

CONSEJO SUPERIOR DE INVESTIGACIONES CIENTIFICAS INSTITUTO PIRENAICO DE ECOLOGÍA

Dr. Ana Moreno Caballud Instituto Pirenaico de Ecología-CSIC Campus de Aula Dei Avda. Montañana, 1005 50059 Zaragoza Spain

Zaragoza, 18th May 2021

Dear Eliza

As the corresponding author, I am submitting the revised version of the manuscript entitled **Spatial variability of northern Iberian rainfall isotope values: investigating atmospheric controls on daily and monthly timescales** written by *Ana Moreno, Miguel Iglesias, César Azorín et al.*, to be considered for publication in *ACP*.

We also include below the notes to the three reviewers, which were previously uploaded to the system. We finally decided, after discussing that issue with you and according to Rev1, to present this work as "Measurement reports" manuscript type.

We look forward to hearing from you.

Sincerely,

LON EUD

Ana Moreno

We have read Rev1's comments about our manuscript and appreciate his/her sincerity. We acknowledge his/her opinion about the novelty and interest of our data. However, we disagree with many of his/her observations as indicated in this response:

1) First, regarding the question of which factor influencing rainfall d180 composition is emphasized in this manuscript, we would like to note that the manuscript refers to a large number of factors and analyze the role they play in the variability of d180rainfall. Our objective is to "assess the principal influencing factors determining rainfall isotopic variability" although we agree we are not always able to **quantify** the effects of every factor since they are frequently playing an overlapping role. We state both in the abstract and in the conclusions the important role played by geographical factors when referring to annual averages and at a spatial approach but temperature and moisture origin and uptake are fundamental factors to explain the seasonality and the differences between "Atlantic" sites and "Mediterranean" ones. Therefore, it is fundamental to characterize the d180rainfall at different sites and at different time scales. For example, in this manuscript we present for the first time in Spain the combination of seven sites to account for the regional spatial variability and the combination of daily and monthly data to account for the temporal scale. This is a huge exercise that for sure will be of interest for this community, even if we don't success on quantifying the effect of every factor separately.

2) Second, Rev1 considers the purpose of this study does not seem to fit the scope of the ACP based on the "local" character of our research. We totally disagree with this remark. In ACP there are many papers focused on a regional approach, not all the studies have general implications for atmospheric science as Rev1 indicates. Here is a short list of recent papers in ACP dealing with rainfall stable isotopes in quite local settings or focused on single events:

- Bonne, J.-L., Masson-Delmotte, V., Cattani, O., Delmotte, M., Risi, C., Sodemann, H. and Steen-Larsen, H. C.: The isotopic composition of water vapour and precipitation in lvittuut, southern Greenland, Atmospheric Chemistry and Physics, 14(9), 4419–4439, doi:https://doi.org/10.5194/acp-14-4419-2014, 2014.
- Bonne, J.-L., Meyer, H., Behrens, M., Boike, J., Kipfstuhl, S., Rabe, B., Schmidt, T., Schönicke, L., Steen-Larsen, H. C. and Werner, M.: Moisture origin as a driver of temporal variabilities of the water vapour isotopic composition in the Lena River Delta, Siberia, Atmospheric Chemistry and Physics, 20(17), 10493–10511, doi:https://doi.org/10.5194/acp-20-10493-2020, 2020.
- Dittmann, A., Schlosser, E., Masson-Delmotte, V., Powers, J. G., Manning, K. W., Werner, M. and Fujita, K.: Precipitation regime and stable isotopes at Dome Fuji, East Antarctica, Atmospheric Chemistry and Physics, 16(11), 6883–6900, doi:https://doi.org/10.5194/acp-16-6883-2016, 2016.
- Jeelani, G., Deshpande, R. D., Galkowski, M. and Rozanski, K.: Isotopic composition of daily precipitation along the southern foothills of the Himalayas: impact of marine and continental sources of atmospheric moisture, Atmospheric Chemistry and Physics, 18(12), 8789–8805, doi:https://doi.org/10.5194/acp-18-8789-2018, 2018.
- Lee, K.-O., Aemisegger, F., Pfahl, S., Flamant, C., Lacour, J.-L. and Chaboureau, J.-P.: Contrasting stable water isotope signals from convective and large-scale precipitation phases of a heavy precipitation event in southern Italy during HyMeX IOP 13: a modelling perspective, Atmospheric Chemistry and Physics, 19(11), 7487–7506, doi:https://doi.org/10.5194/acp-19-7487-2019, 2019.

