REVIEWER 1

General comments

This study presents a new dataset of sedimentary pigments from Lake Borreguil (Sierra Nevada, Spain), in the context of climate change, nutrient sources, and their impact on aquatic productivity in high-mountain lakes. The paper constitutes a valuable contribution to the limited literature on sedimentary pigments as biomarkers of primary producer communities. This paper compares community changes with existing (downscaled) climate data to constrain the environmental factors responsible for the development of the lake ecosystem in the last 440 years.

Overall, these data deserve to be published; however, a clear storyline is difficult to deduce from the large amount of information provided. As we deem this data valuable, we suggest some major changes to enhance the clarity, comparability, attractiveness, and reproducibility of the manuscript.

The paper can benefit from streamlining of the main message e.g., shortening, removal of repetitions and redundant information, restructuring to remove repetitive sections. All of these aspects disturb the flow of the read. Similarly, the number of plots can be reduced; however, there are also plots missing that could aid in understanding the discussion topics and highlight the evidence presented to support the arguments.

Authors often refer to other literature without clearly specifying what they wish to refer to or where the argument is leading. We encourage the authors to pay extra attention to this in the line-by-line comments. Repeating in detail which parts of the literature were relevant would help the reader understand the line of argument, without the need to read all the referenced literature.

ANSWER: Thank you for your thorough revision. We have carefully considered your comments and made the necessary changes. We believe these revisions have significantly strengthened the manuscript. The primary changes are as follows:

- Introduction: Revised and updated in accordance with the suggestions of Reviewer 1 and 2. Objectives have been reduced.
- Materials and Methods: Expanded the "Study Site" and "Methodology" sections with new data. We have also included a new table in the Supplementary Materials.
- Results: Entirely restructured and rewritten following Reviewer 1 and 2 recommendations to describe findings zone by zone. We have added a new table and a new figure to the Supplementary Materials, as requested by the reviewer 1.
- Discussion: Streamlined and more focused. The section has been reduced from eight pages to less than five.

- Figures: The total number of figures in the main manuscript has been reduced from ten to six to improve conciseness. Figures have been amended according to the reviewer’s suggestion.
- Data in repositories- Raw data and statistical data have been placed in open- source platforms Github.

Individual scientific questions/issues ("specific comments")

The title of this contribution sounds as if it were AI-generated and is overselling. We suggest using an alternative title without AI superficial words, such as “striking”, as well as the typical AI-generated format “,:” followed by a question. Here we attach an example of a shorter yet still well-descriptive title suggestion: “Tracking algal changes during the last ~400 years using subfossil pigments in a high- mountain lake, Sierra Nevada, Spain”. If authors used LLM/generative AI model(s) extensively, we encourage them to add this in the form of a disclaimer at the end of the manuscript.

ANSWER: The manuscript title was not created by the IA but it was the suggestion of the first author. Anyway, we have changed it to the proposal title.

Lines 11-14: Ensure that the affiliations are consistent, information such as address, ZIP code, city, state, and country, need to be listed for every institution.
ANSWER: Thank you. We have updated the affiliations of two authors.
1. Carsten Meyer-Jacob. Université du Québec en Abitibi-Témiscamingue Institut de Recherche sur les Forêts (IRF) 445 Boulevard de l’Université, Rouyn-Noranda, QC, J9X 5E4, Canada
2. Teresa Buchaca.Integrative Freshwater Ecology (CEAB-CSIC), C/ acceso a la Cala St. Francesc, 14, 17300, Blanes, Girona (Spain)

Abstract

Please consider shortening the abstract while maintaining focus on the main message within the broader context. Some minor rephrasing can resolve this.

- Line 17 – Suggestion: “have been shown to be” -> “are affected”

ANSWER: Change has been made. See line 19 of the revised manuscript.

- Line 18 – Suggestion: “there is an increasing body of evidence from paleolimnology that indicates”

-> “Paleolimnological studies increasingly indicate”

ANSWER: The sentence has been eliminated following your suggestion.

- Line 19 – Suggestion: “paucity” -> “lack”

ANSWER: Thank you. Paucity has been substituted by lack (Line 20 of the revised manuscript).

- Line 19 – This is very specific for it being the second sentence of an abstract to use diatoms. Is there any less specific data in the publication, such as an algal community, or so? It would also better link to the next sentence, addressing the “overall algal community”

ANSWER: The sentence has been removed in order to shorten and focus the abstract.

- Line 20-22 – Suggestion: “This study investigates the shifts in algal biomass and communities using subfossil sedimentary pigments from the last ~430 years. We use well-dated sediment cores from...”

ANSWER: Thank you. The changes have been made. Line 22.

- Line 21 – Using high-resolution is problematic, as different methodologies assign different meanings to the term 'high-resolution’. We suggest stating the exact temporal or spatial resolution for clarity.

ANSWER: We have included the interval resolution in the sentence: 'high-resolution (2.5 mm intervals). Line 22.

- Line 23-24 Suggestion: “We noted a significant algal biomass increase in the 1970s, which appears to be linked to increasing temperatures and aerosol concentrations.” Thus, the sentence at line 24 becomes repetitive and can be removed.

ANSWER: Thank you for your suggestion. However, note that we refer to changes in algal biomass but also to changes in algal composition and both should be referred. We have changed the sentence to shorten it attending to your suggestion (see lines 23-25): “We noted a significant change in both algal biomass and community composition throughout the core, with a notable intensification since the ca. 1970s, which appears to be linked to increasing temperatures and aerosol concentrations. ~~These changes appear to be linked to climate and atmospheric aerosols”.~~

- Lines 25-26 are somehow repetitive. We suggest merging these sentences into one to avoid redundancy.

ANSWER: It is important to note that we analyzed algal biomass and also algal composition. Following your suggestion, we have joined the two sentences together (lines 25-27):" Algal biomass and composition exhibited two significant shifts approximately between 1740–1840 and from 1970 to the present, with the latter period reaching unprecedented algal biomass concentrations".

- Lines 29-31: As this was already partly mentioned in the sentence at line 24, we recommend adding the enhanced Saharan dust to line 24 and removing this part.

ANSWER: We have done it the other way around. We removed the sentence from line 24 and maintained it in line 29-31. So that the current lines 23-25 would look like this: "We noted a significant change in both algal biomass and community composition throughout the core, with notable intensification since the ca. 1970s, which appears to be linked to increasing temperatures and aerosol concentrations. ~~These changes appear to be linked to climate and atmospheric aerosols"~~.

- Line 33 – Did the "N availability" change over time, or is it rather linked to the N/P ratio? If the latter, a clarification is needed here, e.g., "compared to P, which is brought disproportionally to this lake by Saharan dust."

ANSWER: We have changed the sentence following your suggestion. Please, see lines 32-33: "This is probably due to reduced nitrogen availability compared to phosphorus, which is brought disproportionally to this lake by Saharan dust".

- Line 34-36: We suggest rephrasing to increase the text clarity and remove redundancies, e.g., "The observed changes in the algal community in Borreguil Lake were temporally related to algal community shifts in other Mediterranean sites, suggesting that rising global temperatures, aerosol deposition and enhanced Saharan dust deposition will likely continue to affect the ecological condition of these ecosystems"

ANSWER: Thank you. The sentence has been rephrased (lines 35-36). "The observed changes in the algal community in Borreguil Lake are unprecedented in the last ~400 years in the Sierra Nevada lakes and were temporally related to algal community shifts in other Mediterranean sites suggesting that rising global temperatures, aerosol deposition and enhanced Saharan dust deposition will likely continue to affect the ecological condition of these ecosystems.

- <u>Lines 35-36:</u> It is great to conclude the abstract with the predictions, and we suggest adding the implications or consequences linked to the predictions for the broader context and the importance of studies like this one.

ANSWER: The last sentence has been changes to include your suggestion and reviewer 2 suggestion (Lines 37-38): “Projected increases in global temperatures and Saharan dust deposition will likely lead to an increase in the lake trophic status and significant changes in algal composition in Borreguil Lake”.

Introduction

Overall, the paragraph structure, as well as the structure of the entire section, can benefit from improvements in information distribution.

The novelty of this specific research needs to be more clearly defined, especially in comparison to Jiménez et al. (2018, 2019) and Pérez-Martínez et al. (2020a). The knowledge gap for this study needs to be better stated by the end of the second paragraph, not the end of the introduction. The studies mentioned above have already answered the first RQ and partially answered the second RQ.

We recommend focusing on the novel aspects of the manuscript and decreasing the number of RQ/hypotheses, which include:

1. the in-depth analysis of community shifts, and
2. The relatively longer timescale perspective.

As is noticeable in the discussion section, the current RQs were also merged and discussed in the two main overall discussion sections. This is a clear sign that there is no need for more than the overall 2 RQs.

We recommend outlining the problem and stating what remains unclear after considering all the other studies addressing aquatic ecosystems under stress in the first and second paragraphs. This text is now placed in paragraphs 5 and 6 instead. Parts of the 3rd and 4th paragraphs can be blended into the “study site” section (2.1).

ANSWER: The novelty of this study has been stated at the third paragraph (please, see lines 58-64): “This study has been conducted in Sierra Nevada range (Southern Spain) where, despite the well-documented impact of climate change on their lakes, the long-term response of algal community composition remains unknown. In this study, we analyse changes in algal biomass and community composition over the last 430 years in Borreguil Lake by examining sedimentary pigment concentrations. We relate these changes to environmental variables such as climate and Saharan deposition. Whereas many other studies have addressed changes in a particular group of organisms (e.g. diatoms or cladocerans) during the Industrial era, we focus on an in-depth analysis of shifts in the algal community and adopt a relatively longer timescale, expanding our scope until the Little Ice Age (LIA).”

While other articles from our group have partially addressed the first two research questions presented in the original manuscript, these articles only cover the last 180 years. We are now studying the last 430 years since the Little Ice Age (LIA). This enables us to understand the changes that have occurred since the 1950s and 1960s within a broader timeframe. We believe this to be an important addition to our research compared to previous studies. Moreover, in this work, we analyse changes in algal community composition, providing additional information to that obtained from the study of specific groups of algae (e.g. diatoms) or algal biomass indicators (e.g. chlorophyll-a). The last RQ (changes in algal community) has not been addressed totally or partly in previous research.

We have reduced the research questions from four to two RQ. Please, see lines 105-108: “(1) A previous study of the last 180 years shows a significant increase in algal biomass over the last 50 years. Is this increase also significant when viewed over the extended period of 430 years? (2) Has the composition of the algal community shifted over this interval?”.

Consequently, these two objectives were covered in respective sections of the discussion: 4.1. Changes in algal biomass, 4.2 Changes in algal composition

A big part of the discussion has been relocated in Study site following your suggestion, specifically the present composition of algal communities (Lines 145-154).

Part of the Introduction has been also relocated in Study site (Lines 115-127).

- Lines 46-47: It is somehow unclear what the stress actually is; a high-altitude location and the position in the Mediterranean zone are not themselves a stress. Could you please clarify what stress you mean already in this sentence?

The sentence has been changed to include your suggestion (lines 49-51): “Of particular concern is the case of high mountain lakes in the Mediterranean region, as they face two distinct forms of stress: intensification of climate warming because their high-altitude location and intensification of summer droughts because of their position within the Mediterranean zone (Nogués-Bravo et al., 2008)”.

- Line 51-52: The last sentence of the paragraph brings only information, but no context.

Please, see the changes in lines 55-57: “Additionally, significant amounts of Saharan dust are transported to the Mediterranean (Salvador et al., 2022) and fertilize with elements as Calcium, Phosphorus and Iron the mediterranean ecosystems”.

- Lines 53-67 belong to the study site and can be removed here, then blended with the study site section.

We agree, most of the information about Sierra Nevada lakes has been removed and blended with the study site section, lines 115-127.

- Line 53: A similar phenomenon as in the previous paragraph: the first introductory sentence carries way too little context. Consider adding the crucial argumentation in short already in the introductory sentence, such as here: what makes the lake setting unique? It comes in the paragraph, but it would be great to have it already mentioned in the first sentence.

ANSWER: We already have removed this sentence from the introduction following your suggestion.

Line 58: Is the temperature information from the reference cited in the next sentence? Or where does this come from?

ANSWER: The temperature information comes from the reference cited in the next sentence. We have include the reference in the sentence (lines 116-117):" Since 1864 CE, the Sierra Nevada region has warmed by 1.56 °C, mainly due to increased spring and summer temperatures (Sigro et al., 2024)".

Anyway, this sentence has been moved to the Study site.

- Line 59: Please provide an estimate of the decline in precipitation, particularly from 1975 to 2020 CE. From what number to what number (in mm) or a percentage.

ANSWER: We have included the percentage to the sentence (Lines 117-118): "Summer precipitation has declined, particularly from 1975 to 2020 CE (-13.4% per decade^{-1}) (Sigro et al., 2024)".

This sentence has been moved to the Study site

- Line 64: Where does this information come from? Low-alkalinity – maybe worth mentioning the geological settings of the area.

ANSWER: This information comes from the numerous articles published about Sierra Nevada lakes. We have included a review article as reference to support this information (Medina-Sánchez et al., 2022).

Geological settings of the area are included in the study site section Lines 120-121): "The geologic substrate in lake catchment basins is composed of slow-weathering, metamorphosed siliceous bedrock mainly mica-schist with graphite and mica-schist with feldspar, devoid of carbonated rocks".

Medina-Sánchez, J. M., Cabrerizo, M. J., González-Olalla, J. M., Villar-Argaiz, M., & Carrillo, P. (2022). High mountain lakes as remote sensors of global change. In *The Landscape of the Sierra Nevada: A Unique Laboratory of Global Processes in Spain* (pp. 261-278). Cham: Springer International Publishing.

- Lines 64-65: Repetitive sentence, we suggest removing: “These features make ... and atmospheric factors (refs)”.

ANSWER: We agree, the sentence has been removed.

- Line 68 and elsewhere: Please be careful with the use of “long-term”. From a geological perspective, and even in the context of the Quaternary sciences, 430 years is not a long-term period. Although coming from biology, it’s understandable why one would use the term 'long-term’. Be aware of the discrepancies in understanding this phrase. And, the sentence on line 68 doesn’t bring anything new, thus can be removed.

ANSWER: We agree, the sentence has been removed.

- Paragraph 5 – This is an important paragraph which would benefit from being mentioned at the beginning of the introduction for the context of the importance of this study.

ANSWER: We agree. We removed paragraphs 3 and 4, so this information comes earlier in the discussion. Furthermore, part of this information has been incorporated into paragraph three in the introduction.

- Line 70-71: missing references. They occur in the rest of the paragraph, though they should also be listed in this sentence.

ANSWER: This sentence now includes a reference to a Springer book chapter. This book chapter provides a review of paleolimnological studies conducted in Sierra Nevada lakes (line 72): “Several paleolimnological studies conducted in Sierra Nevada lakes, focusing on the Anthropocene period (last ~200 years), suggest that rising regional air temperatures, reduced precipitation, and increased P- and Ca-rich Saharan dust deposition are the primary drivers of ecological changes observed in six Sierra Nevada lakes (Pérez-Martínez et al., 2022)”.

Pérez-Martínez, C., Conde-Porcuna, J. M., Ramos-Rodríguez, E., Moreno, E., Rühland, K. M., Jeziorski, A., ... & Jiménez, L. (2022). Paleolimnological indicators of global change. In *The Landscape of the Sierra Nevada: A Unique Laboratory of Global Processes in Spain* (pp. 279-291). Cham: Springer International Publishing.

- Line 74 and throughout the manuscript: we recommend avoiding vague wording like “linked”, “connected”, “complex”, or “associated”; if something is linked or complex, then please clarify the link or complexity in full.

ANSWER: The sentence has been changed following your suggestion (line 72-73): “Additionally, shifts in chironomid assemblages in Río Seco Lake (Sierra Nevada) are also driven by increasing temperatures (Jiménez et al., 2019)”.

- Line 77 - “mountain lakes worldwide” - This is somewhat contradictory; the authors state that the Sierra Nevada is unique due to its location. Exposing it to specific Mediterranean drought and warmth, as well as Saharan dust. Other mountain regions are not influenced by those. Yet, this sentence attempts to demonstrate that the shifts in the Sierra Nevada align perfectly with global trends. This would speak against its uniqueness. Please consider resolving this inconsistency in argumentation.

ANSWER: We agree. The sentence has been removed to avoid misunderstanding. Sierra Nevada is unique due to its high altitude and geographical position, which means it is affected by intensified summer droughts and atmospheric deposition. Significant changes have been observed in its lakes since the 1950s and 1960s.

- Lines 81-82: Information in brackets is unusual and should be limited whenever possible in scientific articles.

ANSWER: Thank you for your suggestion. The information in brackets has been incorporated into the sentence. The brackets have been removed (lines 76-78):" Here, we provide detailed analyses of sedimentary pigments and other biogeochemical indicators such as stable isotopes and the elemental composition of organic matter, from the Little Ice Age to the present, specifically the last ~430 years".

- Lines 91 - 93: Are these examples relevant to this study?

ANSWER: It´s one of the few studies that have been conducted on alpine systems but really it´s a study below treeline and more eutrophic lake. We have removed the sentence.

- Lines 93-94: How is this important in this paragraph? Consider removing this information, as it is already mentioned elsewhere in the manuscript.

ANSWER: We agree, the sentence has been removed.

- Line 97 – Consider rephrasing: "The atmospheric" replace with "We expect that atmospheric"

ANSWER: Thank you, the sentence has been changed (lines 89-91): "We expect that atmospheric input of Saharan-derived phosphorus (P) and calcium (Ca) to be particularly relevant given the naturally low levels of these nutrients in the lake (Jiménez et al., 2018; Pérez-Martínez et al., 2020a)".

- Line 101 – If no other nutrients than P are discussed from this study, consider rephrasing: "and nutrient concentrations – particularly phosphorus" -> "phosphorus concentrations"

ANSWER: We agree. The sentence has been changed (lines 89-91): "We expect that atmospheric input of Saharan-derived phosphorus (P) is particularly relevant given the naturally low levels of this nutrient in the lake (Jiménez et al., 2018; Pérez-Martínez et al., 2020a)".

- Line 104 – To clearly state what is new in this study, we recommend adding "in more detail (compared to Jiménez xxx)"

ANSWER: The paragraph has been changed to include the information you suggest (Lines 93-100):" In this study, we examined changes in algal composition over time in Sierra Nevada lakes for the first time by analysing carotenoid pigments. In addition, compared to previous studies of this lake, in which we examined changes in algal biomass (chlorophyll-a) over the past 180 years, we extended the analysis of algal biomass trends by a further 250 years, i.e. to Little Ice Age, the last period with a defined climate change. Finally, we also extended the period of study of paleoenvironmental proxies such as Carbon:Nitrogen ratio of the sedimentary organic matter (C/N) and added the analysis of new variables such as δ15N and Dissolved Organic Carbon in lake water (DOCw). It is also important to note that, in this study, we used climate series obtained specifically for Sierra Nevada summits by Sigro et al. (2024), whereas in previous studies, we used climate series from sites near the Sierra Nevada".

- Line 105 – Please be specific and list the "key biogeochemical variables"

ANSWER: We have specified the biogeochemical variables in the new sentence included above: Carbon:Nitrogen ratio of the sedimentary organic matter (C/N), $\delta^{15}N$ isotopic composition and Dissolved Organic Carbon in lake water (DOCw).

- Line 108 - Could you, somewhere in the manuscript, clarify why the period of 430 years was selected? To what extent is this already answered in the previous study by Jimenez xxx? Is there any clear motivation to target this specific time period?

ANSWER.: This period was chosen to obtain the changes in the community since the LIA, the last period with a defined climate change. Jiménez et al. (2018) analysed changes in Chl-a, but did not analyse changes in algal assemblage composition. Furthermore, the study by Jiménez et al. (2018) covers the period from 1820 to 2013 CE, whereas our study covers the period from 1600 to 2020 CE.

However, the dating of the oldest period of the study (200 to 430 years B.P.) must be interpreted with caution, as ^{210}Pb dating was only obtained for the first ~200 years, and the oldest sample dates were extrapolated.

Even though the oldest sample dating were not absolutely reliable, we think it was worthwhile to show the evolution of algal biomass and composition over a long period of time.

