. 2).”

Lines 191-193: “Another Poaceae sample (PEI19-HTS4-T-1) growing in the vicinity of a hydrothermal spring (~15 cm) revealed an age of 1,580 ± 30 BP indicating incorporation of ^{14}C-depleted carbon (~20%; Table 1, Fig. 2d).”

Lines 260-261: “Moreover, the aquatic plant with the oldest ^{14}C age has a proportion of modern carbon (~ 4%; Table 1, Fig. 2d) supporting that the reservoir ages result from dilution with ^{14}C-free volcanic CO_2.”

(Non-comprehensive) typo/wording suggestions:

L14: Insert comma after “This”.

We have inserted the comma after “This” (line 14).

L17: Change “constrain” to “constraint”.

“Constraint” has been corrected (line 17).

L24: Here, do you mean the “centre of the lake” specifically?

We have clarified in the revised text. Lines 23-24 now read: “Altogether, our findings reveal a spatial variability of up to 14,000 ^{14}C years of the modern reservoir effect between the hot springs and the northern part of the Peinado lake basin”.

L114: Missing word: “littoral [zone]”?

We have added the missing word “zone” (line 117 of the revised text).

L115: Spell out “macrofossil”… Perhaps even “plant macrofossil”.

We now refer to “plant macrofossil samples” (line 118).

L127: “Mile” should read “mille”.

This has been changed (line 149).

L143: "cal CE" is a suffix, and so should come after the date (e.g., "1994-1996 cal CE").

We have modified the text accordingly and in the revised version "cal CE" is placed after the date (lines 154, 155, 189, and 190).

L246: Even though I agree that your explanation is the overwhelmingly most likely one, is "proving" still too strong a word to use?

We agree with the reviewer and changed wording to "revealing" (line 275).

L278: I would say that ">26,000 14C years" is more than "up to several thousand years"?!

It is indeed. We have modified line 308 (former line 278) and now reads as follows: "Radiocarbon dating of modern plants revealed large reservoir effects ranging between >12,000 and >26,000 ^{14}C years within the El Peinado basin."

Referee #2

Page 1

Line 22: Please check if it would make more sense to use here the term "younger" instead of "lower".

We have changed "lower" to "younger" (line 22).

Line 28: The introduction is very well written and the problem investigated and the aim of the study are clearly described. However, I think the manuscript might benefit from a few sentences about reservoir effects in general and/or definitions like the terms "C14-free", "C14-depleted",…. . Please consider adding some sentences.

As suggested, we have revised the Introduction. Line 38 of the revised text now reads:

"Our understanding of the regional and temporal hydroclimatic dynamics in the Altiplano-Puna Plateau is hampered by the difficulty in obtaining accurate chronologies from lacustrine sediments due to the scarcity of terrestrial organic matter and the anomalously old apparent ^{14}C age of waters and hence aquatic samples, known as "reservoir effect" (Grosjean et al., 1995, 1997, 2001; Geyh et al., 1998; Valero-Garcés et al., 2000; Yu et al., 2007)."

We have also added a sentence in line 46:

"Reservoir effects depend on different causes including CO_2 exchange rates between the water and the atmosphere, the internal system mixing dynamics, and the input of ^{14}C-free ('dead') or ^{14}C-depleted carbon either derived from dissolved carbonates, volcanic CO_2 or the inflow of old groundwater (Macdonald et al., 1991; Ascough et al., 2010; Keaveney and Reimer, 2012; Jull et al., 2013; Lockot et al., 2015)."

Page 3

Line 80: I am not familiar with the study area, but as it is written "currently" I asked myself if information is available about the frequency of lake level changes and/or the history of earlier connections of both lake systems. In both cases the authors should add information here.

We have added a sentence in line 84 (former line 80): "Both lakes were connected until ca. 2005 according to satellite images (Villafañe et al., 2021)". We have also included a comment on this in line 278 (former line 250): "…probably related to a lake level lowering of at least 0.6 m and the associated disconnection between Laguna del Peinado and Laguna Turquesa (Villafañe et al., 2021)".

Lines 85 – 98: The climate patterns are well described, but to follow this paragraph even better, the manuscript would benefit from an addition of the climate patterns to Fig. 1.

We have revised Figure 1 and it now includes a map with the climatic moisture sources. Please refer to Figure 1 of the revised manuscript.

Page 6:

Line 139: I have three questions/comments to Table 1:

- **I count six questions marks in the table, e.g. "Hot spring 4?". These uncertainties are not mentioned in the text or the Table caption. Question marks should be explained to avoid confusion.**

We have modified Table 1 caption. In the revised manuscript (line 170) it reads as follows:

"Table 1: ^{14}C ages from El Peinado basin. $F^{14}C$ values were calculated with the package 'rintcal' (Blaauw, 2003). The proportion of dead (^{14}C-free) carbon was calculated with reference to sample PEI19-P-4, considered representative the local atmospheric $F^{14}C$. As a reference, for the year 2019 when these samples were collected, the mean value of atmospheric $F^{14}C$ for the Southern Hemisphere Zone 1-2 is 1.019 (January to May; Hua et al., 2022). The $\delta^{13}C$ values in italic correspond to samples at 24 to 26 cm and 46 to 48 cm, and differ at the sampling depths for ^{14}C. Question marks (?) denote samples where water influence, water mixing, and plants genus and/or species could not be determined with certainty."