- Okazaki, A., Satoh, Y., Tremoy, G., Vimeux, F., Scheepmaker, R. and Yoshimura, K.: Interannual variability of isotopic composition in water vapor over western Africa and its relationship to ENSO, Atmospheric Chemistry and Physics, 15(6), 3193–3204, doi:https://doi.org/10.5194/acp-15-3193-2015, 2015.
- Pfahl, S., Wernli, H. and Yoshimura, K.: The isotopic composition of precipitation from a winter storm a case study with the limited-area model COSMOiso, Atmospheric Chemistry and Physics, 12(3), 1629–1648, doi:https://doi.org/10.5194/acp-12-1629-2012, 2012.
- Steen-Larsen, H. C., Sveinbjörnsdottir, A. E., Peters, A. J., Masson-Delmotte, V., Guishard, M. P., Hsiao, G., Jouzel, J., Noone, D., Warren, J. K. and White, J. W. C.: Climatic controls on water vapor deuterium excess in the marine boundary layer of the North Atlantic based on 500 days of in situ, continuous measurements, Atmospheric Chemistry and Physics, 14(15), 7741–7756, doi:https://doi.org/10.5194/acp-14-7741-2014, 2014.

3) Finally, Rev1 propose presenting our manuscript as a Methodological report instead of a Research report. We agree this can be a good change and, after conversation to the Editor, we will move our manuscript to Methodological report format.

We appreciate very much the constructive comments by Rev2 which have certainly helped to improve our manuscript. Here we comment on some issues related to his/her notes which are of particular importance.

1) Shorten and strengthen the structure of the discussion on the climatic controls. We totally agree with Rev2 ideas to shorten and strengthen the discussion and have mentioned the aspect of the difference in ocean surface isotope composition only once and shortly (now it is only included in section 5.3 and removed from the other lines indicated by Rev2). Contrarily, we have dedicated more space to the controls at the moisture source following Rev2 advice, with new analyses of the moisture uptake (see below).

Another idea to strengthen the structure of the discussion was to remove references to temperature control in p.16 and confine the discussion of temperature and precipitation only to Section 5.2. This is a good idea and, in fact, due to this and following Rev2 comment, we have decided to remove the discussion of temperature and amount of precipitation in relation to the type of rainfall. Thus, the last section of the discussion is only dedicated to changes in d180 according to rainfall types. We have modified Fig. 7 in this regard.

2) Temperature and precipitation amount controls on the variability of the isotopic composition of precipitation. We agree with Rev2 about the relative role that temperature may exert on our data and about the difficulty to explain this role without fully understanding the implied processes and all the overlapping interactions. Therefore, we have modified this section to modulate our results (eg. indicating the correlation numbers without implying causality). We are aware that behind the "temperature effect" there are many other processes and mechanisms not easy to describe just by employing correlation analyses. This idea is again employed in Section 5 introductory paragraph, as Rev2 suggested.

3) Moisture source identification discussion. We greatly appreciate Rev2 suggestions on this topic and have included a new analysis to calculate moisture uptake in all events (850hpa trajectories). This idea was also expressed by Rev 3 and it is possibly the most important change we have made in this new version. We use Baldini's method (Baldini et al., 2010) in a more restrictive way (see also Iglesias González, 2019) to identify the locations where moisture uptake processes have been produced during the 48h before the rainfall samples were collected. Taking into account that Iberian Peninsula is surrounded by ocean, together with the fact that most of the rainfall events analyzed in the investigation were produced by frontal systems and convection events (see synoptic analysis), only 850hPa airmass moisture uptake events have been considered as relevant in our new analysis. In addition, while Baldini et al, (2010) considered moisture uptake processes with an increase in 1h of 0.1 gH2Ov/kgair as significant, in our analysis we only took into account events where moisture uptake process where higher than 0.25 gH2Ov/kgair, so if exists any influence in the rainfall isotopical signal, it would be easier to identify than in other previous studies. With this restricted method, and considering all the events analyzed, more than 3000 moisture uptake events have been identified. These events were analyzed considering seasonal variability and the different locations where the rainfall samples were collected. With this new analysis, we are able to identified changes in the moisture uptake location distribution of the airmasses which

produces rainfall events along the Iberian transect. These results are now discussed in detail and represented in a new figure.