We say in lines 92-94: "In addition, compared to previous studies of this lake, in which we examined changes in algal biomass (chlorophyll-a) over the past 180 years, we extended the analysis of algal biomass trends by a further 250 years, i.e. to Little Ice Age".

- Lines 107-110: see comments on RQ's, this section would need rephrasing according to the new RQs

ANSWER: Research question has been rephrasing and reduced to two (lines 104-107):

1. A previous study of the last 180 years shows a significant increase in algal biomass over

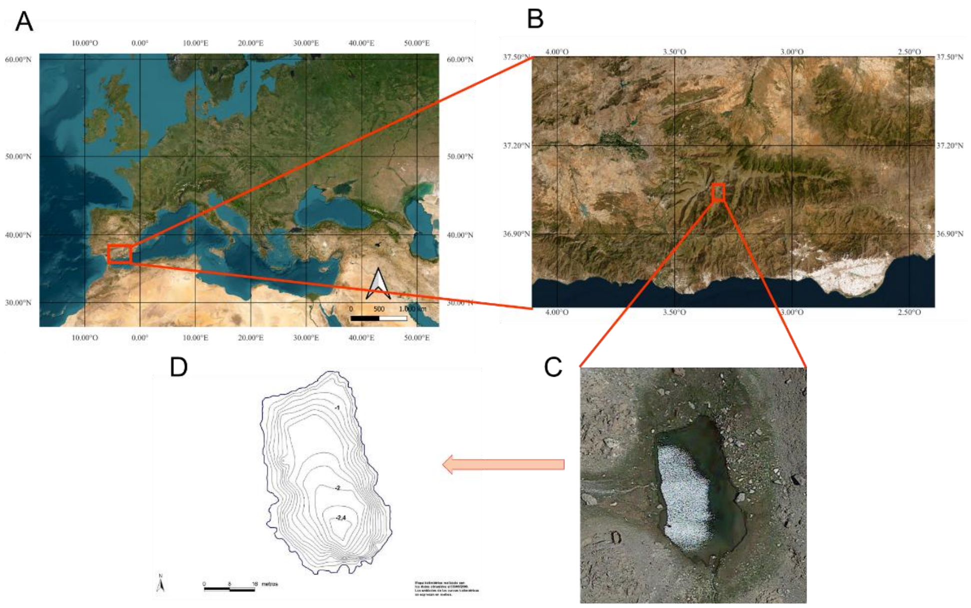
the last 50 years. Is this increase also significant when viewed over the extended period of 430 years?
2. Has the composition of the algal community shifted over this period of time?

- Line 110-112: The last sentence of the introduction section is redundant once the introduction is restructured.

ANSWER: We agree, the sentence has been removed.

Figure 1:

If possible, we suggest using ArcGIS or QGIS software to display a DEM (SRTM) of Spain and its neighbouring countries instead of relying on Google Maps.Furthermore, four maps are displayed, but only three panels (A, B, and C) are described in the figure caption. The orange box in panel A does not represent the map in panel B. Please provide a scale and north arrow for every map. If Figure 1 aims to show where the lake lies in the larger landscape (region), the right part of panel A is needed. However, if the local landscape around the lake and the lake itself and its catchment and bathymetry are the main goals of this figure, then the figure on the right side of panel A can be removed.
ANSWER: You are right, the orange box in panel A does not represent the map in panel B. Figure 1 has been completely revised and redone.



Methods

Overall, the methods lack a significant amount of important information and would benefit from additional details for clarity, reproducibility, and repeatability.

We have added a significant amount of information to the methodology section to improve clarity and reproducibility. Following your suggestions, we have transferred some of the introductory content to the study site section. Furthermore, we have expanded the methodology for analysing pigments, DOC-w and VRS-inferred Chl-a.

This section has grown from less than 5 pages to 6 pages.

Additionally, we have included a new table in the Supplementary Material detailing the main characteristics of Borreguil Lake (Table S1).

2.1 : Study site

This section could be placed separately, apart from methods, if wanted. As already suggested in the introduction, where the study site is also introduced (that section of the introduction can be shortened, and information can be stated mainly here to eliminate repetitive information), it would be beneficial to introduce the bedrock here, thereby placing the low alkalinity information into context. Now it comes a few sentences later, with no further context.

ANSWER: Thank you for your suggestions. We have attended all of it. The study site data of the introduction has been shortened, and information have been relocated in Study site section. Geological data re located in the first paragraph, close to information on low alkalinity (lines 120-123): "The geologic substrate in lake catchment basins is composed of slow-weathering, metamorphosed siliceous bedrock mainly mica-schist with graphite and mica-schist with feldspar, devoid of carbonated rocks. These lakes are typically oligotrophic or oligo-mesotrophic, characterized by cold, oxygen-rich waters with low alkalinity and mineralization (Medina-Sánchez et al., 2022)".

- Line 119-120: Is this information needed for any of the interpretations? If not, this is redundant.

ANSWER: The sentence has been eliminated.

- Lines 124 – 125: The sentence "This study 0.18 ha." deserves to be the first sentence of the section.

ANSWER: We agree. The sentence has been placed first in the section.

Please read the new paragraphs (lines 128-134) in the study site section, which has been thoroughly changed.

- <u>Line 127 –</u> Is there a reference for the watershed vegetation "alpine meadows"? Please add the source of this information.

ANSWER: The source has been added (Line 131). The Flora composition of the alpine meadows has been added in response to a suggestion from Reviewer 2 (see lines 129-132): The lake is surrounded by approximately 0.56 ha of alpine meadows, the flora of which is mainly composed of Cyperaceae, Poaceae and Fabaceae, with frequent genera including Carex, Nardus, Festuca and Scorzoneroides (Pérez-Luque et al., 2015). The wetter meadow shows bryophyte species such as Drepanocladus fluitans."

Pérez-Luque, A. J., Sánchez-Rojas, C. P., Zamora, R., Pérez-Pérez, R., & Bonet, F. J. (2015). Dataset of Phenology of Mediterranean high-mountain meadows flora (Sierra Nevada, Spain). *PhytoKeys*, (46), 89.

- <u>Line 129:</u> The physicochemical data are used further in the discussion (e.g., lines 493-498) but are not stated here; only the reference is provided. We recommend clearly stating the data needed for interpretation in this manuscript, so the reader doesn't need to look them up in the cited publication.

ANSWER: We agree. A short table with location and environmental characteristics of Borreguil Lake has been added to Supplementary Materials. Please see Table S1.

Supplementary Table S1. Location and environmental characteristics of the study lake in Sierra Nevada mountains. Range and mean values (in brackets) or single values of the chemical and biological parameters from water column measurements are shown. Range and mean values are from a minimum of four samples. TP, total phosphorus; TN, total nitrogen; Chl-a, chlorophyll-a.

	Borreguil Lake
Altitude (m asl)	2980
Lake area (ha)	0.18
Catchment area (ha)	50.9
Alpine meadows area (ha)	0.56
Maximum depth (m)	2.5
Maximum volume (m^3)	2070
Conductivity ($\mu S\ cm^{-1}$)	13-15 (14)
Alkalinity ($meq\ L^{-1}$)	0.07-0.1 (0.09)
pH	6.3
Calcium ($mg\ L^{-1}$)	0.8-1.1 (1.0)
Chl-*a* ($\mu g\ L^{-1}$)	1.4-1.7 (1.5)
TP ($\mu g\ L^{-1}$)	13-27 (18)
TN ($\mu g\ L^{-1}$)	180-380 (280)

2.2 Sediment coring and sampling

- Line 136: Is there a particular reason for 26 cm?

ANSWER: It was the sediment core we were able to obtain. Much of them were used in this study.

- Line 137: What was the motivation for the selected sectioning of 0.25cm and, at a certain depth, switching to 0.5cm? Was there any change in lithology (which is completely absent in this manuscript, see the comments below in the section results)? How, was sectioning directly on- site done? Please add more details on core extrusion.

ANSWER: We wanted to have an elevated number of samples in the most modern section of the core because, based on previous results, we expected to see more changes due to climate change and Saharan dust in this part of the core. However, the core was not completely sectioned at 0.25 cm to avoid an excessive number of samples for analyses. There were no appreciable changes in lithology in this 26 cm core.

The core was sectioned with a core extruding apparatus provided by Aquatic research Instruments (http://www.aquaticresearch.com/core_extruding_apparatus.htm). A detailed procedure tutorial is provided in this last web page as well in https://www.queensu.ca/pearl/methods/extruding.php.

We have added Glew et al. (2001) reference in the text (lines 158): “The sediment core was sectioned on-site following Glew et al. (2001) at 0.25 cm intervals for the first 9.25 cm and at 0.5 cm intervals for the remaining sediment”.

References:

- Glew, J.R. 1988. A portable extruding device for close interval sectioning of unconsolidated core samples. J. Paleolimnology 1: 235-239.
- Glew, J.R., Smol, J.P. and Last, W.M. 2001. Sediment core collection and extrusion. pp. 73-105. In: Last, W.M. and Smol, J.P. [Editors]. Tracking Environmental Change Using Lake Sediments. Vol 1: Basin Analysis, Coring, and Chronological Techniques. Kluwer Academic Publishers, Dordrecht.

- Lines 138 – 139: remove “using disposable material to prevent contamination between samples” - this is common sense.

ANSWER: We think this is a crucial point of the method. We have reduced the sentence (lines 160-162): “For each section, 1 cm3 were immediately taken from the centre of the core using disposable material ~~to prevent contamination between samples,~~ and stored in small plastic polipropilene 5 cm3 vials for pigment analysis avoiding the exposure of the sediment to light”.

- Lines 139-140: What type of plastic? PET, PP, HDPE? Volume of the vials? Was there an attempt to keep the headspace minimal to avoid pigment degradation by oxygen? How about temperature? Did you store the samples at -20°C whenever possible, or at least at 4°C? All this information is crucial for the method's replicability. These can shortly be added here or stated.

ANSWER: This information has been added. Please, see lines 160-164: "For each section, 1 cm3 were immediately taken from the centre of the core using disposable material and stored in small plastic polipropilene 5 cm3 vials for pigment analysis avoiding the exposure of the sediment to light. The vials were completely full of sediment to avoid pigment degradation by oxygen, and were kept at 4°C during transportation to the lab. Once in the laboratory, the vials were immediately placed at -80°C and at -20ºC after freeze-drying".

- Line 141: Any reasoning for storing the freeze-dried sample in 4°C instead of a desiccator, frozen, or just at room temperature?

ANSWER: You are right, samples were stored in a dessicator (line 165).

- Lines 140 – 141: remove "for later analyses"

ANSWER: It has been removed from the sentence.

: Chronology

Were the data checked for missing inventory? (https://doi.org/10.1016/j.quageo.2020.101106)

ANSWER: The ^{210}Pbtot profile was complete, i.e. ^{210}Pbtot reaches equilibrium. The dating results have been published on Git-hub and are available for consultation (https://github.com/carmen6363/Dating-results-core-SSBG-21).

- Line 149 – Is the Fukushima accident marker actually used in this study? If not, then this information may be removed.

ANSWER: The Fukushima accident mentioned has been removed. This marker was not used in this study.

2.3 : Carotenoid pigment analysis

- Line 151: Does the number of samples 51 include full chemical replicates (e.g. parallel samples from the core) as well as blanks? Additionally, were the freeze-dried samples stored at room temperature or at the above-mentioned 4°C? It should also be noted that freeze drying has a negative impact on carotenoids; rare carotenoids may not be detected due to degradation during freeze drying. Although these findings have not been published yet, this information should be taken into account when interpreting the pigment data.

ANSWER: The 51 samples do not include chemical replicates or blanks. Typically, a single sediment section from a core does not provide enough material to prepare replicates, and blanks are not used in this type of analysis because high-purity solvents eliminate the risk of contamination. After freeze-drying, sediment samples were stored at −20 °C (we have included it in the text, line 164). Regarding the comment on carotenoid degradation, our observations do not fully support this concern. We compared pigment concentrations in samples processed with and without freeze-drying and found no qualitative differences—the same pigments were detected in both cases. Although minor quantitative differences were observed for a few pigments, these did not outweigh the practical advantages of freeze-drying. Working with fresh sediment introduces challenges such as unknown water content, which affects extraction efficiency. For example, when using fresh sediment, acetone concentration varied between 80-90%, whereas with freeze-dried sediment it was consistently 90%. If water content exceeds 10% during extraction, pigment recovery decreases, likely due to degradation into non-chromophoric compounds that are undetectable by the system.

- Line 153: Was a cooled centrifuge used? Or how was centrifugation achieved at 4°C?

ANSWER: Yes, centrifugation was performed using a refrigerated centrifuge set to 4 °C, as indicated in the methods section (line 176).

- Line 154: We encourage authors to specify any modifications made to the HPLC method. Furthermore, the reproducibility of the pigment analysis based on replicates or internal standards is lacking. This would be most necessary, as different carotenoids usually have quite different reproducibility.

ANSWER: Please, see lines 178-189: "The UHPLC system used was a Waters Acquity UPLC equipped with a PDA detector (λ 300–800 nm), refrigerated autosampler (4 °C), and upgraded with a Hexa/THF kit and 20 µL loop (Waters, Milford, MA, USA). Analytical separations were performed on an HSS C18 SB column (100 × 2.1 mm, 1.8 µm particle size) at 25 °C. The PDA channel was set at 440 nm for pigment detection and quantification. After sample injection (7.5 µL), a constant flow rate of 0.7 mL min^{-1} was maintained. Pigments were eluted using a linear gradient from 100% solvent B (51:36:13 methanol:acetonitrile:Milli-Q water, v/v/v, with 0.3 M ammonium acetate) to 75% B and 25% A (70:30 ethyl acetate:acetonitrile, v/v) over 3 min, followed by a 0.45 min isocratic hold at 75% B and a 2 min gradient to 100% A. Initial conditions (100% B) were restored in 0.65 min, with a 1 min re-equilibration before the next injection. To address reproducibility, chromatographic performance was evaluated by calculating the relative standard deviation (RSD) for retention times and peak areas across replicate injections. All pigments exhibited RSD values within commonly accepted limits for HPLC analyses (typically ≤1% for retention time and ≤2% for peak area), confirming that the method provides consistent and reliable results under the tested conditions".

- Lines 157-161: Please add references for the interpretations of individual pigments. Perhaps a table with the pigment name, its interpretation, retention time (in the used method), molecular weight, and references would be helpful.

ANSWER: Including a detailed table with pigment interpretations, retention times, molecular weights, and references would be more appropriate for a methodological paper rather than the current study, which focuses on paleoenvironmental reconstruction. However, we agree that this information can be useful for readers. Therefore, we propose to include it as supplementary material (Table S2) or refer readers to the comprehensive resource already published in [Buchaca and Catalan, 2007, 2008)], where these details are fully documented.

Buchaca & Catalan (2007): Factors influencing the variability of pigments in the surface sediments of mountain lakes. Freshwater Biology, 52, 1365–1379.

Buchaca, T. and Catalan, J.(2008) On the contribution of phytoplankton and benthic biofilms to the sediment record of marker pigments in high mountain lakes, J. Paleolimnol., 40, 369–383, https://doi.org/10.1007/s10933-007-9167-1.

Supplementary Table S2. Pigments identified from lake sediments included in this study and their taxonomic affinities. The number of functional groups (FG) of the pigment molecular structure is also indicated for carotenoids.

Peak	RT (min)	PeakCode	Full name	lmax (nm)	Taxonomic affinity	FG
1	0.69	Ph_0	Chlorophyllide-a	432, 665	Chl-a degradation product, abundant in diatoms	
2	1.02	Ph_3	Chl-c1	441, 635	Diatoms and Chrysophytes	
3	1.04	Car_7_8	UVA2- Scytonemin	387	Cyanobacteria exposed to high UV radiation	4
4	1.09	Ph_5b	Chl-c2	448, 636	Cryptophyta, Dinoflagellates, Diatoms and Chrysophytes	
5	1.53	Car_5	Fucoxanthin	452	Chrysophytes and diatoms	5
6	1.44	Ph_2	Phaeophorbide-b1	438, 650	Chl-b degradation product (grazing processes)	
7	1.92	Ph_1a	Phaeophorbide-a	411, 664	Chl-a degradation product (grazing processes)	
8	2.19	Ph_2a	Phaeophorbide-b2	438, 650	Chl-b degradation product (grazing processes)	
9	2.32	Car_9z	Asthaxanthin	478	Zooplankton (Crustacea)	4
10	2.40	Car_17d'	Aphanizophyll	450, 477, 507	N2-fixing cyanobacteria	3
11	2.52	Car_10a	Diadinoxanthin	449, 479	Dinoflagellates, chrysophytes and diatoms	3
12	2.71	Car_12_5	Antheraxanthin	448, 477	Charophyceae, Rodophyta and vascular plants	3
13	2.91	Car_18	Alloxanthin	456, 484	Cryptophytes	2
14	3.11	Car_17	Diatoxanthin	456, 485	Diatoms and chrysophytes	2
15	3.21	Car_11	Lutein	448, 475	Chlorophyta	2
16	3.31	Car_12	Zeaxanthin	454, 483	Cyanobacteria, Chlorophytes	2
17	3.55	Car_16	Canthaxanthin	472	Cyanobacteria and zooplankton (Cladocera)	2
18	3.85	Ph_13	Chl-b	462, 650	Chlorophyta	
19	3.95	Car_17_3	Unknown	448, 475		
20	4.21	Ph_19a	allomer Chl-a	433, 663	Chl-a degradation product	
21	4.40	Ph_19	Chl-a	431, 662	Cyanobacteria, algae and vascular plants	
22	4.51	Ph_19e	epimer1 Chl-a	431, 662	Chl-a degradation product	
23	4.65	Ph_19e'	epimer2 Chl-a	431, 662	Chl-a degradation product	

24	4.73	Car_21	Echinenone	470	Cyanobacteria and zooplankton (Cladocera)	1
25	4.95	Ph_23	Phaeophythin-b1	435, 651	Chl-b degradation product (senescence)	
26	4.99	Car_25a	b-carotene	421, 454, 480	Cyanobacteria, algae and vascular plants	0
27	5.07	Ph_25	Phaeophythin-a1	410, 665	Chl-a degradation product (senescence)	
28	5.19	Ph_26	Phaeophythin-b2	436, 651	Chl-b degradation product (senescence)	
29	5.27	Ph_28	Phaeophythin-a2	410, 665	Chl-a degradation product (senescence)	

- Line 159: Just out of interest, the references to zooplankton, how do they actually work? I have not seen these pigments particularly associated with zooplankton before. Hence, illustrating the need to outline the interpretational framework here (see later comments in results).

ANSWER: The interpretational framework for pigments associated with zooplankton is detailed in Buchaca & Catalan (2007): Factors influencing the variability of pigments in the surface sediments of mountain lakes. Freshwater Biology, 52, 1365–1379. In this study, certain pigments (e.g., those derived from zooplankton diet or gut content) were shown to accumulate in sediments and serve as indirect indicators of zooplankton presence and activity, supporting their use in paleoecological interpretation.

- Lines 161–163: Suggestion: “there is a ... deposited and buried (ref)” -> “carotenoids are less prone to degradation to colourless compounds (ref).”

ANSWER: Thank you. The sentence has been changed (lines 196-197): ”We based our interpretation on carotenoid profiles instead of phorbins because carotenoids are less prone to degradation to colourless compounds (Buchaca and Catalan, 2008)”.

- Line 164: Were the 10 selected pigments calibrated to concentrations, or only the peak areas used to determine the relative abundance %? Is there any reasoning behind expressing pigments as relative abundances? The closed sum at 100% needs to be justified, as a preselection of pigments is used. If the aim is to compare only specific primary producers, this is possible, but if the aim is to explore shifts of the entire PP community, the absolute concentrations, in umol/DW or umol/OC or OM, would be a better way to present the data. We further encourage authors to report the retention times and relative downcore abundance changes (peak area) of unknown pigments in the supplementary or any open-source database. This helps the pigment community. Additionally, an example of a chromatogram (for example, at 460nm) with annotations may be included in the supplementary data to enhance the reproducibility of the modified HPLC method. This makes the research more reproducible, transparent, usable, and citable.