As clarified in the "Reply to Referee #2" file, some of the question marks are indeed discussed in both the original and revised manuscript (lines 204-206 and 264-268 of the revised text).

- **The first two samples result in two calibrated ages each. It should be explained why this is the case.**
- **Please explain why not all radiocarbon ages have been calibrated.**

In the revised manuscript we have deleted calibration from Table 1 because it is not essential for the discussion and distracts from the main focus of the study (see also comment to Referee #1).

Page 9:

Line 172, 174, 180: The authors refer to Figure 4 only. Its orientation becomes clear only in comparison to Figure 1. However, I wish either an indication of e.g. "western hot spring", a north arrow or maybe a numeration of the hot springs as indicated in Fig. 1 with sample names added to Fig. 4. Otherwise, this paragraph might not be understandable without comparison to Fig. 1. Moreover, Fig. 1 should be referred in addition to Fig. 4.

We have modified Fig. 4 and included the names of the samples as well as an arrow indicating north. We now also refer to Fig. 1 (lines 188, 196, 198, 202, and 206).

Page 12:

Line 257: How do the authors proceed with the sediment core and develop the chronology? I would suggest to implement this information here or somewhere later in the manuscript.

We have included this information in the Conclusions of the revised manuscript in line 318:

"This problem might be solved by either dating truly terrestrial material like pollen or by applying independent dating methods like U/Th. Both, however, have also deficiencies so that constructing chronologies in environments such as that of Laguna del Peinado lake remains a major challenge. Nevertheless, the characterisation of spatial variations in reservoir effects has the potential to better assess the underlying processes influencing radiocarbon ages in a lake even if it does not fully solve the problem of reservoir effect temporal changes."

Line 263: Are there lithological indications that would support the hypothesis of a hiatus in the sediment core?

We have added a comment in line 291 (former line 263) of the revised manuscript: "We do not observe lithological indications in the sediment core neither for a substantial sedimentation rate change nor for a hiatus in the record. However, since detection of a hiatus is not always straightforward, we cannot fully exclude this possibility."

Page 16:

Line 380-381: Please check if the published year should be changed to 2022, as indicated on the journal's homepage

It is now corrected to 2022.

Other changes made by the authors

Line 13: We have replaced "often" with "commonly".

Line 41: We have added the reference "Yu et al., 2007".

Line 91 and 97: We have added "; Fig. 1".

Line 104: We have modified the title of Figure 1 based on the suggested changes. It now reads: "Figure 1: Location and type of samples collected in the El Peinado basin during 2019 (© Google Earth 2020, Maxar Technologies, CNES/Airbus). Sediment core samples are indicated in italics. Left top corner: map of South America with the Altiplano-Puna Plateau highlighted in brown and the climatic moisture sources (SAMS: South American Monsoon System, SHPW: Southern Hemisphere Pacific Westerlies). The red square marks the approximate location of the El Peinado basin in the Puna Plateau of NW Argentina."

Line 111: We added "/modern".

Line 153: We added "away".

Line 176-Figure 2: We have modified the lettering (or numbering) of the images within the figure to separate two samples that were previously indicated together (now 'c' and 'd'). We have also added in the bottom corners of each image a pie chart in reference to the result of the "mixing model" suggested by Referee #1. Therefore, we have made the necessary changes in the text with reference to the new order of the images (lines 156, 158, 160, and 193). The modified figure caption now reads as follows: "Figure 2: Modern samples: (a) and (b) terrestrial, (c) aquatic and (d) terrestrial by the western shore hydrothermal spring, (e) aquatic from the southern shore hydrothermal spring, (f) lake littoral, (g) aquatic from the top of the lake short core. The pie charts in the bottom corners show the estimated proportion of modern and dead carbon for each sample (Table 1)."

Line 179-Figure 3: We no longer include the sedimentation rates.

Line 183: We modified the first sentence and moved it to the Introduction to include there information on the reservoir effect in general as suggested by Referee #2 (see comment above). We also replaced "that lead to the" by "^{14}C-free or ^{14}C-depleted carbon causing", and added an "s" at the end of reservoir effect.

Line 210-Figure 4: We have modified the figure as suggested. The figure caption now reads: "Figure 4: Aerial view of Laguna del Peinado from the northeast and all radiocarbon dates obtained from modern surface samples. For a top view, please refer to Figure 1."

Line 233: We replaced "dead" by "free".

Line 234: We added "Fig. 1".

Line 270: We have added "Fig. 2a, 2b, and 4".

Line 301: We have replaced "age" by "effect".

Lines 333-335: We have added in the Acknowledgements section the following: "We would also like to acknowledge Prof. Dr. Tomasz Goslar, head of the Poznan Radiocarbon Laboratory, for providing us with the necessary information on the samples treatment. We are grateful for the comments and suggestions of the two anonymous reviewers who helped us to improve this manuscript".

Line 342: We have added "and E/1001/-Integrar".

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