4) Seasonality of moisture sources: We mention now in the introduction the role of seasonality of moisture sources. This idea is later revised in the discussion (section 5.3) where it becomes clear after the moisture origin and uptake study.

5) Meteorological context of precipitation events: We appreciate the suggestions made by Rev2 about other studies related to the isotopic composition of different precipitation types. Most of those references are now included in the text. However, Rüdisühli et al. 2020 is included in section 2 when describing which weather system dominates precipitation in which season on the Iberian Peninsula.

Minor comments

We include here our responses to other (minor) comments from Rev2 (those not included here in the letter were minor comments and were just corrected following reviewer's suggestions).

- Rev2 considers the abstract is not well structured and not totally clear in the factors we propose as the most important ones. We agree about the fact that all investigated processes are overlapping and we just show a consistent picture from different angles. We have used this idea in the abstract (also it appears at the beginning of the Discussion section).

- Regarding our text about the NAO, we agree with Rev2 who considered it was out of scope since it was not later used in the analyses performed. Thus, we have removed it from this revised version.

- We agree about the importance of the sampling procedure and have highlighted that only at one site (from the 7 sites) we used an automated system (at El Pindal site). More details on that self-built system are now presented. Unfortunately, at the time of the experiment (2006-2008), we have not compared our system with rainfall collected by other automated or manually procedures.

- According to Rev2 comment, we have moved to the method section lines 392-399 in page 13 and better described our method to calculate and represent back trajectories.

- In line with Rev2 about the reason to use disaggregated precipitation time series, we want to remark we follow the procedure carried out by Millan et al. (2005) to account for the meteorological origin of every rainfall event. We decided to apply this approach to analyze differences between isotopes as a function of three moisture source regions, i.e.: (i) Atlantic frontal systems; (ii) convective-orographic storms; and (iii) easterly advections over the Mediterranean sea (back-door cold fronts). It is well-known in Meteorology that the atmospheric dynamics and evaporation behind precipitation from these three components are different. Thus, this procedure allowed us to discern one of the principal influencing factors (type of precipitation) determining rainfall isotopic composition and variability. For instance, we found higher isotopic measurements associated with convective-orographic storms. This explanation was already included in the text, in Methods, but slightly improved in this new version.

- We have prepared new figures following Rev2 suggestion of splitting Fig 3 into different panels (one per station). We add to every site, the deuterium excess and total precipitation. We include those figures in the supplementary material since we agree they are important but not the focus of this manuscript.

- Rev2 emphasizes the large daily variability of our dataset and this idea is now included in the text. Similarly, we have include a sentence in the short introductory paragraph of the Discussion section making reference to the high complexity of the hydrological cycle with many processes playing a role (or a combination of processes interacting) in the formation of the rainfall isotopic signal.

- In discussion section 5.1, Rev2 points to two sentences or paragraphs that should be moved to section 5.3 and section 5.2. We understand his/her reasons, and have modified these sections to avoid repetitions. Still, the interaction among processes and drivers make inadequate a rigid organization in the discussion and, inevitably, some effects are already introduced in 5.1.

- As suggested by Rev2, we incorporate more often the classification of our studied sites into subregions (Cantabrian coast, Iberian range, Pyrenees and Mediterranean) in the paper (already used to describe the sites, section 2).

- Rev2 indicates that we don't show the important role played by the moisture source in a quantitative and methodologically convincing way. We don't totally agree with this criticism, specially since in Figure 5, the role of moisture source, underlined by the air masses trajectories, is quantified (in percentages of rainfall and in isotopic values). To us, it is evident in Fig.5 the two dominant sources in Borrastre sites, as indicated in the text. In Table 4, the three main synoptic patterns are also quantified for the seven studied sites. In any case, the moisture uptake study carried out in this new version represents a more convincing way of representing the role of the air masses origin.