ANSWER: Concentrations were calculated as nmol/g DW for all pigments, including the 10 selected marker pigments. These 10 pigments were chosen because they are well-preserved and taxonomically informative, allowing us to assess the relative dominance of major algal and cyanobacterial groups throughout the sediment profile. The rationale for selecting marker pigments in palaeolimnology generally follows two criteria: (i) specificity to a taxonomic group (usually at a resolution of Division or Class level) or ecological process (e.g., grazing) and (ii) low lability (low number of functional groups), ensuring persistence in the sedimentary record. Chlorophylls are typically more labile than carotenoids, although some degradation products (e.g., phaeophytins) can be highly persistent (Scheer, 1991). Carotenoid stability depends on molecular structure and tends to decrease with the number of functional groups (Britton et al., 1995; Sinninghe Damsté & Koopmans, 1997).

The selected pigments provide coverage of all major algal and cyanobacterial groups. To address the reviewer's suggestion, we have included as supplementary material a plot showing pigment profiles of the 10 selected pigments expressed as concentrations (nmol PGM / g DW) (Figure S2) and additional methodological details like the retention time and maximum absorption spectra (lmax) of all pigments detected as a supplementary table (Table S2).

Our goal is to assess changes in community composition rather than the absolute amount of pigments present in each period, so we chose to report pigment concentrations in terms of relative abundance. This approach helps us to avoid the risk of changes in sedimentation rates affecting pigment concentrations.

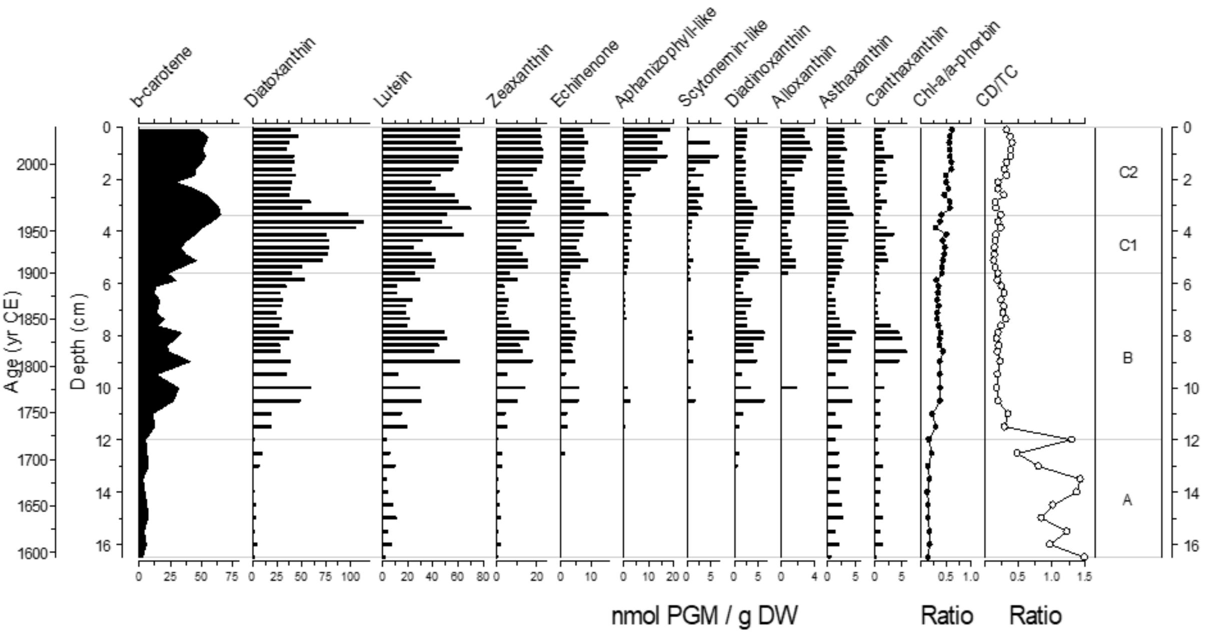
To assess lake productivity, we used ng/g DW of β-carotene and mg/g DW of chlorophyll-a.

Britton, G. (1995). Structure and properties of carotenoids in relation to function. *The FASEB Journal*, *9*(15), 1551-1558.

Scheer, H. (1991). Structure and occurrence of chlorophylls. En H. Scheer (Ed.), *Chlorophylls* (pp. 3–30). CRC Press.

Sinninghe Damsté, J. S., & Koopmans, M. P. (1997). The fate of carotenoids in sediments: an overview. *Pure and applied chemistry*, *69*(10), 2067-2074.

Supplementary Figure S2. Pigment profiles of the 10 selected marker pigments and b-carotene expressed as concentrations (nmol PGM / g DW) and the ratios of Chl-a/a-phorbin as a Chla preservation index and the ratio CD/TC (chlorophyll-a derivatives vs. total carotenoids) as indicative of higher contribution of autochthonous production when the ratio is low. Four main biostratigraphic zones were established based on constrained incremental sum of squares partitioning of pigment concentration with Rioja-package version (0.9-15.2) (Juggins, 2017) using R version 3.5.3 (R Core Team 2019).



2.4 : Biological and paleoenvironmental data analysis

- Line 167-168: Was an existing decalcification method used? If so, please cite it. If not, please specify volumes of added HCl.

ANSWER: This information has been included in the text (see lines 205-207): “For elemental analyses of carbon and nitrogen content of the organic matter, a sediment fraction of ~ 1 g of wet weight (WW) sediment was freeze-dried. To remove inorganic carbonates, samples were acidified in situ by adding 1M HCl dropwise until effervescence ceased and overnight at 50 ºC”.

- Line 171-172: This sentence could be used as the explanation why these analyses were done, and thus we suggest moving it to the start of this paragraph to make the general statement about C/N.

ANSWER: We agree, the sentence has been moved to the start of the paragraph (Lines 202-03).

- Line 172: The reference of Meyers and Ishiwatari (1993) is missing in the reference list and is likely misplaced. Other references are meant here.

ANSWER: We agree. We have left in the text only the reference of Meyers and Teranes (2001).

- Line 175: Was the isotope analysis done on different samples than C/N? Also, please add the number of samples used/analyzed by both methods, as well as the method of accuracy (error or SD).

ANSWER: The isotope analysis were done on the same samples than C/N analysis.

This information has been included in the text (line 208): “Nitrogen isotope (δ15N) composition was determined from the same bulk sediment organic matter used for the C/N analyses. It was performed on the remaining carbonate-free sediment samples using the elemental analyser Flash HT Plus coupled to IRMS DELTA V Advantage at the CIC".

Line 232: A total of 51 samples were used for C/N, δ15N and for VRS-inferred Chl-a and DOCw analyses.

- Line 177-179: This sentence is not needed in methods, as the data are discussed later. Please consider our previous comment regarding the use of vague words like "complex".

ANSWER: The sentence has been eliminated

- Lines 180-183: We encourage authors to introduce both methodologies briefly so that the reader can easily follow the data interpretation. Additionally, it would be advisable to explain the motivation behind using these methods and why relying solely on the TOC from the elemental analysis and sedimentary pigments is insufficient. Are these data, anyhow, comparable to observational data from the current lake water column?

ANSWER: We revised the method section and added a brief explanation of both methodologies to the text (lines 213-231): "Sediment samples were analysed for inferred trends in total lake-water organic carbon following the methodology described in Meyer-Jacob et al. (2017) and sedimentary chlorophyll-a including its derivatives (VRS-inferred chl-a) were analysed according to Michelutti et al. (2010) at the Paleoecological Environmental Assessment and Research Laboratory (PEARL), Queen's University (Canada). Briefly, prior to analyses, lyophilized sediment samples were sieved (125 µm mesh) to remove the influence of particle size and water content on the spectral signal. Next, VNIR spectra were recorded with a FOSS XDS Rapid Content Analyzer in diffuse reflectance mode. Each sediment sample spectra represents a mean of 32 scans at 2 nm resolution in the wavelength range from 400 to 2500 nm. For VRS-inferred chl-a, the absorbance peak of chlorophyll-a between 650 and 700 nm was considered in relation to calibration samples covering a range of sedimentary chlorophyll-a concentrations, as measured by high performance liquid chromatography (HPLC). Sedimentary VRS chl-a concentrations were inferred using log transformed data from Michelutti et al. (2010) with the equation: chl a + derivatives = EXP (0.83784 * LN (peak area 650–700 nm) + (−2.48861)).Our chlorophyll-a inferences, which include all algal and cyanobacterial groups, also include all chlorophyll isomers and its major derivatives (pheophytin and pheophorbide) (Michelutti et al., 2010; Michelutti and Smol, 2016). For inferred water TOC, an orthogonal partial least squares regression model between VNIR spectral information (400-2500 nm) and measured surface-water TOC concentrations in 345 lakes was used to reconstruct lake-water TOC levels from sediment samples (Meyer-Jacob et al, 2017). In our case, lake-water TOC is considered virtually equivalent to DOC in lake water due to the dominant contribution of DOC in the Sierra Nevada lake's organic carbon pool

(Mladenov et al., 2008), a pattern also observed in lakes with similar characteristics (Meyer-Jacob et al., 2019). From this point onward, analyzed TOC in water will be referred to as DOCw, as we consider relative changes in TOC to reflect relative changes in DOC in the water."

The VNIRs method we use is fast and efficient and importantly captures contributions from all algal and cyanobacteria groups, as well as all the isomers of chlorophyll-a and its main diagenetic products (i.e. pheophytin-a and pheophorbide-a).

Lake-water DOC is a key driver of aquatic ecosystem functioning and cannot be reliably inferred from sedimentary TOC, as temporal changes in these two indicators are not necessarily synchronous. Our lake-water TOC/DOC inference approach has been shown to successfully track long-term trends in water-chemistry monitoring data over several decades, and previous studies have demonstrated the long-term effects of land use, acid deposition, and climate change on TOC and DOC dynamics in lakes across Canadian, the UK, and Sweden (Meyer-Jacob et al. 2015; 2017; 2019). Observational data for lake-water TOC or DOC in the study lake is limited to only a few samples and therefore does not allow an evaluation of longer-term trends.

We do not rely solely on DOCw (TOC) values; we also use the C/N ratio, ^{15}N and, moreover, chlorophyll-a and B-carotene as explanatory variables. Additionally, we use climate and aerosol data as explanatory variables. We believe this constitutes an important group of variables with which to relate sedimentary pigments.

DOCw values enable us to analyse the light conditions in the lake and correlate them with the presence of UV-protective pigments. The C/N ratio enables us to track the evolution of algal biomass and possible nutrient limitation, and the 15N ratio enables us to link changes in abundance to the presence of nitrogen-fixing pigments. Finally, aerosol and instrumental climate data are powerful variables with which to explain pigment dynamics. Although more variables could be included in a study, we believe that our results are robust and can be adequately explained by these variables.

The obtained DOC values are similar to those published for Sierra Nevada lakes, although there is no data available for Borreguil Lake. However, there are data for different Sierra Nevada lakes in Mladenov et al. (2008). This article reports values ranging from 0.047 to 0.300 mmol/L (i.e. 0.6 to 3.6 mg/L) in several Sierra Nevada lakes. Our values range from 3.5 to 4.5 mg/L.

Meyer-Jacob C., Tolu J., Bigler C., Yang H., Bindler R. 2015. Early land use and centennial scale changes in lake-water organic carbon prior to contemporary monitoring. Proc. Natl.

Acad. Sci. 112(21): 6579–6584.

Meyer-Jacob, C., Michelutti, N., Paterson, A.M., Monteith, D., Yang, H., Weckstrom, J., Smol, J.P. and Bindler, R., 2017. Inferring past trends in lake water organic carbon concentrations in northern lakes using sediment spectroscopy. Environmental Science & Technology, 51(22), .13248-13255.

Meyer-Jacob C, Michelutti N, Paterson AM, Cumming BF, Keller W, Smol JP (2019) The browning and re-browning of lakes: divergent lake-water organic carbon trends linked to acid deposition and climate change. Sci Rep 9:16676. https:// doi. org/ 10. 1038/ s41598- 019- 52912-0

Mladenov, N., Pulido-Villena, E., Morales-Baquero, R., Ortega-Retuerta, E., Sommaruga, R., & Reche, I. (2008). Spatiotemporal drivers of dissolved organic matter in high alpine lakes: Role of Saharan dust inputs and bacterial activity. *Journal of Geophysical Research: Biogeosciences*, *113*(G2).,

- Line 181 – Please state the full name of the method VRS when it is first mentioned.

ANSWER: The full name has been included (line 214): "sedimentary chlorophyll-*a* including its derivatives (visible reflectance spectroscopy (VRS) inferred chl-*a*) according to ……".

- Lines 184-185: The assumption that TOC = DOC is bold, but if it's based on linked monitoring data to surface sediment data, it can be used. Though one should be careful, as with changing vegetation in the watershed, this may not be applicable to older stages of the lake. Additionally, the water TOC model of Meyer-Jacob et al. is based on northern lakes. Is it really applicable to this mountainous lake? A more conservative interpretation should be considered, such as relative changes in TOC to reflect relative changes in DOC in the water. It is more likely that the 1:1 relationship does not exist, as there is at least a fractionation of DOC being recycled, and only a small part is buried: e.g., in the Boreal region, where the carbon burial is expected to be higher, the mean C burial is 22% from the incoming C to the lake: https://doi.org/10.1002/2013JG002345. Ultimately, does the DOC data alter the entire narrative? Isn't the TOC data sufficient?

A) The DOC/TOC ratio in oligotrophic lakes is greater than 0.9 (Wetzel, 2001, p 748). Specifically, in Chapter 23, Table 23.1 Wetzel shows that in most lakes, the DOC exceeds the POC by a factor of about 6:1 to 13:1. Other remote lakes where a high DOC/TOC ratio has been measured are discussed in Rosen (2005). Mattson et al. (2005) also found that the majority of TOC is in dissolved form, and Kokelj et al. (2009) found that DOC accounted for 96% of TOC (DOC–TOC Spearman rank correlation $r^2 = 0.99$, p-value < 0.0001). Evans et al. (2017) assumed that TOC ~ DOC in their study. Meyer-Jacob found that, on average, DOC comprised 87% of the TOC pool in 241 alpine and Arctic lakes. Members of our research group (led by Professor I. Reche) have measured DOC and TOC in Sierra Nevada lakes and found that DOC accounts for more than 90% of the total TOC concentration. Unfortunately, these data have not been published.

As we say in the previous question, lake-water DOC cannot be reliably inferred from sedimentary TOC. Our lake-water TOC/DOC inference approach has been shown to successfully track long-term trends in water-chemistry monitoring data over several decades, and previous studies have demonstrated the long-term effects of land use, acid deposition, and climate change on TOC and DOC dynamics in lakes across Canadian, the UK, and Sweden (Meyer-Jacob et al. 2015; 2017; 2019).

Evans, C. D., Malcolm, I. A., Shilland, E. M., Rose, N. L., Turner, S. D., Crilly, A., ... & Monteith, D. T. (2017). Sustained biogeochemical impacts of wildfire in a mountain lake catchment. Ecosystems, 20(4), 813-829.

Meyer-Jacob, C., Tolu, J., Bigler, C., Yang, H., & Bindler, R. (2015). Early land use and centennial scale changes in lake-water organic carbon prior to contemporary monitoring. Proceedings of the National Academy of Sciences, 112(21), 6579-6584.

Meyer-Jacob, C., Michelutti, N., Paterson, A. M., Monteith, D., Yang, H., Weckstrom, J., ... & Bindler, R. (2017). Inferring past trends in lake water organic carbon concentrations in northern lakes using sediment spectroscopy. Environmental Science & Technology, 51(22), 13248-13255.

Meyer-Jacob, C., Michelutti, N., Paterson, A. M., Cumming, B. F., Keller, W., & Smol, J. P. (2019). The browning and re-browning of lakes: Divergent lake-water organic carbon trends linked to acid deposition and climate change. *Scientific reports*, *9*(1), 16676.

Rosén P. (2005): Total organic carbon (TOC) of lake water during the Holocene inferred from lake sediments and near-infrared spectroscopy (NIRS) in eight lakes from northern Sweden. – Biogeochemistry 76: 503-516.

Wetzel, R. G. (2001). Limnology: lake and river ecosystems. 3rd edition. Academic Press, San Diego.

B) There is no published data on vegetation changes in Borreguil Lake, but there are data from a nearby lake in Sierra Nevada named Río Seco. This study found that there have been no significant changes to the vegetation surrounding the lake in the last 400 years.

Anderson, R. S., Jiménez-Moreno, G., Carrión, J. S., & Pérez-Martínez, C. (2011). Postglacial history of alpine vegetation, fire, and climate from Laguna de Río Seco, Sierra Nevada, southern Spain. *Quaternary Science Reviews*, *30*(13-14), 1615-1629.

C) TOC model of Meyer-Jacob et al. is based on northern lakes. Like many of the northern lakes studied by Meyer-Jacob et al. (2017), the lakes in the Sierra Nevada are located above the tree line and surrounded by meadows. Furthermore, the climate and precipitation range in the Sierra Nevada are similar to those of the lakes studied by Meyer-Jacob et al. (2017). The depth, alkalinity and trophic state of Borreguil Lake are also within the range of the lakes studied by Meyer-Jacob et al. (2017).

D) A more conservative interpretation should be considered-We agree that a more conservative interpretation should be considered, taking into account that not all the DOC is buried. The last sentence of this paragraph has been amended in the text (see line 230): "From this point onwards, analysed TOC in water will be referred to as DOCw, as we consider relative changes in TOC to reflect relative changes in DOC in the water".

E) Does the DOC data alter the entire narrative? Isn't the TOC data sufficient?

While we do not consider the DOC data to alter the entire narrative, we specifically want to refer to DOC lake water and its influence on the UV light climate in the lake. We discuss changes in DOCw in relation to changes in scytonemin, a pigment that protects against UV radiation. The concentration of DOC is inversely related to water transparency (Morris et al., 1995), thereby reducing solar radiation in the water column.

2.5 Climate data

As these builds on previous publications, we suggest keeping this section brief, considering using only one of the two wNAO or SPI to simplify the story and avoid constantly discussing the differences between them. If possible, it would be even better to show the Zr/Al ratios from this nearby lake as a proxy for the deposited dust.

ANSWER: We agree, wNAO has been deleted from the manuscript.

As we report in our manuscript the Zr/Al data is described in an already published paper (see lines 243-247): "Jiménez et al. (2018) demonstrated that the SPI and wNAO indices can be utilized as predictors of the transport and intensity of Saharan dust events in Sierra Nevada, reflecting atmospheric P and Ca deposition trends in this region. These indices exhibited strong correlations with the zirconium-to-aluminium (Zr/Al) ratio—a proxy for Saharan dust deposition—measured in a sediment core from one of the Sierra Nevada lakes, as well as with Saharan calcium concentrations in an ice core from the French Alps, which are indicative of Saharan dust events (Preunkert and Legrand, 2013).".

We do not think it makes sense to show the Zr/Al ratio and its relationship with the Saharan dust proxies, as this would require considerable expansion of the text, which is not desirable by the reviewer. Moreover, the Zr/Al ratio data from Río Seco Lake covers a shorter time period than the SPI and wNAO series (Jiménez et al., 2018). Therefore, we believe that it is more appropriate to use the SPI and wNAO series once their relationship with Zr/Al ratio and with the Saharan calcium concentrations in an ice core from the French Alps has been previously demonstrated. All this data and its relationships are extensively explained in Jiménez et al. (2018). As the reviewer says, we try to keep this section brief and it has already been considerably extended by including details of the pigment and TOC/DOC analyses.

- Line 194: Why was Pearson correlation used? Were the data tested for normality before?

ANSWER: Yes, data were tested by Kolmogorov- Smirnov y Shapiro-Wilk tests for normality (all p> 0.5). It has been added to the sentence (line 239): "Data were tested for normality before the analysis".

Lines 209-211: Please clarify the binning method: Were the data binned into arbitrary sampling intervals, and then an average was taken?

ANSWER: The data were binned in exactly the same intervals as those obtained from the sediment core. This information has been added to the text (see lines 253-255):"The annually resolved climate and dust metrics were averaged over the period of accumulation for each dated interval, i.e. the data were binned in exactly the same intervals as those obtained from the sediment core, thereby integrating the instrumental data with the paleolimnological data".

- Lines 205 – 208: Please consider rephrasing this long sentence into two shorter ones.