- We have updated our conclusions following main changes carried out in the new revised version. However, we are not including more discussion on the manuscript about *d*-excess to not complicate it. A previous version of this study incorporated *d*-excess data and we found too complicated for us to integrate it in the discussion. We prefer to make those data available (*d*-excess is represented for every site in the new supplementary figure and data are available at the supplementary tables) giving the opportunity to other experts on that subject to use this large dataset.

Cited references

Baldini, L. M., McDermott, F., Baldini, J. U. L., Fischer, M. J., and Möllhoff, M.: An investigation of the controls on Irish precipitation δ18O values on monthly and event timescales, Clim Dyn, 35, 977–993, https://doi.org/10.1007/s00382-010-0774-6, 2010.

Iglesias González, M. I.: Variabilidad climática del noroeste de la península ibérica durante los últimos 1500 años, descrita por espeleotemas de diversas cuevas del principado de asturias, http://purl.org/dc/dcmitype/Text, Universidad de Oviedo, 2019.

We appreciate very much the constructive comments by Rev3 which have certainly helped to improve our manuscript. The level of detail of his/her comments is extraordinary and very helpful. Here we comment on some issues related to his/her notes which are of particular importance.

General comments

1) Organization of the manuscript and writing. We appreciate the recommendations from Rev3 to improve the structure of the manuscript (eg. merging chapters 4 and 5) and, accordingly, have modified the organization ending with a Results and Discussion section divided in six subsections. We also avoid repetitions, such as the two previous sections in the text where we talk about meteoric water lines. Some of these ideas are in line with those proposed by Rev2 to shorten the manuscript, so we think the final version is certainly improved and have increased its readability.

2) Source regions and backward trajectories. Rev3 considers insufficient our study of back trajectories to discriminate the moisture source at the study transect. We agree with this argument and it probably represents the largest change we have carried out in this version. In fact, it is true, that most trajectories have an origin in the NW, but they later follow a sometimes quite complicated path with different options of moisture uptake. Therefore, we agree that the study of the trajectories alone is not able to represent the processes we want. Therefore we, first, have replaced Fig. 5 by S1 as Rev3 suggested to obtain our results from trajectories extracted the last 1 or 2 days. Second, we have performed a new analysis to calculate moisture uptake in all events (850hpa trajectories). We use Baldini's method (Baldini et al., 2010) in a more restrictive way (see also Iglesias González, 2019) to identify the locations where moisture uptake processes have been produced during the 48h before the rainfall samples were collected. Taking into account that Iberian Peninsula is surrounded by ocean, together with the fact that most of the rainfall events analyzed in the investigation were produced by frontal systems and convection events (see synoptic analysis), only 850hPa airmass moisture uptake events have been considered as relevant in our new analysis. In addition, while Baldini et al, (2010) considered moisture uptake processes with an increase in 1h of 0.1 gH2Ov/kgair as significant, in our analysis we only took into account events where moisture uptake process where higher than 0.25 gH2Ov/kgair, so if exists any influence in the rainfall isotopical signal, it would be easier to identify than in other previous studies. With this restricted method, and considering all the events analyzed, more than 3000 moisture uptake events have been identified. These events were analyzed considering seasonal variability and the different locations where the rainfall samples were collected. With this new analysis, we are able to identified changes in the moisture uptake location distribution of the airmasses which produces rainfall events along the Iberian transect. These results are now discussed in detail and represented in a new figure.

Specific comments (in red, the Rev3 comments)

Line 2. The title refers to "climate controls" on the variability of isotopic composition in rainfall. The study itself though seems more to be focused on meteorological processes such as moisture pathways and weather regimes/precipitation types of rain days. Perhaps, the authors may consider to use or add another term such as "weather", "meteorological", or "atmospheric"?

Good suggestion! We use atmospheric.

Line 39-40. Perhaps, besides referring to the dataset, this concluding sentence may also refer to the analysis that helps to understand rainfall isotope variability in relation to meteorological / atmospheric processes and geographic influences?

Good suggestion! Change "dataset" by "analyses".

Lines 73 and 74. The term "trajectories" is perhaps quite technical for the introduction. Instead, a term that refers to actual physical processes, such as "air mass origins" or "air mass transport" may be more appropriate.

Done

Lines 80-85. This is a crucial paragraph as it outlines what the intention of the study is, and what it adds to previous studies as outlined in the text above. The thought behind the first sentence "In this paper ..." is not clear to me. Is the approach, based on multiple stations new and is that the main selling point of the paper? Or is this study presenting a comprehensive analysis based on multiple stations across the Atlantic-Mediterranean transect? In the first case, the authors may write "we introduce a new approach...", and in the latter case, "we present a comprehensive / multiple perspective analysis on daily and monthly ...". Also, is it really new that a study considers multiple stations across a region? If other studies followed such an approach, perhaps for other regions, this may deserve attention in the introduction to provide context for this study, for example by adding a new paragraph. In addition, this paragraph may explicitly refer to the atmospheric processes and geographic factors that influence the isotopic rainfall variability that are addressed in this study to guide the reader's expectations.

We are presenting a comprehensive analyses based on multiple stations, not certainly "a new approach" since there are many studies using multiple stations. Some of those previous references are included now in the introduction. We also added some information about the processes and factors we are going to address in the manuscript.

Line 87. This section addresses besides the site description and climate also the different weather regimes that bring precipitation over the northern Iberian Peninsula. This may be reflected in the title of the section.

Done.

Lines 103-104. The phrase "also easterly advections over the Mediterrean Sea" sounds somewhat vague. Please, rewrite, perhaps in the direction of "fronts that approach the Iberian Peninsula from the east (backdoor cold fronts)"..

Done

Lines 119-122. While reading this paragraph I somehow lost the storyline. The first sentence refers to the dominant source regions and seems to follow as a conclusion from the text above, while the next sentence introduces the four different climate zones. The authors may consider to add the first sentence to the paragraph above (or elsewhere), and to start a new paragraph with the second sentence.

Yes, we agree and have removed the first sentence.

Then, the introduction of the four climate zone regions is hard to follow; It may help rephrase this sentence as, for example, "Below, the seven stations are grouped into four regions and described in terms of their climatology". Also, it feels somewhat chaotic to refer at this stage

multiple times to Figure 4 while Figures 2 and 3 have not yet been discussed. Is it necessary to include the line "Regional meteorological data are provided in Figure 4A."?

Done. We have removed references to Fig. 4 that were unnecessary.

Lines 123-127. Can this paragraph be shortened by saying "The sites of El Pindal and Oviedo..." and removing the sentence on lines 126-127 "Additionally, ... in this study."?

Done

Line 197. To what "Meteorological data" is referred? If this is the air temperature and precipitation, please, remove the brackets, and rephrase the sentence to place more emphasis on these meteorological variables, for example, as "Air temperature and precipitation are obtained from the closest meteorological stations over the sampling periods, as indicated in Table 1, to investigate "

Done

Line 292. Usually, when referring to the ERA-Interim analysis Dee et al. (2011) is cited.

Done.

Lines 211-238. In this paragraph I feel quite overwhelmed by the many references to Tables and Figures for which here only the applied methodology is described (e.g., Tables 3, 4, and 5 and Figure 5). I would recommend to only refer explicitly to the Tables and Figures when discussing the scientific results, not when describing the used (statistical) methods.

I partially agree about this... but citing tables here is quite necessary to refer to the place where the reader can find the data associated to that analysis. We have kept the references to Tables and removed those to Figures.

Lines 223-224. Which reanalysis data are the HYSPLIT simulations using? This should briefly be mentioned, including the resolution of the underlying reanalysis.

Done. We have included this brief sentence: "GDAS (Global Data Asimilation System) have been used in Hysplit simulations with 0.5^ox0.5^o spatial resolution".

Line 226. One should be cautious with referring to the origin of the rainfall using an analysis that is solely based on air parcel trajectories without taking into account the uptake of moisture along its pathways. The part of the sentence may be rephrased in the direction of "to generate a vector representing the mean trajectory of the air mass transport associated with the precipitation".

Yes, we agree. In this new version, a procedure to consider moisture uptake is included (see general comments above)

The titles of sections 4.1 and 4.2 may be rephrased as "Daily rainfall isotopic variability" and "Monthly rainfall isotopic variability".

Done

Line 245. It may be helpful to refer to a study that presented the Global Meteoric Water Line. More importantly, a reader may expect after these two lines (244-247) an interpretation and discussion of the local meteoric water lines. What do we learn from the analysis? How do these local meteoric water lines compare to other regions? Later on, I realized that lines 281-

285 further discuss this subject. The manuscript could benefit to describe this aspect at one place only (see also general comment 1).