The sentence has been split into two shorter ones. Moreover, we have changed the website of the SPI, as it has recently been relocated (lines 248-251): "The Sahel precipitation index (SPI), extending back to 1900 yr CE, was accessed from the NOAA Physical Sciences laboratory (https://psl.noaa.gov/data/timeseries/month/SAHELRAIN/). It provides a standardized rainfall index data for the Sahelian zone of northern Africa".

2.6 : Statistics

The overall impression from this section is that the study includes an excessive number of statistical data treatments without clearly stating the motivation or the need behind using the individual statistical approaches. Every statistical analysis is biased by the selection of variables, and thus, the selection of variables needs to be justified by the aim/RQ that the data should answer. Thus, we strongly recommend using descriptive statistics selectively and with clear objective reasoning.

We encourage authors to make their workflow, as well as data, open source on GitHub or other open- source platforms. This makes the paper stronger, more applicable, citable, and especially reproducible.

ANSWER: Statistical analyses have been reduced and their objective clearly stated. Please, see our answers to specific comments on Statistics.

The analyzed pigment and paleoenvironmental data (https://github.com/carmen6363/Pigment-article-variables-2026) and statistical information (https://github.com/carmen6363/RDA-pigment) have been published in open- source platform Github. The GLM analysis codes have not been published because we performed the analysis directly from the R Commander window.

Dating results have been also published (https://github.com/carmen6363/Dating-results-core-SSBG-21).

The analyzed pigment and paleoenvironmental data will be also published in the PANGAEA repository. We have previously utilized this platform for datasets related to the Sierra Nevada core analyses. The current dataset has already been submitted and is undergoing the standard review process by PANGAEA; consequently, a DOI has not yet been assigned.

Line 213 - 215 – It may be hased as follows: "Before statistical analysis, data were checked for normality and further standardized using a Hellinger transformation to remove asymmetry and incomparabilities in the total variance." We also suggest adding the reasoning behind the selection of the data transformation.
ANSWER: In the first sentence of the paragraph, we refer to both explanatory and explicative variables. Following your recommendations, we will add reasoning of transformation to each set of variables.
The sentences remain as follows (lines 258-261): "Before statistical analyses were applied, data were transformed to reduce the asymmetry of the distributions and to standardise them, i.e. remove incomparabilities in the total variance. The pigment variables (expressed as relative abundance) were square root transformed (Hellinger transformation) to minimize the impact of dominant species and handle the sparse nature of the dataset (presence of many zeros)".

- Line 215: An example of how text can be shortened throughout the manuscript for better readability: "on the basis that" =>"because"

ANSWER: Thank you.

Line 216-217 – Please state the explanatory variables and add the reasoning behind using the specific data transformation
ANSWER: The sentences have been changed (lines 258-262): "The explanatory biological and paleo-environmental variables (C/N ratio, DOCw, chlorophyll-a and β-carotene) were logarithmically transformed followed by a relative scale transformation (y'=yi/ymax) to remove asymmetry and incomparabilities in the total variance. The δ15N values were used untransformed as these met the conditions of normality and homoscedasticity. Finally, the

climate and dust metric variables (MATA, APA, wNAO and SPI) were relative-scale transformed".

Pigment data, expressed in relative abundance were Hellinger transformed which is a measure recommended for clustering or ordination of species abundance data (Legendre and Birks, 2012), having into account this kind of data contains many zero values. Hellinger focuses on relative abundance, mitigating the problem of zero values. Moreover, the square-root component of the transformation stabilizes variance, preventing highly abundant species from masking the patterns of rare ones.

Physical variables are not expressed in the same physical units and need to be transformed to eliminate physical dimension before being used together for ordination. Relative scale variables transformation ranges the values, eliminating physical dimensions (Legendre and Birks, 2012). By centering and scaling, you shift the focus to the shape of the variation rather than the magnitude.

Both transformations linearize relationships since most multivariate techniques (like PCA or Clustering) rely on Euclidean distance.

Legendre, P., & Birks, H. J. B. (2012). From classical to canonical ordination. In *Tracking environmental change using Lake sediments: Data handling and numerical techniques* (pp. 201-248). Dordrecht: Springer Netherlands.

- Line 219-220 – These sentences can be changed, or placed elsewhere in the paragraph, as we suggest using the context of those further up in the paragraph to clarify the reason why the data were transformed. The d15N sentence can be incorporated further above, e.g. after the sentence about transformation.

ANSWER: The sentence has been replaced (see lines 262-266):" The explanatory biological and paleo-environmental variables (C/N ratio, DOCw, chlorophyll-a and β-carotene) were logarithmically transformed followed by a relative scale transformation (y'=yi/ymax) to remove asymmetry and incomparabilities in the total variance. The δ15N values were used untransformed as these met the conditions of normality and homoscedasticity. Finally, the climate and dust metric variables (MATA, APA, wNAO and SPI) were relative-scale transformed".

- Lines 221-222: The sentence starting "Changes in ... algal groups" can be removed, as it is already introduced in the pigment section

ANSWER: The sentence has been removed

- Lines 222-225: An example of shortening the text for increasing the readability of the section:"The number of significant pigment zones (CONISS; constrained hierarchical clustering; Grimm, 2011) was determined using the broken stick model."

ANSWER: Thank you for your suggestion. The paragraph has been changed (lines 268-269):

”The number of significant pigment zones (CONISS; constrained hierarchical clustering; Grimm, 2011) was determined using the broken stick model (Bennet, 1996)”.

- Lines 227: It is somehow unclear why the data were split, and two separate PCAs were conducted.

Two separate PCA analyses were conducted for two time periods: The first period covers ~430 years for which no climate or atmospheric data are available (from ~1600 CE to the present), and the second period covers ~115 years for which climate and atmospheric data are available (from ~1850 CE to the present).

However, the whole paragraph has been changed to solely describe the PCA performed on 430 years data (lines 269-272). We have done this to reduce the amount of space taken up by text and graphics, and because it is not necessary to show both PCAs when discussing the article.

- Lines 235: Similar to the comment above, it is not explained why AIC was selected.

A sentence has been added to explain why AIC was selected (Line 284): “AIC was selected because it helps identify the most parsimonious model for explained the variation in algal community assemblage”.

- Lines 247-248: “for the period ... CE; 22 samples)” - Yes, this is logical, but it would be better to read at the top before the GLM and RDA details are mentioned. Similar to other parts of this section, the purpose of the used statistics is not well explained.

ANSWER: The sentence has been placed at the top of the paragraph (current lines 273-275).

The purpose of using GLM was explained in line 240 (current line 241-275): “Generalized linear models (GLM) analysis was conducted to explore drivers of the main variables reflecting primary producer abundance (VRS-inferred Chl-*a* and β-carotene)”.

We miss to explain the purpose of using RDA analysis and it has been added (lines 273-274): “Redundancy analysis (RDA) was conducted to identify the explanatory variables of algal assemblage changes”.

- Lines 252 – To follow the FAIR principles of publishing scientific results, we encourage authors to publish their code on any open-source platform.

ANSWER: Raw data (https://github.com/carmen6363/Pigment-article-variables-2026) and statistical information (https://github.com/carmen6363/RDA-pigment) have been published in open- source platforms Github. The GLM analysis codes have not been published because we performed the analysis directly from the R Commander window.

Dating results have been also published (https://github.com/carmen6363/Dating-results-core-SSBG-21).

The analyzed pigment and paleoenvironmental data will be published in the PANGAEA repository. We have previously utilized this platform for datasets related to the Sierra Nevada core analyses. The current dataset has already been submitted and is undergoing the standard review process by PANGAEA; consequently, a DOI has not yet been assigned.

A section on Data availability has been added before Reference section.

Results

To support the readability and clarity of the manuscript's main message, we suggest adding an interpretation in the results section to present the data. In the Results and Discussion section, we keep the line-by-line comments minimal as this section needs restructuring. However, many of the textual comments given above are applicable throughout the manuscript. In a potential second round, we can focus on the Results and Discussion sections in detail.

For example:

“N2 fixing cyanobacteria (Aphanizophyll) are sporadically present from x CE to x CE and increase exponentially after x CE. *This rise coincides with low d15N values, both hinting at NOx transformation and fixation.* “

Another example:

“Echinenone is rather stable throughout the record, meaning that the production of the pigment is constant. *Either its primary producer (cyanobacteria) or its transformation pathway (degradation of other pigments into echinenone) is constantly operating, regardless of climate and environmental forcing.*”

Furthermore, please consider describing the entire dataset (VRS-Chl-a, C/N, dN15, pigments) on a zone-by-zone basis. Alternatively, one can proceed pigment by pigment, as in the examples above, and choose a consistent structure to follow. If the zone-by-zone structure is chosen, a combined interpretation is expected at the end of each zone.

We have completely rewritten the Result section. In the revised manuscript, we provide a zone-by-zone description of the entire dataset, including pigments and other paleoenvironmental and climate variables. As you suggested, we have changed the pigment description to include interpretative sentences.

The subsection 3.2 Trends in pigment composition, 3.3 Instrumental climate data, 3.4 Atmospheric deposition variables and 3.5 Trends in other paleoenvironmental variables have been merged into a single, more cohesive section now titled: "3.2 Trends in pigment composition and other paleoenvironmental variables". This change improves the synthesis of the data and allows for a better connection between different variables.

3.1 : Geochronology:

In a study conducted on a sediment core, the core/sediment picture and description, as well as material characterisation, are expected, yet absent in this manuscript. This may impact several aspects of data interpretation. If possible, we encourage authors to include this information in both the manuscript and the figures, such as the age-depth model.

ANSWER: Please, take into account that in Paleolimnology the core is sectioned at field y no description and analyses as magnetic susceptibility or lithology are usually made. We know the core consists of peaty clays and silty clays in a uniform composition. We have included that in the manuscript (line 295): " The Borreguil Lake record consists of 26 cm of peaty clays and silty clays".

We have published the ^{210}Pb and ^{137}Cs data in the open-access Github repository (see https://github.com/carmen6363/Dating-results-core-SSBG-21).

- Line 259: Please consider displaying both models (CIC and CRS) side by side in the supplementary material. Further, we encourage authors to publish their entire dataset, including the 210Pb and 137Cs data, at any of the open-access data repositories prior to publication, so the data can be linked via DOI to this publication.

ANSWER: ^{210}Pb dates for this core are unequivocal in the two standard (CRS and CIC) models (Appleby et al 1978) give very similar results.

We have published the ^{210}Pb and ^{137}Cs data in the open-access Github repository (see https://github.com/carmen6363/Dating-results-core-SSBG-21).

Figure 2:

We suggest overlaying (A) in (B), or indicating the marker points of 137Cs on the panel B model, as the y-axis is currently not aligned to the same scale. It is advisable to follow the author's guidelines of the journal to fulfil the criteria of readability (e.g., font type and font size). Furthermore, a light grey shading may be used to express the uncertainty intervals. Lastly, as stated in the text, the age-depth model is reliable only until a depth of 6.63cm, but the pigment data are presented all the way to a depth of 14cm.

Thus, the extrapolated ages should be included in the age-depth figure as a dotted line described as "extrapolated".

ANSWER: We agree. Figure 2 has been modified in line with your suggestions. As we repeatedly point out in the text, the age-depth model is only reliable up to a depth of 6.63 cm. We emphasise the most recent period in which the age-depth model is reliable in our results and discussion for this reason.

As mentioned in the caption of the figure 3, “Dates prior to 1850 should be interpreted with caution, as the age provided is beyond the confidence dating provided by stable radioisotopes”. Also, extrapolate dates are marked with a '~' symbol (e.g. ~1800, ~1600).

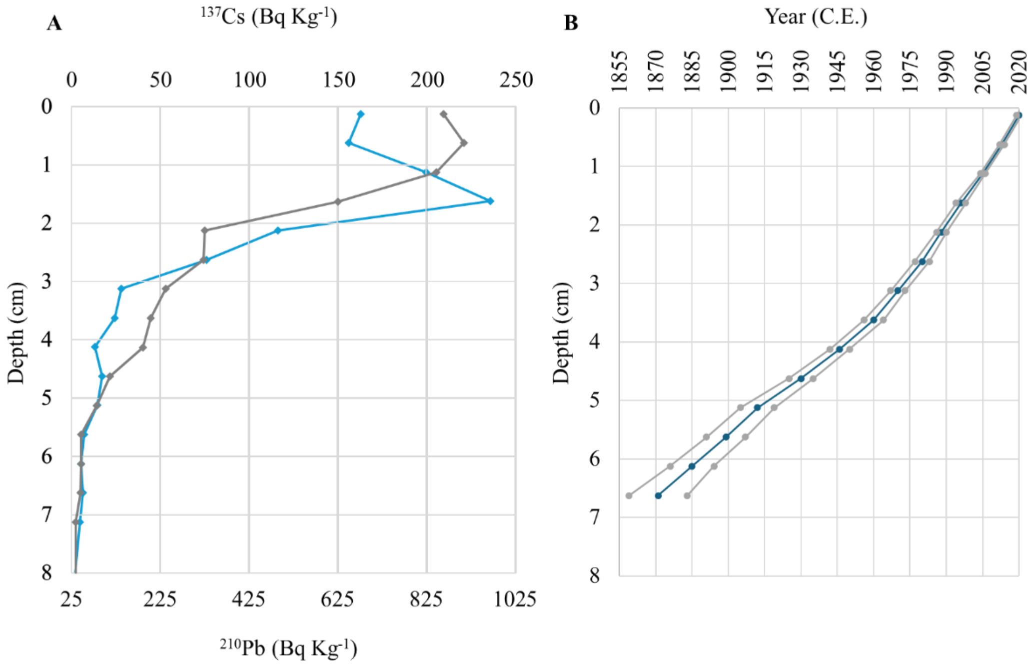


Figure 2: A) Radiometric chronology showing ^{210}Pb (grey line) and ^{137}Cs (blue line) activity (bq kg^{-1} dried sediment) from sediment core SSBG-21 extracted from Borreguil Lake. B) ^{210}Pb-estimated age (using the constant rate of supply model) versus core depth (black line) and the associated errors (grey lines).

3.2 Trends in pigment composition:

For better readability and message conveying, we suggest presenting the pigment data alongside the chronological description of sediments/lithology and all other proxies. This would affect Figure 3.

- Line 310: It is unclear how the PCA of the climatic data periods explained the total pigment variation. What data were used for this PCA? Was it a merged dataset of pigment data and climate data? If so, this was not clearly explained in the methods.

ANSWER: Probably our sentence was confusing. We don´t mean that climate data PCA explained the total pigment variation. In any case, this PCA has been removed to reduce the length of the manuscript and the number of figures following your suggestion.

Figure 3:

It is advisable to use colours to distinguish between the different pigments, guiding the reader more easily through the various primary producers. If authors decide to keep the pigments in relative abundance % (which we don't suggest), a stacked bar plot would be easier to constrain the community shifts. Furthermore, plotting the lithology, TOC, C/N, β-carotene, and VRS-inferred Chl-a data along with the pigments would be useful in supporting certain interpretations. Optionally, PC1 & 2 from pigment PCA can also be added, along with the other data, to complete the dataset for interpretation.

For the extrapolated ages, we recommend removing age markers or keeping only 1600 and 1800 as the uncertainty of these ages is high.

It is advisable to follow the journal's guidelines to fulfil the criteria of readability (e.g., font type and font size), as some x-axes have no tick labels. The CONISS clustering dendrogram can be either reduced in width or displayed simply as boxes that delineate clusters.

ANSWER: Figure 3 has been revised according to your suggestions. Previous Fig 3, fig 4 and fig. 8 have been merged in a single figure.

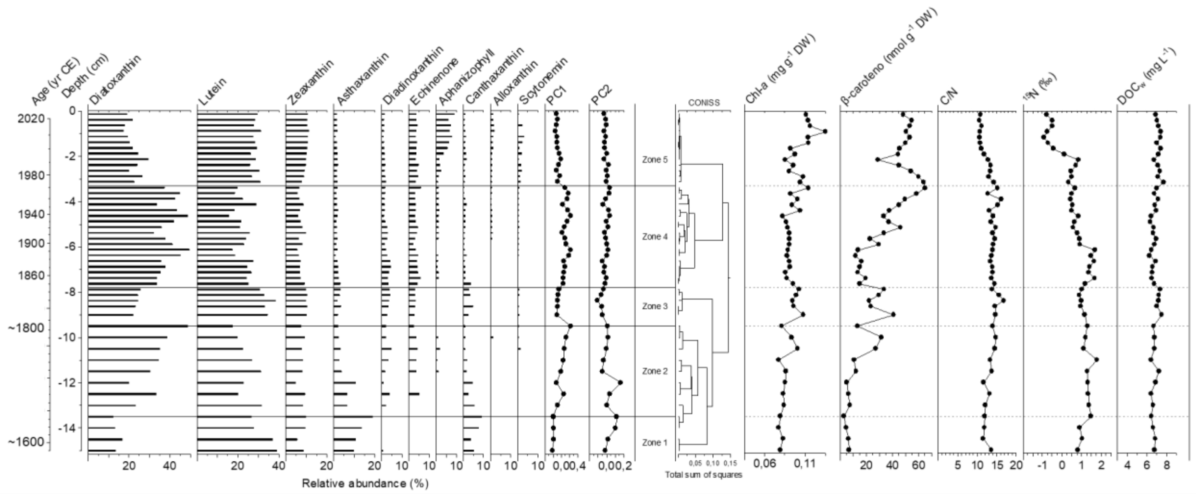


Figure 3: Relative abundance diagrams of the sedimentary pigments and evolution of selected paleoenvironmental variables recorded in the sediment core SSBG-21 in Borreguil Lake. The axis 1 and 2 of the PCA performed on pigment data and the result of a cluster analysis of pigment assemblages data using constrained incremental sum of squares (CONISS) are shown. The black lines represent the main zonation identified by the broken stick model. The data are plotted against the sediment depth (cm, primary y-axis) and on age (yr CE, secondary y-axis). Dates prior to 1850 should be interpreted with caution, as the age provided is beyond the confidence dating provided by stable radioisotopes.

Sections 3.3 and 3.4

We suggest merging sections 3.3 and 3.4 to remove redundancies and clarify the data used for

Figure 7. It is unclear whether these are the binned modelled data or if these data originate from the sedimentary record. Further, in the section 3.4, it is recommendable to provide a full description of the SPI, MATA, or refer to another paper? Optionally, these data can be presented in the context of the zone-by-zone description.

ANSWER: We have merged both sections 3.3 and 3.4 in the zone-by-zone presentation of the results.

It is unclear whether these are the binned modelled data or if these data originate from the sedimentary record. Sorry, we do not understand your question. We specify in the text and section heading all climate data are modelled from instrumental data. Anyway we have added it in the figure caption. We refer in many occasions to Sigró et al (2024) where description of climate data are available.

We refer to Becker at al. (2013) for a full description of the SPI (line 352).

Becker, A., P. Finger, A. Meyer-Christoffer, B. Rudolf, K. Schamm, U. Schneider, and M. Ziese, 2013: A description of the global land-surface precipitation data products of the Global Precipitation Climatology Centre with sample applications including centennial (trend) analysis from 1901-present. Earth Sys. Sci. Data, doi:10.5194/essd-5-71-2013.

Figure 4:

Confusing to have two PCA's. If there is no specific reason why these two time periods were split, please consider removing the second PCA; If removing the second PCA, parts of figures 3, 4 and 8 may be merged in one. Furthermore, please consider using a more intuitive temporal scale, such as whole-round non-diagonal numbers (e.g., 2000, 1950, 1900, 1850, 1800). Similar to other figures, please follow the journal guidelines for figures in terms of fonts, font size, and line width. Additionally, consider using a different line style for PC1 and PC2 to accommodate colour-blind individuals.

ANSWER: Following the reviewer's suggestions to streamline the results, the second PCA has been removed, and the figures have been reorganized throughout the revised manuscript. We have reduced the total number of figures from 10 to 6. Specifically, the information previously contained in Figure 4 has been merged into the current Figure 3 to provide a more integrated view of the data.

Figure 5:

Please refer to the comments above regarding the reduction of PCAs and statistical analysis.

Preferably, an objective summary of the full dataset should be presented in the results section.

To enhance the readability of the PCA, we recommend using arrows/vectors for the variables (pigments). Optionally, a colour-coding of the sites/samples by their respective CONISS pigment cluster would help the reader to understand the data and their interpretation. And finally, if the CONISS clusters are used, a confidence ellipse of these clustered data would guide the reader in the statistical significance of the clusters (clearly seen potential overlaps).