We have better organized this section and merged the two places where the meteoric water lines were described. Comparison with other sites in southern France is now included.

Line 253. This synchronicity is quite remarkable as, according to this study, precipitation across the northern Iberian Peninsula is controlled by different weather regimes. May this suggest, along with later findings that show similar isotopic rainfall along the western and eastern coasts, that the elevation and temperature effects dominate the isotopic signatures in precipitation?

We think that this case is quite singular since it represents the influence of an Atlantic front passing over a large region of the IP and affecting our sites in a similar way (high precipitation amount, very negative isotopes). It may be difficult to extrapolate this quite exceptional situation to the whole record and extract general conclusions.

Lines 261-268. Here I miss again a discussion and interpretation of the results. Simply phrasing the main findings without interpretation leaves the reader guessing what to take away from the text. Later on, I realized that the text from line 286 onwards seems to continue with this analysis. Please, discuss one subjects at one place in the manuscript.

This text was just a presentation of the data since this section was in Result chapter in the previous version of the manuscript. Now we have included the discussion of the data, adding information previously on line 286 onwards.

Line 315. In fact, when considering the above and following analysis, I get the impression that the elevation and/or temperature effect has the strongest influence on the rainfall isotopic variability (in the order of 2 permil) as compared to all other factors. Or is this too simplistic?

It is true that elevation and temperature are important to explain averaged values (eg. annual means) but not enough to explain daily variability. For that scale, we need to account for the air mass history (moisture origin and moisture uptake, type of rainfall, etc).

Lines 454-456. Another study that found similar differences in the isotopic signature in precipitation from convective versus stratiform precipitation in the Mediterranean is Lee et al. (2019). Citing this study may strengthen the text here.

Yes, this paragraph is enriched with new references provided by both Rev2 and Rev3.

Lines 460-462. The sentence "Backdoor cold fronts heavy precipitation and flooding (Llasat et al., 2007)" already appeared in section 2 (lines 107-109) and is thus repetitive. Please, remove the sentence at one of the two locations.

Done. We remove it from the discussion (section 4.6).

Lines 493-495. I cannot follow the sentence. Please, clarify and correct if necessary. In addition, how are outliers defined in Figure 7?

Following recommendation by Rev2, this last paragraph associating rainfall types with precipitation amount or temperature has been removed to be speculative. We have also simplified Fig. 7 to show only the variation of d18O associated to the three rainfall types

Tables. Overall, I find the information in the Tables quite overwhelming, and I wonder if the information can be reduced without losing relevant information. For example, the multiple use

of "n=" in the cells of Table 2 could be avoid by choosing another notation, perhaps providing the number of samples between brackets after the d18Op values, or simply by removing "n=" in all cells and providing adequate description on top of the columns or in the Table title/caption.

Done. Table 2 is simplified according to these suggestions.

Lines 223-238. One of the main methodologies of the study is defining the different weather regimes that are linked to the rain events and d18Op values. Upon first reading I missed how these different weather regimes are defined, and realized that lines 231-236 address this method. I would recommend to make this methodology more visible by renaming the title of section 3.4. In addition, more information should be provided on how these different synoptic situations are defined, allowing for potential reproduction of the results. Is this analysis subjective or based on an automated detection algorithm?

We have changed the title in the method section to make this methodology more visible and provide more information about where to find the criteria (subjective) to define the three different synoptic situations.

We have also changed all the typos and other errors indicated by Rev3 in "Technical comments" thus improving this new version of the manuscript.

Cited references

Baldini, L. M., McDermott, F., Baldini, J. U. L., Fischer, M. J., and Möllhoff, M.: An investigation of the controls on Irish precipitation δ 180 values on monthly and event timescales, Clim Dyn, 35, 977–993, https://doi.org/10.1007/s00382-010-0774-6, 2010.

Iglesias González, M. I.: Variabilidad climática del noroeste de la península ibérica durante los últimos 1500 años, descrita por espeleotemas de diversas cuevas del principado de asturias, http://purl.org/dc/dcmitype/Text, Universidad de Oviedo, 2019.