ANSWER: The second PCA has been removed from the manuscript. The new figure 5 (previous fig. 4) is shown.

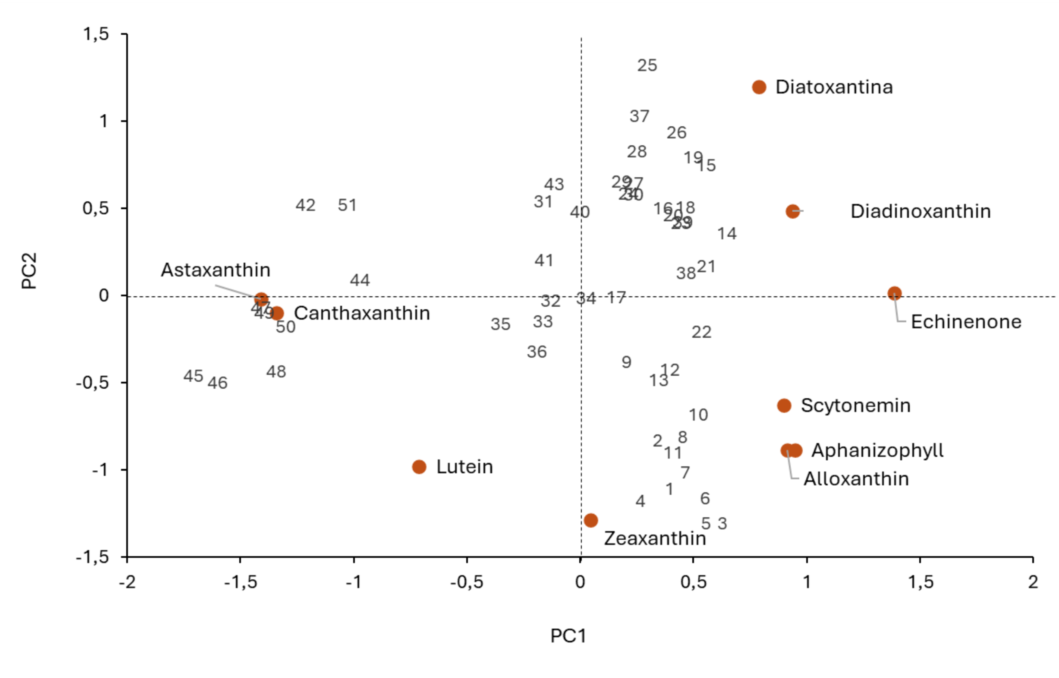


Figure 5: Principal component analysis (PCA) biplots of the pigment assemblages from sediment core SSBG-21 extracted from Borreguil Lake for the last ~430 years. Numbers refer to the sample sites being 1 the most modern sample and 51 the oldest sample.

Figure 6:

For increased readability, we suggest adding transparency of the individual datapoints and using regular x-label scaling (e.g., 1860, 1870; 1880, 1900). Why are wNAO and SPI showing opposite trends? It is probably best to keep only one metric in the paper; this makes the story shorter and more understandable. Please verify that the SPI unit should be cm [area unit]. month^{-1}; as this is data from another publication. Further, we encourage authors to make a general discussion figure, with 3-4 pigments (Diatox, Lut, Echi, Aphanizo), the SPI, MATA, total Chl-a, C/N, N15, timescale boxes marked on it (e.g. LIA, MCA, etc.), and pigment zones marked on it. This would also replace Figures 8 and 9, enhancing the main message delivery to the reader.

ANSWER: Your suggestions have been taken into account when constructing the new figures, and the wNAO has been removed from the manuscript. SPI and wNAO show opposite trends because they measure different things: wNAO shows positive values when the weather is dry and Saharan dust enters the European atmosphere, while SPI shows negative values when the Sahel drought is more severe and there is more dust in the atmosphere. Substantial negative SPI values indicate severe drought conditions, significantly enhancing dust emission and atmospheric loading. Both variables have been considered as drivers of Saharan dust deposition. We have included an explicative sentence in text (lines 361-636): "The SPI record indicated a relatively stable and dry period (negative values) from around 1970 yr CE to the present, with the lowest values observed during the 1980s-1990s yr CE (Fig. 6). Substantial negative SPI values indicate severe drought conditions, significantly enhancing dust emission and atmospheric loading (Jiménez et al., 2018)".

Moreover, we have added an explicative sentence in M&M (line 251): "Negative SPI values indicate severe drought conditions, significantly enhancing dust emission and atmospheric loading".

The rainfall unit used for the SPI is cm per month.

We believe it is important for readers to see the complete pigment record. Therefore, we have merged the pigment data with the PCA results and the biogeochemical variables into a single figure (now Figure 3). This effectively reduces the total number of figures. However, we decided not to include the climate variables in this combined figure to ensure clarity and readability.

The SPI, MATA and APA (current Fig 4, previous Fig. 6) have been reformatted so that the new Fig. 3 and Fig. 4 be easily comparable.

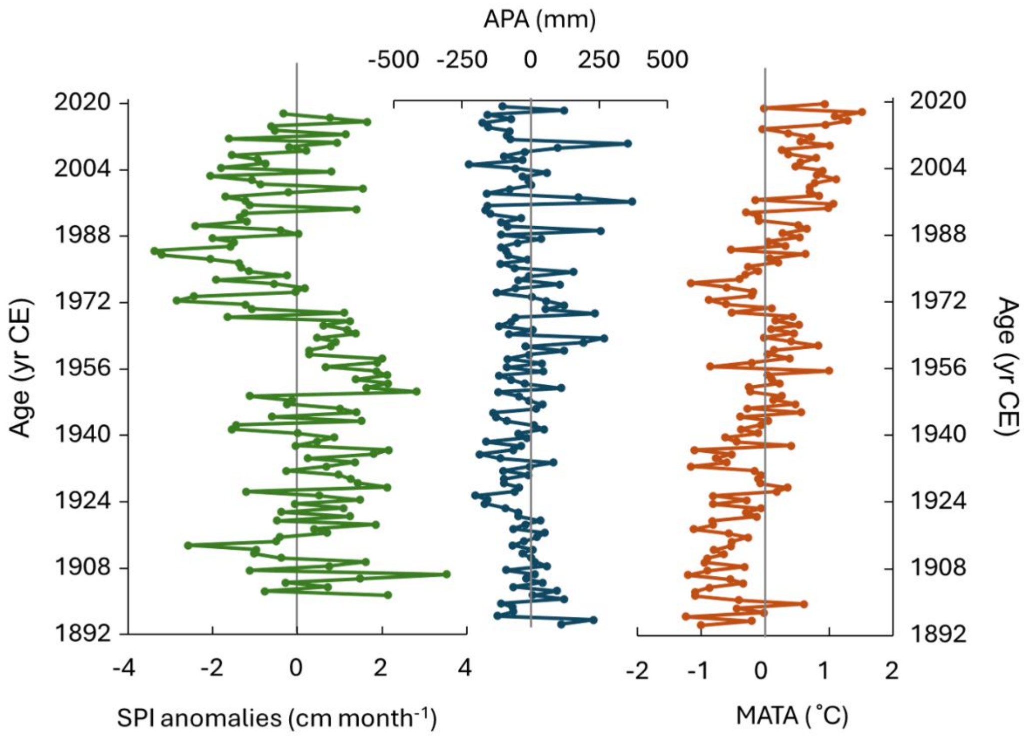


Figure 4: Evolution of the climatic variables MATA (mean annual temperature anomalies for air), and APA (annual precipitation anomalies) at the summits of Sierra Nevada for the period 1894-2020, and Saharan dust metric SPI (Sahel Precipitation Index). All are instrumental data. Temperature and precipitation data were obtained from Sigró et al. (2024). The anomalies of the SPI are calculated with respect to 1900 and 2013, and based on June through October averages for each year.

Figure 7:

Similar to other figures, please follow the journal guidelines for figures in terms of fonts, font size, and line width. Additionally, an interpretation of the PC axes would be helpful to convey the main interpretation of the data. Since this PCA is performed on the climate data, which is not new in this paper, consider moving this figure to the supplementary material or blending it in the overall discussion plot as line plots.

ANSWER: Figure 7 has been removed to shorten the manuscript. The PCA on climate and atmospheric variables has been also removed.

Section 3.5:

This section could be merged with the chronological description of sediments, pigments, and other proxies. We recommend updating Figure 3 according to our suggestions and referring to it in this text, wherever it will be placed in the restructured results.

ANSWER: Figure 3 has been updated and section 3.5 has been moved forward in the text, now following the description of pigments zone by zone.

Figures 8 & 9:

Please, see the comments in Figure 6.

ANSWER: Thank you. Figures have been updated. Please see current Fig. 3.

Section 3.6:

Describing PCA and RDA results can be challenging; Good job! Please, try to limit overlap with the zone-by-zone description.

Figure 10:

This figure requires revision, as all the names of the pigments are overlapping in the middle of the plot and are not readable. The reasoning behind doing an RDA with only two variables is missing. Please explain the purpose of doing the RDA this way either in the methods or in the results. If forward selection doesn't allow us to use more variables, then we have the option to just show the results of the two drivers in a table. It may also be potentially interesting to perform partial RDA and variance partitioning. https://pmassicotte.github.io/stats-denmark-2019/07_rda.html#/ and show results of variance partitioning (in a table or supplementary).

ANSWER: We performed the RDA with all the climate and atmospheric variables (MATA, APA, and SPI). This is stated in Methodology section: Statistical analysis. In this figure we show only the explanatory variables selected by the RDA model. Forward/Backward stepwise model was used to extract the variables that significantly increased the amount of explained variation of pigment data.

We considered other paleoenvironmental variables as chlorophyll or beta-carotene to be not independent of algal assemblage pigment variables and therefore not suitable for inclusion in RDA. For this reason, we assessed the influence of climate and atmospheric variables on algal biomass variables (chlorophyll and beta-carotene) separately using GLM analysis. Furthermore, since reliable dating, climate, and atmospheric data are only available for n = 22 samples, we limited the ordination analysis to a maximum of three variables to avoid over-parameterization and ensure model robustness.

Nevertheless, to ensure the robustness of our findings, we performed an additional RDA using the full set of paleoenvironmental variables: Chl-a, β-carotene, DOCw, C/N, $\delta^{15}N$, MATA, APA and SPI. After applying VIF = 5, the results indicate that MATA, SPI and DOCw are the explanatory variables of the pigment assemblage:

Model: rda(formula = pigment ~ tm_annual + SPI.relative + DOCw, data = Dataset)

	Df	Variance	F	Pr(>F)
tm_annual	1	0.47293	7.9693	0.003 **

```
SPI.relative   1   0.27944  4.7088  0.022 *
DOCw           1   0.21742  3.6638   0.037 *
Residual      18  1.06820
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
> RsquareAdj(Nombre)
$r.squared
[1] 0.5998631
$adj.r.squared
[1] 0.5331736
```

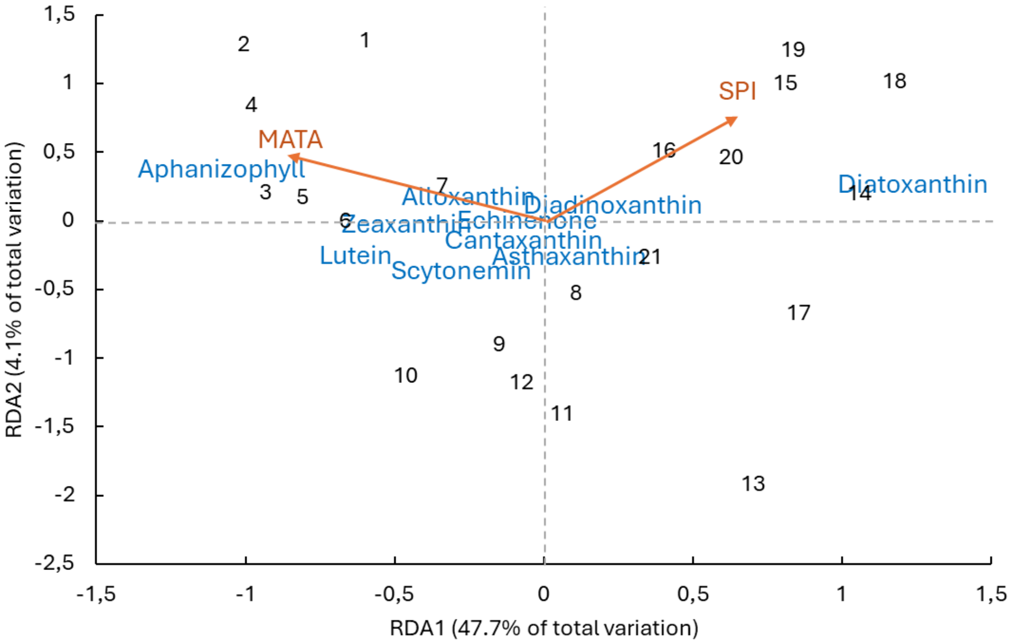


Figure 6: The results of the Redundancy Analysis (RDA) ordination plot showing the relationship between pigment assemblages data from sediment core SSBG-21 and the climate and atmospheric variables. The selected explanatory variables by the RDA analysis are shown: MATA (Mean annual air temperature) and SPI (Sahel Precipitation Index as proxy of Saharan dust input). Only the first two RDA axes are shown. Sample sites (representing sediment intervals) are also shown, being "1" the most modern interval and "22" the oldest one. See Table 1 for more details of the analysis results.

Discussion

In general, the discussion section would benefit from restructuring, grouping and linking processes together to interpret certain phenomena. If the RQ are reduced to 2 as we suggest in the introduction section, the structure of the discussion section must change accordingly. Please consider these changes to improve the clarity and readability of this manuscript. To reduce the number of redundancies, it is preferable to go topic by topic or time zone by time zone; currently, it is a combination of both. The main section headers (4.1 and 4.2) are too long and should be a maximum of 8 words long. Often, a substantial amount of literature is provided, but its relevance and context remain unclear (examples outlined below). It is essential to clearly outline the point one wants to discuss, present the evidence, and highlight the relevant aspects of the literature, ultimately working towards comparison and conclusion. This will enhance the readability of the manuscript

ANSWER: Thank you for your suggestions. The discussion has been shortened from 8 pages to less than 5. We have responded to your comments point by point.

4.1 : increasing biomass?

ANSWER: The subsection 4.1 title has been shortened and changed to Changes in algal biomass.

Paragraph 1:

Lines 419 – 420: Suggestion: "The long-term changes in primary production and algal biomass were assessed using VRS-inferred Chl-a B-car and C/N"

Example of inconsistent paragraph logic structure: This first sentence brings the readers' expectations to the discussion of these biomass and community changes. However, as the paragraph continues, there is no explanation of these changes; instead, it presents a range of values (which belong to the results) and discusses the origin of OM. Thus, we encourage authors to revise the section and paragraph structure in a way that logically groups the information, and the logic is clearly communicated so the reader knows what is connected to what and why.

ANSWER: We agree. We tried to demonstrate that our data were consistent with those previously published for Sierra Nevada lakes and other similar bodies of water. However, we agree that this is not necessary. The paragraph has been removed for the sake of clarity.

The interpretation of the C/N ratio follows the common way, but it is advised to consider N-limitation as an alternative explanation. If N is limited, N-fixating cyanobacteria may still result in high productivity and influence the C/N ratio, rather than the changes in the N source. This is also evident in the d15N data. And if pollen data are available from this lake or a nearby lake, these two potential explanations can be distinguished. As Section 4.2.3.2 discusses the N2-fixing cyanobacteria, it would be advisable to link these interpretations together and restructure the discussion to reduce redundancy and repetition. This would also help to streamline the main message and shorten the manuscript.

Thank you for your suggestion about C/N interpretation. We have included this possible explanation and connected C/N values with N-fixating cyanobacteria. Please see lines 536-539: "If N is limited, N2-fixing cyanobacteria (aphanizophyll) may still result in high productivity and lower the $\delta^{15}N$ data as the isotopic signature of N_2 is also low. The decline of the C/N ratio after 1970 could be partly the result of an enrichment of organic matter in nitrogen due to nitrogen fixation".

Relative abundance data for pollen is available for nearby Lake Rio Seco (Anderson et al., 2011). This record covers the entire Holocene period and shows no significant changes over the past 400 years except for the appearance of cultivated species pollen (Olea…). The authors conclude that the lake was probably above the elevation of the tree line throughout the Holocene.

Anderson, R. S., Jiménez-Moreno, G., Carrión, J. S., & Pérez-Martínez, C. (2011). Postglacial history of alpine vegetation, fire, and climate from Laguna de Río Seco, Sierra Nevada, southern Spain. *Quaternary Science Reviews*, *30*(13-14), 1615-1629.

Paragraph 2:

It is essential to clarify the main message that is to be conveyed in this paragraph. Is it: a). Biomass declines in the LIA? or b). Are crustaceans abundant during the LIA? or c). Chl-a and b-car are similar and good indicators of total productivity. The paragraph should focus on a single topic; otherwise, it's very difficult to follow.

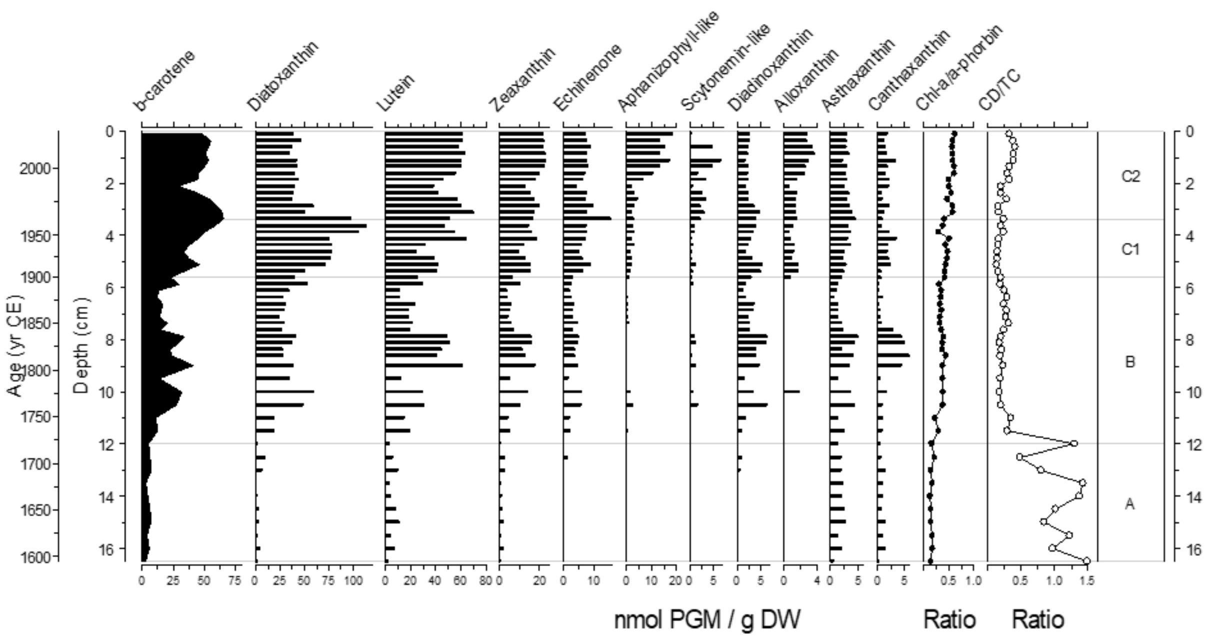
ANSWER: Algal biomass (chlorophyll a and β-carotene) shows very low values from 1600 to 1750 during the Little Ice Age (LIA), but peaks between 1750 and 1800 (still during the LIA). This peak coincides with a period of minimum temperatures according to Jiménez-Moreno et al (2023). We do not have a clear explanation for this biomass peak. Therefore, we hypothesize that it could be due to low grazing pressure. However, several authors have highlighted the 1750–1800 period as an era of climatic extremes in the Iberian Peninsula. We hypothesize that these extreme conditions may explain the fluctuations observed in chlorophyll-a and β-carotene concentrations during this interval

We have addressed these issues in lines 447-458: "The long-term changes in primary production and/or algal biomass throughout the core was assessed by the combined analysis of VRS-inferred chl-a and β-carotene, indicators of algal biomass. Both variables show similar trends throughout the core. From the bottom of the core to around 1750 yr CE, both remained stable and low, reaching their lowest levels. This low biomass period may be linked to low temperatures during the Little Ice Age (LIA), as shown by Jiménez-Moreno et al. (2023) for Río Seco Lake in Sierra Nevada and from Oliva et al (2018) for the Iberian Mountains. Between ~1750 and 1800 CE, concentrations of chl-a and β-carotene fluctuated, reaching peak levels, despite this period coinciding with the lowest temperatures of the Little Ice Age (LIA) in the Sierra Nevada, according to Jiménez-Moreno et al. (2023). Although we do not have a clear explanation for this increase, we hypothesize that this phenomenon may be attributed to reduced herbivory pressure (reduction of astaxanthin and canthaxanthin), as the development of herbivore populations is constrained by the brief duration of ice-free periods. However, Oliva et al. (2018) indicate that the period 1760–1800 CE was a time of climate extremes in the Iberian Mountains, which could explain the fluctuations observed in chl-a and β-carotene in the Sierra Nevada during that period".

Line 433: Could this peak in the pigments be related to better preservation during the cold period? Until here, there is no explanation in the manuscript of how the preservation of the pigments was somehow constrained. As the lake is only 2.8m deep and the light penetrates all the way to the bottom, the preservation of pigments may be a very important factor when interpreting. Maybe the relative changes reflect only the changes in the preservation conditions? If any method or data analysis was used to constrain the preservation, it should be stated in the methods section.

ANSWER: Although we do not have a clear explanation for this increase, we hypothesize that this phenomenon may be attributed to reduced herbivory pressure, as the development of herbivore populations is constrained by the brief duration of ice-free periods.

The hypothesis that pigment preservation is strictly climate-dependent is inconsistent with our observations. We calculated the Chl-a/a-phorbins ratio throughout the entire profile (Figure S2, Supplementary data). The smooth, continuous variation in this ratio, with no abrupt shifts, indicates that there is no preservation peak associated with the LIA. In addition, the highest Chl-a concentrations have been recorded since the 1960s, a period characterized by the highest temperatures and the most prolonged ice-free intervals in the lake's recent history.



Supplementary Figure S2. Pigment profiles of the 10 selected marker pigments and b-carotene expressed as concentrations (nmol PGM / g DW) and the ratios of Chl-a/a-phorbin as a Chla preservation index and the ratio CD/TC (chlorophyll-a derivatives vs. total carotenoids) as indicative of higher contribution of autochthonous production when the ratio is low. Four main biostratigraphic zones were established based on constrained incremental sum of squares partitioning of pigment concentration with Rioja-package version (0.9-15.2) (Juggins, 2017) using R version 3.5.3 (R Core Team 2019).

Paragraph 3:

This paragraph covers what is after the LIA, but it offers just a description of trends, which is rather results than discussion. There is a lack of context regarding what this means in relation to the RQs. E.g., if biomass follows temperature, then temperature is a driver. When describing a time period, please offer an interpretation. An example could be: *"During the XY timeframe, P was abundant, and xx algae thrived. This appears to be the case across the Sierra Nevada, vouching for a regional driver. Fe/Zr data in study Y shows that the increasing biomass follows dust, here we also find a strong correlation with dust during this time window (table X). We therefore suggest that dust drives the production of biomass."*

Paragraph has been changed. See lines 459-462: "At the end of the LIA (1800-1840 yr CE), a temperature peak at the Sierra Nevada summit was recorded (García-Alix et al.,2020; Jiménez-Moreno et al., 2023) which probably resulted in a major algal biomass increase, consistent with trends in other alpine lakes (Hu et al., 2014; Huo et al., 2022; Lami et al., 2010; Oleksy et al., 2020). Moreover, the C/N peak ~1840 yr CE, shortly after the LIA, suggests an influx of external organic matter, potentially driven by thaw-induced transport."

Take into account that, before 1905, no data on Saharan deposition was available.

Paragraph 4:

Paragraph should not be composed of 2 sentences only. Please find an alternative location for this information so it's well-placed in context.

ANSWER: This paragraph has been joined with the next one.

Paragraph 5:

This paragraph comprises literature only with no clear structure which would link it to the data. Consider following an order from far away to closer by, or the opposite. Further, this type of information is rather expected in the introduction.

ANSWER: The paragraph has been shortened and focused. It has been merged with other paragraph of the discussion. Please see lines 462-474: "The main changes in VRS-inferred chl-a and β-carotene occurred from ~1920 yr CE onwards, when a striking increase in both variables was observed. This biomass increase has no precedent in the last 400 years in the lake and according to GLM analysis (Table 2), climate factors—particularly rising temperatures—were identified as the main drivers of these changes. In alpine lakes, higher temperatures and lower precipitation may extend the ice-free period (Anderson et al., 1996; Rogora et al., 2018), increasing the growing season and resulting in a higher accumulation of annual algal biomass. Moreover, longer ice-free seasons in alpine lakes boost light availability, water temperature, and solute inputs through increased snowmelt and weathering (Preston et al., 2016; Sommaruga-Wögrath et al., 1997) all enhancing biological production (Douglas and Smol, 2010; López-Merino et al., 2011). These facts were observed across the Sierra Nevada lakes (Jiménez et al., 2018) and across arctic and alpine lakes (Adrian et al., 2009; Rühland et al., 2008, 2015). We therefore suggest that temperature controls how much biomass is produced in the lake. This means that the big rise in temperature and fall in rain in summer in Sierra Nevada (Sigro et al., 2024) could be the reason for the large and unprecedented increase in the amount of algae since the 1960s yr CE to the present".

Paragraph 6:

Similar to other paragraphs, there is a lack of flow, and it is unclear whether the paragraph addresses the biomass or communities.

Line 457: It would be useful to have the C/N values plotted along with the pigments and the PCs, not only the algal mass. This can be resolved by reorganizing the content of the figures as suggested throughout the review.

ANSWER: Following your suggestion, the figures have been revised and updated to enhance clarity and presentation.

Line 460 – Right, now the question is: What does this study add to what is already known?

ANSWER: This study indicates that the post-1970 increase in algal biomass have no precedent in the last 400 years and is due to the warming in the Anthropocene (please, see lines 471-474): "We therefore suggest that temperature controls how much biomass is produced in the lake. This means that the big rise in temperature and fall in rain in summer in Sierra Nevada (Sigro et al., 2024) could be the reason for the large and unprecedented increase in the amount of algae since the 1960s yr CE to the present."

See also lines 464-465: "This biomass increase has no precedent in the last 400 years in the lake and according to GLM analysis (Table 2), climate factors—particularly rising temperatures—were identified as the main drivers of these changes".

Line 464 – This part is somehow repetitive and claims that everything is important; if all of it remains vague, what is the conclusion?

ANSWER: The sentence has been removed and the paragraph considerable shortened. Please, see lines 462-474: "The main changes in VRS-inferred chl-*a* and β-carotene occurred from ~1920 yr CE onwards, when a striking increase in both variables was observed. This biomass increase has no precedent in the last 400 years in the lake and according to GLM analysis (Table 2), climate factors—particularly rising temperatures—were identified as the main drivers of these changes. In alpine lakes, higher temperatures and lower precipitation may extend the ice-free period (Anderson et al., 1996; Rogora et al., 2018), increasing the growing season and resulting in a higher accumulation of annual algal biomass. Moreover, longer ice-free seasons in alpine lakes boost light availability, water temperature, and solute inputs through increased snowmelt and weathering (Preston et al., 2016; Sommaruga-Wögrath et al., 1997) all enhancing biological production (Douglas and Smol, 2010; López-Merino et al., 2011). These facts were observed across the Sierra Nevada lakes (Jiménez et al., 2018) and across arctic and alpine lakes (Adrian et al., 2009; Rühland et al., 2008, 2015). We therefore suggest that temperature controls how much biomass is produced in the lake. This means that the big rise in temperature and fall in rain in summer in Sierra Nevada (Sigro et al., 2024) could be the reason for the large and unprecedented increase in the amount of algae since the 1960s yr CE to the present".

Paragraph 7:

Line 467 – repetitive information

ANSWER: This paragraph has been removed. New paragraph has been written to explain Saharan P fertilization. Lines 475-485: "Additionally, Saharan dust may increase nutrients in Borreguil Lake, boosting algal production. Sierra Nevada has experienced an increase in the frequency of dust events particularly since the 1970s yr CE. Longer ice-free periods may have further amplified dust exposure, boosting nutrient availability and promoting algal growth (Korbee et al., 2012; Saros et al., 2005; Yang et al., 1996). Saharan dust inputs have been shown to influence the structure and composition of biological communities in Sierra Nevada lakes. For example, phosphorus inputs enhance algal biomass, leading to higher chlorophyll-*a* concentrations (Carrillo et al., 1990; Morales-Baquero et al., 2006). Similar Saharan dust fertilization effects have been observed in Pyrenean (Camarero and Catalán, 2012). Recent results from Borreguil Lake show that, in 2022 yr CE, following a massive influx of Saharan dust in Sierra Nevada, there was a significant increase in phytoplankton water column density and a shift in the lake algal composition, with a marked increase in the percentage of cyanobacteria (Supplementary Fig. S1. Thus, the increase in algal biomass in Borreguil Lake since the 1970s can be attributed primarily to temperature and secondarily to Saharan P input"

Line – 469-471: "Longer ice-free … promoting algal growth" - not necessarily, considering that algea bloom in spring, nutrients need to be available in spring. If these nutrients are lying on the ice, and are released at once during spring melt, it only makes algae thrive more. If the dust is not accumulated on the ice, it might sink to the bottom during winter without being used, e.g. a larger part of the load goes to burial.

ANSWER: We agree that large part of the dust can go burial. However, major Saharan dust deposition occurs during spring and summer time in the western Mediterranean area (see in Morales-Baquero et al 2006a; Morales-Baquero & Pérez-Martínez 2016). Therefore, during the ice-free period, the lakes receive dust inputs that the algae can use to enhance their growth. This has already been proven for Sierra Nevada lakes by researchers from our research group. For example, in a limnological study conducted over two years, Morales-Baquero et al. (2006a) demonstrated that Saharan phosphorus (P) inputs enhance chlorophyll-a in Sierra Nevada lakes. As we specify in former lines 473–474, “phosphorus inputs enhance algal biomass, leading to higher concentrations of chlorophyll-a (Carrillo et al., 1990; Jiménez et al., 2018; Morales-Baquero et al., 2006; Pulido-Villena et al., 2006)”.

Algae do not bloom in Sierra Nevada lakes in spring because they are generally covered in ice at that time. Furthermore, the dynamics of algal biomass in these lakes are not clearly patterned and fluctuate. See the figure below, obtained from Morales-Baquero et al. (2006b), showing the Chl-a levels in two Sierra Nevada lakes (La Caldera and Río Seco) over three years.

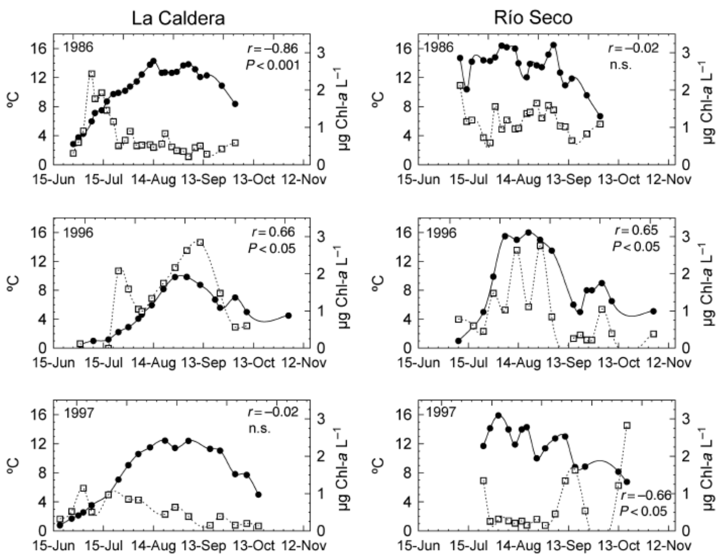


Fig. 2 Time course of temperature (solid line) and chlorophyll *a* (broken line) in La Caldera and Río Seco lakes during the 3 years studied (*r* = Pearson correlation between variables).

Morales-Baquero R., Pulido-Villena E. & Reche I. (2006a) Atmospheric inputs of phosphorus and nitrogen to the Southwest Mediterranean region: biogeochemical response of high mountain lakes. Limnology and Oceanography, 51, 830–837.

Morales-Baquero, R., Carrillo, P., Barea-Arco, J., Pérez-Martínez, C., and Villar-Argaiz, M.(2006b) Climate-driven changes on phytoplankton–zooplankton coupling and nutrient availability in high mountain lakes of Southern Europe, Freshw. Biol., 51, 989–998, https://doi.org/10.1111/j.1365-2427.2006.01545.x, 2006.

Morales-Baquero, R. and Pérez-Martínez, C. (2016) Saharan versus local influence on

atmospheric aerosol deposition in the southern Iberian Peninsula: Significance for N and P inputs, Global Biogeochem. Cycles, 30, 501–513, https://doi.org/https://doi.org/10.1002/2015GB005254.

Line 475 – Please be selective in referencing: e.g., referring to the "United States lakes" – the USA is a big country, and to place this interpretation into a comparable context, it is advisable to specify why and where it is relevant.
ANSWER: We agree, references to Brahney have been removed.

Line 478: It is not clear where the data in the supplementary figure S3 originates from. Are these monitoring data? If so, these were not introduced in the sampling section. If they are from another publication, it would still be useful to clarify that those are monitoring data of the water column, not from the sediment.

ANSWER: These data are monitoring data of the water column. We specify it in the M&M section (please, see lines 142-143): "To determine the composition of the phytoplankton, several water column samples were taken at the deepest zone in Borreguil Lake". It is also specified in the figure caption and in the discussion text (lines 481-483): "Recent results from Borreguil Lake show that, in 2022 yr CE, following a massive influx of Saharan dust in Sierra Nevada, there was a significant increase in phytoplankton water column density and a shift in the lake algal composition, with a marked increase in the percentage of cyanobacteria (Supplementary Fig. S1)".

Paragraph 8:

The start of the paragraph is good, but what is missing is the own data part. This should be added to this paragraph in a logical argument.
ANSWER: The paragraph has been changed. Please, see lines 478-479: "It is conceivable that nitrogen deposition has influenced the increase in algal biomass observed in Borreguil Lake and the sharp decline in the C/N ratio post-1970 CE could reflect this N enrichment in the lake".

4.2 Community changes?

ANSWER: The subsection 4.2 title has been shortened and changed to Changes in the composition of the algal community
Line 490-493: The authors introduce here 4 classes of planktonic community, and it is very

unclear how these classes were defined, and where these data originate from. Are these from monitoring data as questioned just above? Similarly, for the diatoms, where does the information about periphytic species come from?

ANSWER: This paragraph has been moved to the 'Study site' section as a characterization of the study lake (lines 145-148). There, we explain where these data originate. Furthermore, a reference to the study in which diatoms from Borreguil Lake were analysed has been included. Please see the new paragraph from line 145 to line 154: “The planktonic algal community of Borreguil Lake currently consists of four classes, dominated by Chlorophyceae and Cyanobacteria, followed by Chrysophyceae and Cryptophyceae (Supplementary Fig. S1). Chlorophyceae is the most diverse group, with 17 taxa. Most of the diatoms are periphytic species (Llodrà-Llabrés et al., 2024).

In the lakes of the Sierra Nevada, the planktonic community coexists with three additional algal communities (Cuesta Linares et al., 2003; Sánchez Castillo and Morales Torres, 1980): (1) a bog-associated algal community comprising Chlorophyceae (particularly Zygnemataceae and Desmidiaceae), along with diatoms and cyanobacteria, some of which are nitrogen-fixing (such as *Nostoc*, *Anabaena*, and *Cylindrospermum*); (2) epilithic biofilms primarily made up of cyanobacteria and diatoms and epipelic diatoms; and (3) algal mats predominantly composed of filamentous Zygnemataceae and cyanobacteria growing at the littoral and/or bottom of the lake”**.**

Line 493: Why is Ca important? Why not Fe or S? One can argue with any element; thus, it must be justified why Ca is important, if this helps your argument.

ANSWER: You are right. These two paragraphs have been removed to reduce the length of the manuscript and focus on our paleolimnological results.

Line 500: This may be true, but both diatoms and chrysophytes are observable in the sediment through their siliceous tests. One can then check these under the microscope to confirm the relative changes in their abundances.

ANSWER: Unfortunately, the data we have on diatom and chrysophyte siliceous remains only shows relative abundance, not absolute abundance. Therefore, we cannot make assumptions about changes in their absolute abundance.

Line 508 – These observed algal groups are important, perhaps best mentioned in the methods, and which pigments trace which group in your case.

ANSWER: Zygnemataceae and desmidiaceae currently belong to class Zygnematophyceae. It has been included in several places in the manuscript (see lines 151, 153, 195 in M&M; lines 317 in results; lines 569 and 574 in Discussion).

513-517 - This should have been clear from the methods, and it does not require any additional discussion.

ANSWER: The paragraph has been eliminated

536-537 – If not conclusive, then exclude from the manuscript. This makes it difficult to read.

ANSWER: The paragraph has been eliminated

557-559 – There is a repetitive occurrence of the same conclusion; in general, the whole community seems to be driven by climate change and SPI. In the discussion, one may go beyond this and discuss certain (selective) important exceptions that bring the interpretations closer to the conclusion statements.

ANSWER: You are right. This information were repetitive. Paragraph has been considerably shortened.

564 – This is a rather general statement with little conclusive power.

We agree, the paragraph has been removed

574-583 – We recommend shortening this literature review to the essential.

ANSWER: The paragraph has been considerably shortened as well as the cited references. Please, see lines 523-533.

Section 4.2.3.3 about UV perception comes way too late. Many aspects of the interpretation can be explained by this phenomenon, but the reader must wait until the very end. Already, the fact that there are more carotenoids than phorbins would hint that the UV is an important driver. Consider restructuring the discussion so that the processes and drivers are more closely linked to the data.

ANSWER: The presence of higher amounts of carotenoids relative to phorbins does not necessarily imply that UV radiation was a dominant driver in this sediment record. Although UV-screening pigments such as scytonemin can indeed increase under high UV exposure, in our case the occurrence of scytonemin is more consistent with the ecology and habitat of the producing cyanobacteria than with changes in UV regime.

Scytonemin-producing cyanobacteria in this lake predominantly grow as epilithic or benthic communities attached to stones or shallow substrates. These habitats receive sufficient light and UV simply because they are located within the littoral or shallow benthic zone, and the organisms cannot migrate vertically to avoid exposure. Consequently, they synthesize scytonemin as a constitutive protective mechanism, independent of broader changes in UV flux to the lake surface. The input of scytonemin to the sediment therefore reflects the contribution of these benthic communities rather than a lake-wide increase in UV radiation.

Furthermore, as shown in previous work (Buchaca & Catalan, 2008), several diagenetic and environmental factors can lower the phorbin-to-carotenoid ratios during sediment deposition and early diagenesis (e.g., differential degradation, oxygen availability, sedimentation rate, productivity, nutrient ratios; see also Swain 1985). These processes can explain the decrease in phorbins relative to carotenoids without invoking a UV-driven mechanism.

Taken together, the observed ratio in this core is more parsimoniously explained by (i) the contribution of benthic cyanobacteria that naturally produce scytonemin due to their fixed, high-light habitats, and (ii) known diagenetic patterns that differentially affect phorbins and carotenoids. Therefore, UV radiation itself is unlikely to have been the main driver of the ratio change in this sedimentary record.

Conclusions

Lines 668-671 "During this recent … and geochemical response" may be reduced to "During this recent interval, algal biomass reached levels unprecedented in the four-century record, pointing to the intensified influence of climate warming and dust on Sierra Nevada Lake environments."

ANSWER: Thank you. The sentence has been changed

Line 671-672 No proxy is mentioned for ice-free periods, Saharan dust deposition in the lake (only from model inferred), evapoconcentration, hydrological input (no XRF), and lake levels. To the best of the readers knowledge, no data has been presented that is deterministic with respect to any of these parameters.

ANSWER: The sentence has been eliminated.

688 "forecasting future projections" – The Authors did not discuss or present how it contributes to future forecasting; this is a somewhat pretentious statement. Just stick to the facts, they are cool enough. There is no need to oversell this research; it is interesting as it is. Quite the opposite, the smaller and more detailed the better. There is none of the Nitrogen and UV stories here, for example.

ANSWER: The last two sentences has been eliminated.

The nitrogen and UV subjects are mentioned in the above paragraph (lines 588-593): "Two major ecological shifts, occurring around ~1840 CE and ~1970 CE, appear to reflect periods of intensified environmental forcing and marked ecosystem responses. The first coincided with the end of the Little Ice Age, signalling an early warming phase and an associated rise in algal productivity. The second, more pronounced shift, reflects the compounded impact of warmer and drier climate conditions and intensified dust-driven nutrient enrichment. This led to a fundamental restructuring of the algal community, with previously unseen abundances of N_2-fixing cyanobacteria, cryptophytes, and UV-resistant taxa, while diatoms declined".

In the discussion, we explain that nitrogen fixation and UV-protected algae are consequences of atmospheric input and warming.

The data availability section is completely missing at the end of the manuscript. As already mentioned, several times throughout the review, we highly encourage authors to publish their entire dataset at any of the open-access data storage platforms, as well as their statistical and data analysis code on GitHub or any other repository.

ANSWER: We have included a data availability section.

Raw data (https://github.com/carmen6363/Pigment-article-variables-2026) and statistical information (https://github.com/carmen6363/RDA-pigment) have been published in open-source platforms Github. The GLM analysis codes have not been published because we performed the analysis directly from the R Commander window.

Dating results have been also published (https://github.com/carmen6363/Dating-results-core-SSBG-21).

The analyzed pigment and paleoenvironmental data will be published in the PANGAEA repository. We have previously utilized this platform for datasets related to the Sierra Nevada core analyses. The current dataset has already been submitted and is undergoing the standard review process by PANGAEA; consequently, a DOI has not yet been assigned.

REVIEWER 2

Llodra-llabres et al conducted a paleoenvironmental study of an alpine lake in the Sierra Nevada Mountains of Spain. They focused on using algal pigment concentrations and pigment composition to evaluate changes in the algal community in through time, attempting to link changes to climatic changes and atmospheric deposition. I think this effort has merits and could become an excellent paper, but as it stands needs major revisions.

ANSWER: Dear Reviewer2, thank you for your comments. We have carefully addressed the comments and criticisms provided. We believe these revisions have significantly strengthened the manuscript. The primary changes are as follows:

- Introduction: Revised and updated in accordance with the suggestions of Reviewer 1 and 2. Objectives have been reduced.
- Materials and Methods: Expanded the "Study Site" and "Methodology" sections with new data. We have also included a new table in the Supplementary Materials.
- Results: Entirely restructured and rewritten following Reviewer 1 and 2 recommendations to describe findings zone by zone. We have added a new table and a new figure to the Supplementary Materials, as requested by the reviewer 1.
- Discussion: Streamlined and more focused. The section has been reduced from eight pages to less than five.
- Figures: The total number of figures in the main manuscript has been reduced from ten to six to improve conciseness. Figures have been amended according to the reviewer's suggestion.
- Data in repositories- Raw data and statistical data have been placed in open- source platform Github.

General comments

- In general, the manuscript could use a stronger narrative, more cohesive arguments and clearer focus. At present, the study reads like an exploratory or "fishing" exercise: many variables are measured, compared broadly, and interpreted post hoc. This approach makes it difficult to build a clear, mechanistic story and does not lend itself to a cohesive narrative.

ANSWER:The manuscript has been changed substantially following your and reviewer 1 suggestions. Figures have been updated and discussion have been shortened and focused to our results. Please read the new version of our manuscript.

All of the key elements are present and the dataset is interesting, but the analyses need to be more directed. Specifically, hypotheses should be clearly articulated and explicitly linked to the data and figures. For example, if the authors propose that nitrogen deposition is not important but dust inputs are, they should present proxies for both alongside the relevant pigments in the same figure and directly evaluate those relationships. The likely mechanisms should also be clearly described. Paleo data inevitably limit causal inference, but plausible mechanistic links supported by statistics can still be developed.

- The manuscript also includes an excessive number of figures that do not highlight the key take-home results. The main text should be limited to ~4–5 figures, each telling a clear and focused story aligned with the primary conclusions. Currently, many plots present time on both axes or show variables independently, making it difficult or impossible to visually evaluate the relationships being claimed.

ANSWER: Figures have been reduced from ten to six figures. They have been changed following your and reviewer 1 comments. These updates facilitate a more intuitive visual evaluation of the relationships described in the text, ensuring better alignment between our graphical representations and the discussion of the results

For example, former Fig 3, fig 4 and fig 8 have been merged in a single figure.

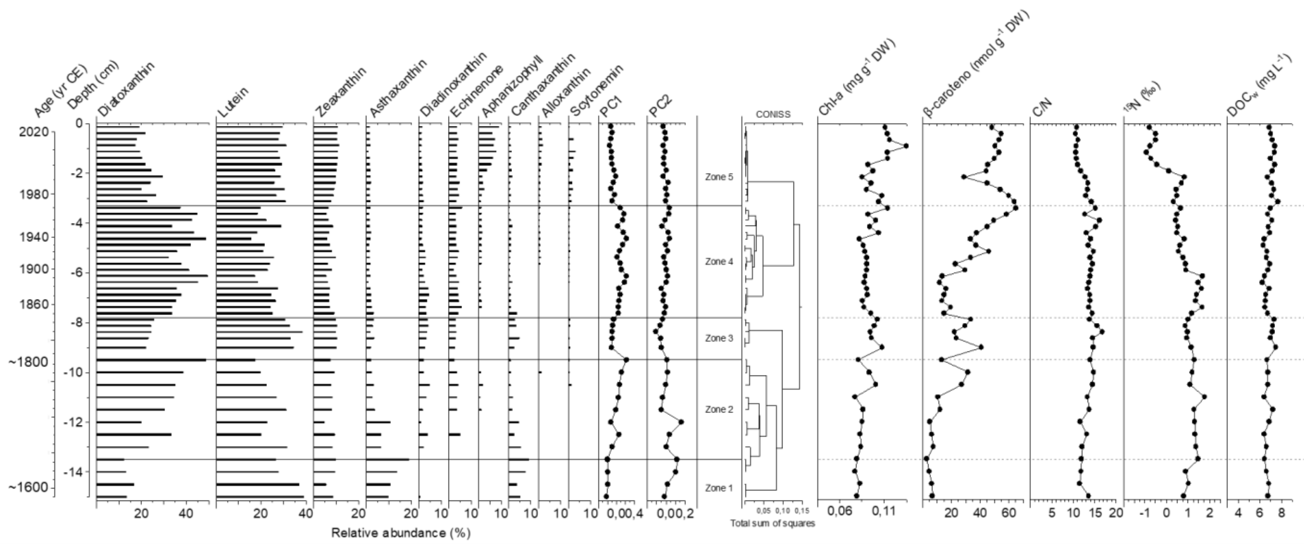


Figure 3: Relative abundance diagrams of the sedimentary pigments and evolution of selected paleoenvironmental variables recorded in the sediment core SSBG-21 in Borreguil Lake. The axis 1 and 2 of the PCA performed on pigment data and the result of a cluster analysis of pigment assemblage data using constrained incremental sum of squares (CONISS) are shown. The black lines represent the main zonation identified by the broken stick model. The data are plotted against the sediment depth (cm, primary y-axis) and on age (yr CE, secondary y-axis). Dates prior to 1850 should be interpreted with caution, as the age provided is beyond the confidence dating provided by stable radioisotopes.

Greater care is needed when drawing conclusions from correlations. For example, showing a statistical relationship between pigment PCA results and a climate metric does not demonstrate that dust is driving pigment changes. The manuscript does not clearly articulate the mechanisms behind these relationships, instead relying heavily on literature examples. While prior work provides context, it cannot substitute for evidence within this dataset. Similarly, C/N ratios are not exclusive indicators of algal versus terrestrial material. Interpreting them this way, particularly in a lake above treeline, is questionable. These types of interpretations suggest insufficient consideration of what each proxy truly represents, its limitations, and the mechanisms that could explain observed changes.

We agree that PCA does not demonstrate that dust is responsible for pigment changes. This is not stated in the manuscript. On the contrary, we state that PCA analysis is used "to summarize the major patterns of variability in pigment assemblages into a few axes" (lines XX)

To demonstrate that dust is driving pigment changes we use RDA analysis "Redundancy analysis (RDA) was conducted to identify the climate and atmospheric explanatory variables of algal assemblage changes" (line 273 of the revised manuscript)

We have shortened the discussion, particularly the literature examples, from eight pages to five pages, and explained the mechanisms behind the observed relationships between pigment and explanatory variables.

C/N is now discussed in relation to possible N limitation of the primary production (please, see lines 535-539): "Saharan phosphorus input into the lake may have caused a stoichiometric imbalance, leading to N-limitation of the primary production that favoured potentially N2-fixing cyanobacteria (aphanizophyll). If N is limited, N2-fixing cyanobacteria (aphanizophyll) may still result in high productivity and lower the $\delta^{15}N$ data as the isotopic signature of N_2 is also low. The decline of the C/N ratio after 1970 could be partly the result of an enrichment of organic matter in nitrogen due to nitrogen fixation".

However, the interpretation of organic material origin is also being maintained.There are numerous studies in which the authors interpret the C/N ratio in terms of the origin of the organic material (terrestrial versus algal) in alpine lakes around the world. For example, Hu et al. (2014) in two alpine lakes in Tibet, Thevenon et al. (2012) in and alpine lake in Switzerland, Jiménez-Espejo at al. (2014) in a Sierra Nevada Lake, Wolfe et al. (2003) in Rocky Mountain lakes or Lami et al. (2000) in eight European alpine lakes.

Both interpretations could be correct. The lake is above the tree line, but it has an extensive catchment area compared to its surface area (lake area = 0.187 ha; catchment area = 50.9 ha; catchment:lake ratio = 272). For example, Hu et al. (2014) state that C/N ratio values in an alpine lake depend on terrestrial organic matter inputs (again reflecting the greater catchment-to-lake ratio). The C/N ratio of this lake is 39, which is much lower than that of Borreguil Lake. Furthermore, Borreguil Lake is also above the tree line, but it is surrounded by alpine meadows (0.563 ha), which can contribute with organic material to the lake.

Of course, the two explanations presented above are not mutually exclusive, and this has been reflected in our manuscript.

- Hu, Z., Anderson, N. J., Yang, X., & McGowan, S. (2014). Catchment-mediated atmospheric nitrogen deposition drives ecological change in two alpine lakes in SE Tibet. Global Change Biology, 20(5), 1614-1628.

- Thevenon, F., Adatte, T., Spangenberg, J. E., & Anselmetti, F. S. (2012). Elemental (C/N ratios) and isotopic (δ15Norg, δ13Corg) compositions of sedimentary organic matter from a high-altitude mountain lake (Meidsee, 2661 m asl, Switzerland): Implications for Lateglacial and Holocene Alpine landscape evolution. *The Holocene*, *22*(10), 1135-1142.

- Jiménez-Espejo, F. J., García-Alix, A., Jiménez-Moreno, G., Rodrigo-Gámiz, M., Anderson, R. S., Rodríguez-Tovar, F. J., ... & Pardo-Igúzquiza, E. (2014). Saharan aeolian input and effective humidity variations over western Europe during the Holocene from a high altitude record. Chemical Geology, 374, 1-12.

- Lami, A., Guilizzoni, P., & Marchetto, A. (2000). High resolution analysis of fossil pigments, carbon, nitrogen and sulphur in the sediment of eight European Alpine lakes: the MOLAR project. Journal of Limnology, 59: 15-28.

- Wolfe, A. P., Van Gorp, A. C., & Baron, J. S. (2003). Recent ecological and biogeochemical changes in alpine lakes of Rocky Mountain National Park (Colorado, USA): a response to anthropogenic nitrogen deposition. Geobiology, 1(2), 153-168.

- The discussion section is also unfocused, reads largely as a literature review, and is overly long. It should instead concentrate on interpreting the results of this study specifically.

ANSWER: The discussion has been shortened and focused. The previous discussion extended to eight pages, whereas the new discussion extends less than five pages. We have reduced the references to previous work and examples of literature and explained the mechanisms behind the variable relationships.

- In addition to the issues with the narrative and presentation, I think more explicit data could be used to link the algal proxies to deposition. If you are not going to use proxies of either yin your own sediment record (which would be best). You could at least temporally compare them to your data; there are well constrained records of both dust and N deposition in this region, why not explicitly use those in your statistical evaluations and data presentation.

ANSWER: Unfortunately, we could not find any records of nitrogen deposition for such a long period of time. Holtgrieve at al. (2011) develop a signal of anthropogenic Nitrogen deposition to Northern America continent. Anyway, these data are not available. In Spain, there are records of nitrogen deposition in the Pyrenees dating back to the 1990s (Camarero & Catalan, 2012). In the Sierra Nevada, our research group measured total nitrogen deposition in 2001–2002 (Morales-Baquero et al., 2006).

We found extensive data on nitrogen emissions in Europe, but none on nitrogen deposition. In this regard, Engardt et al. (2017), that used historical data to simulate the deposition of acidifying and eutrophying pollutants in Europe between 1900 and 2050, relied on N emission data (rather than deposition data) to construct their model. These authors used data from the European Air Chemistry Network (EACN), which operated from the 1950s to the early 1980s, as well as more recent emission data from the EMEP monitoring network from 1990 onwards (https://www.ceip.at/).

There are dust deposition data in our region, but the records are also short. Vincent et al. (2016) compiled the existing records in the Western Mediterranean (see Table 1). The longest record (Palma de Mallorca, Spain) collects data from 1982 to 2003. Jiménez-Espejo et al (2014) reconstruct Saharan dust input for the Holocene from a sediment core of a Sierra Nevada lake. However, the number of samples representing 430 years (1600 to the present) is limited and not evenly distributed in Jiménez-Espejo study. The possibility of making statistical analysis with this data is reduced.

Camarero L, Catalan J (2012) Atmospheric phosphorus deposition may cause lakes to revert from phosphorus limitation back to nitrogen limitation. Nat Commun 3:1118

Engardt, M., Simpson, D., Schwikowski, M., & Granat, L. (2017). Deposition of sulphur and nitrogen in Europe 1900–2050. Model calculations and comparison to historical observations. Tellus B: Chemical and Physical Meteorology, 69(1), 1328945.

Holtgrieve, G. W., Schindler, D. E., Hobbs, W. O., Leavitt, P. R., Ward, E. J., Bunting, L., ... & Wolfe, A. P. (2011). A coherent signature of anthropogenic nitrogen deposition to remote watersheds of the northern hemisphere. *Science*, *334*(6062), 1545-1548.

Jiménez-Espejo, F. J., García-Alix, A., Jiménez-Moreno, G., Rodrigo-Gámiz, M., Anderson, R. S., Rodríguez-Tovar, F. J., ... & Pardo-Igúzquiza, E. (2014). Saharan aeolian input and effective humidity variations over western Europe during the Holocene from a high altitude record. *Chemical Geology*, *374*, 1-12.

Vincent, J., Laurent, B., Losno, R., Bon Nguyen, E., Roullet, P., Sauvage, S., ... & Bergametti, G. (2016). Variability of mineral dust deposition in the western Mediterranean basin and south-east of France. Atmospheric Chemistry and Physics, 16(14), 8749-8766.

Detailed comments.

1. Consider quantifying the "richness" of nutrients in dust by comparing them to local bedrock and calculating enrichment factors. Saharan dust is generally low in organic matter and phosphorus, and phosphorus is often mineral-bound and slow to become bioavailable. Also, "rich in alkalinity" is imprecise; consider rephrasing as "rich in alkalizing minerals" or "minerals that increase acid-neutralizing capacity."

ANSWER: You are right. The sentence has been changed (lines 125-126): "Saharan dust input fertilizes in phosphorus, calcium, and alkalizing minerals the oligotrophic lakes that receive it (Morales-Baquero et al., 2006b)".

Saharan dust is rich in phosphorus for the phosphorus-starved ecosystems that receive it, despite containing only modest concentrations by geochemical standards. For example, in the Mediterranean basin, major sources of P to atmosphere are Saharan dust (Migon and Sandroni 1999; Guieu et al. 2002). When aerosols come in contact with water, a part of the particulate inorganic phosphorus associated with the particles can dissolve into orthophosphate form that may be assimilated by the biological community (Ridame & Guieu, 2002). Phosphorus solubility varied substantially across studies and dust sources but some researchers found values of solubility close to 40% in water (Herut et al., 1999; Pulido-Villena et al.,2010)

The influence of Saharan dust nutrients in Sierra Nevada lakes has been previously proved by members from our research group (Morales -Baquero et al., 2006; Reche et al., 2022).

Alkalinity has been changed to alkalizing minerals.

Guieu, C., Loÿe-Pilot, M. D., Ridame, C., & Thomas, C. (2002). Chemical characterization of the Saharan dust end-member: Some biogeochemical implications for the western Mediterranean Sea. *Journal of Geophysical Research: Atmospheres*, *107*(D15), ACH-5.

Herut, B., Krom, M. D., Pan, G., & Mortimer, R. (1999). Atmospheric input of nitrogen and phosphorus to the Southeast Mediterranean: Sources, fluxes, and possible impact. *Limnology and Oceanography*, *44*(7), 1683-1692.

Migon, C., & Sandroni, V. (1999). Phosphorus in rainwater: Partitioning inputs and impact on the surface coastal ocean. *Limnology and Oceanography*, *44*(4), 1160-1165.

Morales-Baquero, R., Pulido-Villena, E., & Reche, I. (2006). Atmospheric inputs of phosphorus and nitrogen to the southwest Mediterranean region: Biogeochemical responses of high mountain lakes. *Limnology and Oceanography*, *51*(2), 830-837.

Pulido-Villena, E., Rerolle, V., & Guieu, C. (2010). Transient fertilizing effect of dust in P-deficient LNLC surface ocean. *Geophysical Research Letters*, *37*(1).

Reche, I., Mladenov, N., Pulido-Villena, E., & Morales-Baquero, R. (2022). Atmospheric inputs and biogeochemical consequences in high-mountain lakes. In *The Landscape of the Sierra Nevada: A Unique Laboratory of Global Processes in Spain* (pp. 293-306). Cham: Springer International Publishing.

Ridame, C., & Guieu, C. (2002). Saharan input of phosphate to the oligotrophic water of the open western Mediterranean Sea. *Limnology and Oceanography*, *47*(3), 856-869.

Em dashes seem to be a mark of AI writing these days aren't normally used so frequently in standard text. This is just my personal opinion, but they should be restricted to when you really need to highlight something. I would remove them.

ANSWER: Em dashes have been removed

174 – High C/N ratios can also reflect nitrogen limitation and/or diagenesis, both of which may be more plausible interpretations given that the lake is above treeline.

ANSWER: As we stated above C/N can be interpreted in terms of N limitation or in terms of organic material origin. Wolfe et al. (2003) note that the C/N ratio is associated with N availability, but it can also be interpreted in terms of the origin of organic material (terrestrial vs algal) in alpine lakes In Rocky Mountains.

On the other hand, there are numerous articles in which the authors interpret the C/N ratio in terms of the origin of the organic material (terrestrial versus algal) in alpine lakes around the world. For example, Hu et al. (2014) in two alpine lakes in Tibet, Thevenon et al. (2012) in and alpine lake in Switzerland, Jiménez-Espejo at al. (2014) in a Sierra Nevada Lake or Lami et al. (2000) in eight European alpine lakes.

Both interpretations can be plausible. The lake is above the tree line but has an extensive catchment area compared to the lake area (Lake area= 0.187 ha; Catchment area =50.9 ha; Catchment :lake ratio = 272). For example, Hu et al (2014) state that the C/N ratio values in an alpine lake are dependent of the terrestrial organic matter inputs (again a reflection of the greater catchment to lake ratio). The C/N ratio of this lake is 39, much lower than those of Borreguil lake. Moreover, The lake is above the tree line but is surrounding by alpine meadows (0.563 ha) that can apport organic material to the lake.

Of course, the two explanations presented above are not mutually exclusive, and this has been reflected in our manuscript.

- Hu, Z., Anderson, N. J., Yang, X., & McGowan, S. (2014). Catchment-mediated atmospheric nitrogen deposition drives ecological change in two alpine lakes in SE Tibet. Global Change Biology, 20(5), 1614-1628.

- Thevenon, F., Adatte, T., Spangenberg, J. E., & Anselmetti, F. S. (2012). Elemental (C/N ratios) and isotopic (δ15Norg, δ13Corg) compositions of sedimentary organic matter from a high-altitude mountain lake (Meidsee, 2661 m asl, Switzerland): Implications for Lateglacial and Holocene Alpine landscape evolution. *The Holocene*, *22*(10), 1135-1142.

- Jiménez-Espejo, F. J., García-Alix, A., Jiménez-Moreno, G., Rodrigo-Gámiz, M., Anderson, R. S., Rodríguez-Tovar, F. J., ... & Pardo-Igúzquiza, E. (2014). Saharan aeolian input and effective humidity variations over western Europe during the Holocene from a high altitude record. Chemical Geology, 374, 1-12.

- Lami, A., Guilizzoni, P., & Marchetto, A. (2000). High resolution analysis of fossil pigments, carbon, nitrogen and sulphur in the sediment of eight European Alpine lakes: the MOLAR project. Journal of Limnology, 59: 15-28.

• Wolfe, A. P., Van Gorp, A. C., & Baron, J. S. (2003). Recent ecological and biogeochemical changes in alpine lakes of Rocky Mountain National Park (Colorado, USA): a response to anthropogenic nitrogen deposition. Geobiology, 1(2), 153-168.

Results

Can you show the Chlorophyll-a to Pheophytin-a ratio, this will assist with interpreting degradation. I know some of these pigments are more or less susceptible to degradation, like diatoxanthin.

ANSWER: Regarding degradation, and given that this is a critical aspect for interpreting the pigment profile, we selected marker pigments following two criteria: (1) specificity to a taxonomic group (generally at the Division or Class level) and (2) low lability (i.e., a low number of functional groups), which ensures their persistence in the sedimentary record. Carotenoid stability depends largely on molecular structure and generally decreases with the number of functional groups (Britton et al., 1995; Sinninghe Damsté & Koopmans, 1997).

The set of 10 selected pigments provides coverage of all major algal and cyanobacterial groups while avoiding markers that, although highly indicative of specific taxa, such as fucoxanthin for diatoms, are too labile to be reliably used in down-core analyses. For this reason, we avoided chlorophyll-based pigments and focused on carotenoids. In particular, diatoxanthin is a highly reliable marker for diatoms due to its low number of functional groups, which makes it considerably more stable than fucoxanthin (Supplementary Table S2).

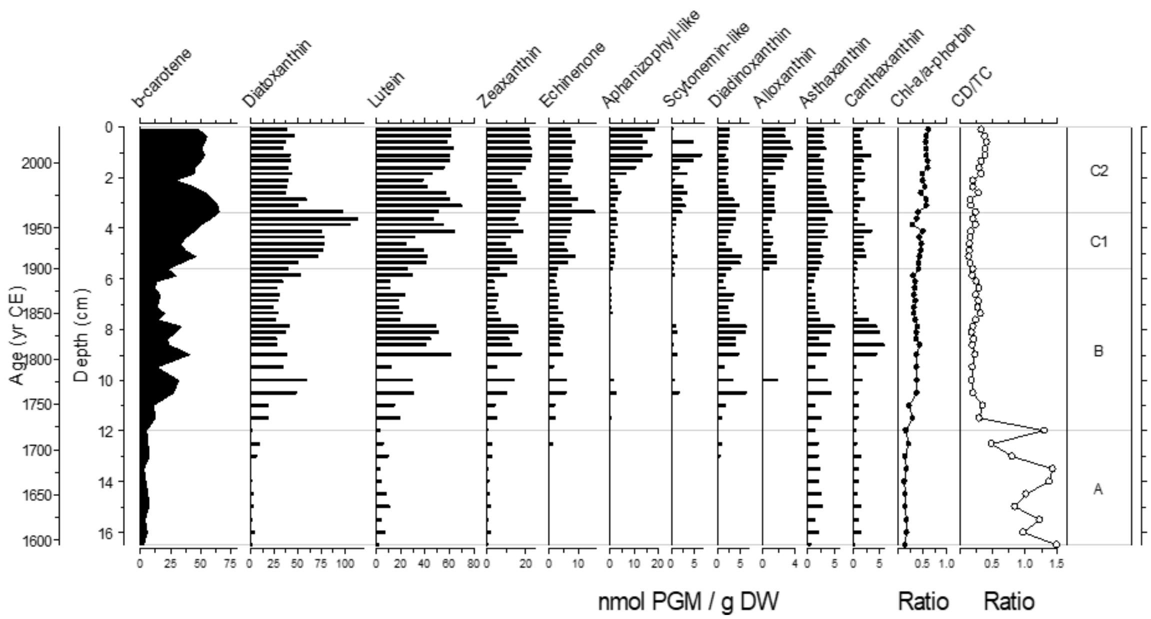
We also analysed Chl-a and several Chl-a degradation products, including chlorophyllide a, one allomer and two epimers of Chl-a, one pheophorbide, and two pheophytins (Table S2). We defined the sum of Chl-a and its derivatives as a-phorbin. Additionally, we calculated a Chl-a preservation index using the Chl-a/a-phorbin ratio. This index ranges from 0 (low Chl-a preservation) to 1 (high Chl-a preservation) (Buchaca & Catalan, 2007). When plotted along the sediment record, the index shows slightly higher values in the more recent sediments, ranging from 0.25 to 0.5 (Figure S2 in Supplementary). We interpret these results as evidence that pigment degradation is not a major concern in this record.

We have included a new table and a new figure in Supplemenatry Material.

Britton, G. (1995). Structure and properties of carotenoids in relation to function. *The FASEB Journal*, *9*(15), 1551-1558.

Buchaca, T., & Catalan, J. (2007). Factors influencing the variability of pigments in the surface sediments of mountain lakes. *Freshwater biology*, *52*(7).

Sinninghe Damsté, J. S., & Koopmans, M. P. (1997). The fate of carotenoids in sediments: an overview. *Pure and applied chemistry*, *69*(10), 2067-2074.



Supplementary Figure S2. Pigment profiles of the 10 selected marker pigments and β-carotene expressed as concentrations (nmol PGM / g DW) and the ratios of Chl-a/a-phorbin as a Chla preservation index and the ratio CD/TC (chlorophyll-a derivatives vs. total carotenoids) as indicative of higher contribution of autochthonous production when the ratio is low. Four main biostratigraphic zones were established based on constrained incremental sum of squares partitioning of pigment concentration with Rioja-package version (0.9-15.2) (Juggins, 2017) using R version 3.5.3 (R Core Team 2019).

Supplementary Table S2. Pigments identified from lake sediments included in this study and their taxonomic affinities. The number of functional groups (FG) of the pigment molecular structure is also indicated for carotenoids.

Peak	RT (min)	PeakCode	Full name	lmax (nm)	Taxonomic affinity	FG
1	0.69	Ph_0	Chlorophyllide-a	432, 665	Chl-a degradation product, abundant in diatoms	
2	1.02	Ph_3	Chl-c1	441, 635	Diatoms and Chrysophytes	
3	1.04	Car_7_8	UVA2- Scytonemin	387	Cyanobacteria exposed to high UV radiation	4
4	1.09	Ph_5b	Chl-c2	448, 636	Cryptophyta, Dinoflagellates, Diatoms and Chrysophytes	
5	1.53	Car_5	Fucoxanthin	452	Chrysophytes and diatoms	5
6	1.44	Ph_2	Phaeophorbide-b1	438, 650	Chl-b degradation product (grazing processes)	

7	1.92	Ph_1a	Phaeophorbide-a	411, 664	Chl-a degradation product (grazing processes)	
8	2.19	Ph_2a	Phaeophorbide-b2	438, 650	Chl-b degradation product (grazing processes)	
9	2.32	Car_9z	Asthaxanthin	478	Zooplankton (Crustacea)	4
10	2.40	Car_17d'	Aphanizophyll	450, 477, 507	N2-fixing cyanobacteria	3
11	2.52	Car_10a	Diadinoxanthin	449, 479	Dinoflagellates, chrysophytes and diatoms	3
12	2.71	Car_12_5	Antheraxanthin	448, 477	Charophyceae, Rodophyta and vascular plants	3
13	2.91	Car_18	Alloxanthin	456, 484	Cryptophytes	2
14	3.11	Car_17	Diatoxanthin	456, 485	Diatoms and chrysophytes	2
15	3.21	Car_11	Lutein	448, 475	Chlorophyta	2
16	3.31	Car_12	Zeaxanthin	454, 483	Cyanobacteria, Chlorophytes	2
17	3.55	Car_16	Canthaxanthin	472	Cyanobacteria and zooplankton (Cladocera)	2
18	3.85	Ph_13	Chl-b	462, 650	Chlorophyta	
19	3.95	Car_17_3	Unknown	448, 475		
20	4.21	Ph_19a	allomer Chl-a	433, 663	Chl-a degradation product	
21	4.40	Ph_19	Chl-a	431, 662	Cyanobacteria, algae and vascular plants	
22	4.51	Ph_19e	epimer1 Chl-a	431, 662	Chl-a degradation product	
23	4.65	Ph_19e'	epimer2 Chl-a	431, 662	Chl-a degradation product	
24	4.73	Car_21	Echinenone	470	Cyanobacteria and zooplankton (Cladocera)	1
25	4.95	Ph_23	Phaeophythin-b1	435, 651	Chl-b degradation product (senescence)	
26	4.99	Car_25a	b-carotene	421, 454, 480	Cyanobacteria, algae and vascular plants	0
27	5.07	Ph_25	Phaeophythin-a1	410, 665	Chl-a degradation product (senescence)	
28	5.19	Ph_26	Phaeophythin-b2	436, 651	Chl-b degradation product (senescence)	
29	5.27	Ph_28	Phaeophythin-a2	410, 665	Chl-a degradation product (senescence)	

Also, I know its common to plot pigments etc by relative abundance with similar sized x axis, but it makes it hard to see the temporal changes in indicator pigments where the concentrations are less than 10%. Can you plot your explanatory variables along side your data? You have a mix of graphs with time on the y or x axis, which makes it difficult to evaluate temporal relationships. Pick one way of showing the data, and stick to it. There is a lot of figure space given to PCA plots and plots that are repetitions (d15N vs Aphanizophyll (repeated from fig 3), but I would rather see more interpretable graphs, e.g. showing temporal changes in proxies alongside the predictor variables as well as predictor plots.

ANSWER: Manuscript figures have been thoroughly changed following your and reviewer 1 suggestions. Please, see the new figures.

All the figures have now the same axis to evaluate temporal relationships. Some figures have been merged (see figure 3). d15N vs Aphanizophyll figure has been removed.

You should use statistics to evaluate whether or not you are observing significant changes in your proxies through time.

ANSWER: We are sorry, but we are not sure what you are asking for. We considered applying a regression or another model to the proxies and time, but we are not sure if that is what you mean. We hope you can clarify this for us in the second round of reviews.

Why is the C/N ratio being compared to climate and atmospheric variables, what are you specifically testing here?

ANSWER: You are right. The C/N ratio can result from changes in climate, such as an increase in temperature enhancing algal production, provoking changes in degradation, or N limitation. However, it is difficult to determine the mechanism behind it using our data.

C/N is not a considered response variable in this article, in which we discuss the increase in algal biomass (chlorophyll-a and β-carotene). Moreover, we do not use this result in the Discussion section, so it makes no sense to include it in the Results section. This analysis has been eliminated.

Regarding dust, I did not see Zr/Al or similar elemental proxies presented. Were these measured, or are you relying on external studies to make these links for you? It is important to show how dust inputs are linked to your dataset. Plotting dust indices (e.g., wNAO, SPI) directly alongside pigment proxies would help establish clearer connections between drought, dust production, and specific pigment responses. Or better yet, find other clear data streams showing the relevant data. I'm not saying these relationships don't exist in the real world or even in your data, I'm just saying that they way you have presented the data is very cursory and does not make it clear.

ANSWER: The relationship between Zr/Al ratio from Río Seco Lake and Saharan Ca from an ice core from the Alps with Saharan deposition drivers (wNAO and SPI) were extensively explained in Jiménez et al (2018).

In Jiménez et al., (2018) we compared the sedimentary Zr/Al ratios directly measured in Río Seco Lake sedimentary record and the Saharan Ca concentration record derived from an Alps ice core (Preunkert & Legrand, 2013) with the wNAO (winter North Atlantic Oscillation) index and the Sahel precipitation index records as representatives of Saharan deposition tendencies in Sierra Nevada. The Zr/Al ratio from RS and the Ca record derived from the Alps ice core show a highly significant correlation ($r = .785$, $p = 8.8 \times 10^{-4}$). The significant correlations found between Zr/Al ratio and both indices SPI (r = _0.862, p = 2.0 x 10-6) and wNAO (r = .361, p = .049) and between the Ca ice core and SPI (r = -0.448, p = 2.8 x 10-5). These results support the assumption that we can consider wNAO and SPI to be predictors of the transport and intensity of Saharan dust events in Sierra Nevada and representative of P deposition trends in Sierra Nevada

We believe that it is clearly stated in the M&M section that we use the drivers of Saharan dust deposition to evaluate deposition in the Sierra Nevada (please, see lines 242-248): "The intensity of Saharan dust emission and transport has been linked to wNAO (winter North Atlantic Oscillation; Moulin et al., 1997) and to the Sahel drought (Chiapello et al., 2005; Moulin and Chiapello, 2004). Furthermore, Jiménez et al. (2018) demonstrated that the SPI and wNAO indices can be utilised as predictors of the transport and intensity of Saharan dust events in Sierra Nevada, reflecting atmospheric P and Ca deposition trends in this region. These indices exhibited strong correlations with the zirconium-to-aluminium (Zr/Al) ratio—a proxy for Saharan dust deposition—measured in a sediment core from one of the Sierra Nevada lakes, as well as with Saharan calcium concentrations in an ice core from the French Alps, which are indicative of Saharan dust events (Preunkert and Legrand, 2013). Consequently, SPI was employed for this purpose in the present study".

Please note that the wNAO variable has been removed from the revised manuscript in response to a comment from Reviewer 1.

We do not believe to be necessary to show again the graph and to write about this relationship in M&M and Results. These are data and results from a previous article and we are trying to shorten the manuscript following your and reviewer 1 suggestion.

Jimenez, L., Rühland, K. M., Jeziorski, A., Smol, J. P., & Pérez-Martínez, C. (2018). Climate change and Saharan dust drive recent cladoceran and primary production changes in remote alpine lakes of Sierra Nevada, Spain. *Global change biology*, *24*(1), e139-e158.

Preunkert, S., & Legrand, M. (2013). Towards a quasi-complete reconstruction of past atmospheric aerosol load and composition (organic and inorganic) over Europe since 1920 inferred from Alpine ice cores. *Climate of the Past*, *9*(4), 1403-1416.

Line 415, did you explicitly show markers of increased dust deposition in your lake or is this entirely based off of other publications from nearby lakes? You haven't not made this relationship clear or explicit.

ANSWER: No, we do not have markers of Saharan dust deposition in the study lake but in a nearby lake (Río Seco Lake, Jiménez et al., 2018).

In Jiménez et al., (2018) we compared the sedimentary Zr/Al ratios directly measured in Río Seco Lake sedimentary record and the Saharan Ca concentration record derived from an Alps ice core (Preunkert & Legrand, 2013) with the wNAO (winter North Atlantic Oscillation) index and the Sahel precipitation index records as representatives of Saharan deposition tendencies in Sierra Nevada.

The Zr/Al ratio from Río Seco Lake and the Ca record derived from the Alps ice core show a highly significant correlation ($r = .785$, $p = 8.8 \times 10^{-4}$). Significant correlations were found between Zr/Al ratio and both indices SPI ($r = -0.862$, $p = 2.0 \times 10\text{-}6$) and wNAO ($r = .361$, $p = .049$) and between the Ca ice core and SPI ($r = -0.448$, $p = 2.8 \times 10\text{-}5$). These results support the assumption that we can consider wNAO and SPI to be predictors of the transport and intensity of Saharan dust events in Sierra Nevada and representative of P deposition trends in Sierra Nevada.

To avoid confusion, we explicitly state "Saharan dust deposition drivers" instead of 'Saharan dust deposition' (line 445). Furthermore, we believe that it is clearly stated in the M&M section that we use the drivers of Saharan dust deposition to evaluate deposition in the Sierra Nevada (please, see lines 242-248): "The intensity of Saharan dust emission and transport has been linked to the Sahel drought (Chiapello et al., 2005; Moulin and Chiapello, 2004). Furthermore, Jiménez et al. (2018) demonstrated that the Sahel Precipitation Index (SPI) can be utilised as predictor of the transport and intensity of Saharan dust events in Sierra Nevada, reflecting atmospheric P and Ca deposition trends in this region. These indices exhibited strong correlations with the zirconium-to-aluminium (Zr/Al) ratio—a proxy for Saharan dust deposition—measured in a sediment core from one of the Sierra Nevada lakes, as well as with Saharan calcium concentrations in an ice core from the French Alps, which are indicative of Saharan dust events (Preunkert and Legrand, 2013). Consequently, SPI was employed for this purpose in the present study".

Please note that the wNAO variable has been removed from the revised manuscript in response to a comment from Reviewer 1.

Preunkert, S., & Legrand, M. (2013). Towards a quasi-complete reconstruction of past atmospheric aerosol load and composition (organic and inorganic) over Europe since 1920 inferred from Alpine ice cores. Climate of the Past, 9, 1403–1416.

Jimenez, L., Rühland, K. M., Jeziorski, A., Smol, J. P., & Pérez-Martínez, C. (2018). Climate change and Saharan dust drive recent cladoceran and primary production changes in remote alpine lakes of Sierra Nevada, Spain. Global change biology, 24(1), e139-e158.

Why are you comparing your results to examples of lakes below treeline?

ANSWER: There are few examples of pigment analysis in alpine lakes. We have tried to eliminate references to non-alpine lakes. Following reviewer 1's suggestion, the paragraph containing examples of non-alpine lakes (former lines 420-425) has been removed in order to shorten the discussion.

The discussion should be written as to highlight your main take home messages. What have we learned from your study, specifically? It is ok to compare and contrast to other studies, but this discussion opens paragraphs with other publications, more like a literature review, and is consequently excessively long.

ANSWER: Discussion has been shortened and focused in our results following your and reviewer 1 suggestions. Discussion has been reduced from eight pages to less than five pages.

Personal opinion, but the titles of the subheadings are a bit long

ANSWER: Subheading titles have been considerably shortened

439 How are you defining 'alpine', alpine is above treeline. Not all these publications are for lakes in the alpine. Mountain does not equal alpine. Please also be precise with citations.

ANSWER: You are right, alpine is above the tree line and some of the lakes referenced are nor alpine lakes. We have changed alpine lakes by mountain lakes (line 461).

645 would you expect very colored DOC at this elevation though? … a few lines later you mention a bog. Is this in the catchment? What vegetation is in the bog? You probably should include that in the site description!

ANSWER: No, we do not expect very colored DOC at this elevation.

In relation to our mention to bog: Sorry, we meant a peat bog (i.e. wet alpine meadows) surrounding the lake. Algae can develop in these habitats. We have changed the sentences to avoid misunderstanding (lines 567-570): "It seems reasonable to suggest that the observed increase in scytonemin and zeaxanthin may be linked to the growth of cyanobacteria, Chlorophyceae and Zygnematophyceae in radiation-exposed environments within the lake and associated wet alpine meadows (Hauer et al., 1997)".

We have included a brief description of the alpine meadow vegetation in the study area (lines 129-132): "The lake is surrounded by approximately 0.56 hectares of alpine meadows, the flora of which is mainly composed of Cyperaceae, Poaceae and Fabaceae, with frequent genera including Carex, Nardus, Festuca and Scorzoneroides (Pérez-Luque et al., 2015). The wetter meadow shows bryophyte species such as Drepanocladus fluitans".

Pérez-Luque, A. J., Sánchez-Rojas, C. P., Zamora, R., Pérez-Pérez, R., & Bonet, F. J. (2015). Dataset of Phenology of Mediterranean high-mountain meadows flora (Sierra Nevada, Spain). *PhytoKeys*, (46), 89.