nature portfolio

Peer Review File

Andean volcanism, ocean fertilization, marine ecosystem turnover, and global cooling in the Late Miocene

Corresponding Author: Barbara Carrapa

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

** Please ensure you delete the link to your author home page in this e-mail if you wish to forward it to your coauthors **

Dear Professor Carrapa,

Apologies for my delay in sending an editorial decision letter. Your manuscript titled "Drivers of Cenozoic global cooling and the impacts of the Andes on ocean fertilization and Late Miocene Cooling" has now been seen by 2 reviewers, whose comments are appended below. You will see that they find your work of some potential interest. However, they have raised quite substantial concerns that must be addressed. In light of these comments, we cannot accept the manuscript for publication, but would be interested in considering a revised version of your article that is presented not as a Review Article but as a Research Article that is focused on showing your modeling results on the effect of Andean Miocene volcanism on ocean fertilization, and that fully addresses the concerns raised by both reviewers.

We hope you will find the reviewers' comments useful as you decide how to proceed. If additional work allows you to either incorporate or refute these criticisms, we will be happy to look at a substantially revised manuscript. If you choose to take up this option, please either highlight all changes in the manuscript text file, or provide a list of the changes to the manuscript with your responses to the reviewers.

When resubmitting, please provide a point-by-point response to the reviewers' comments. Please submit your responses as a separate file, distinct from your cover letter where you can add responses to the Editors' comments that you do not want to be made available to the reviewers. Word files are preferred. We recommend that any figures, tables or graphs that are included in the response to reviewers are also included in the main article or Supplementary Information.

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Please do not hesitate to contact us if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Carolina Ortiz Guerrero, Ph.D.
Associate Editor, Communications Earth & Environment
Consulting Editor, Communications Sustainability

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

In this paper, Carrapa et al. review mechanisms driving pCO2 change over the Cenozoic and test some of them in cGENIE experiments. They also provide atmosphere particle simulation (using HYSPLIT code) to track pathway of Andean volcanic dust together with CESM simulations with activated marine biogeochemistry to discuss the effect of Andes volcanism on southern ocean productivity. They then discuss how this would have affect multi-centennial ocean carbon fluxes and make a tentative link to pCO2 and cooling over the late Miocene.

I find this paper interesting and it provide a good review of processes impacting long term pCO2. Such review and results worth being publish. The results the authors obtained on the effect of Andes volcanism is particularly interesting, so is the methodology but this part would be better highlighted and have more space for discussion in a standalone paper. I think the modeling approach is appropriate.

I would however highlight that the focus of the paper is currently not really clear and the main message difficult to catch. I think it comes from the choice to combine a long review on processes driving Cenozoic global cooling with a more focus study on the Andes. Both could be easily the focus of a single paper, as each part anyway comes with independent modeling design; and actually it really reads like two independent studies have been pasted together to make a review paper. It though does not have the style of a formal review as it introduces new material. The main drawback of this, to my opinion is that there are too many information that are not well articulated together and that the choice of performed simulations is not well justified and can seem a bit random without having to dig to the method section/supplementary material. I made some comments that could help the authors reorganize the paper so the focus is more clear and the reader does no get lost with the amount of information. Note that I am not a carbon cycle specialist and are not able to provide a thorough review on the mechanism for global Cenozoic cooling or cGENIE aspect of the paper. I will let another reviewer comment on this.

Main comments :

I think the outcome of this study would be way larger would the authors decide to focus on 1) a thorough review of Cenozoic cooling trends and processes - which they did very well , 2) refocus on reviewing volcanism contribution to climate cooling, including the effect of Andes on the Miocene or on ocean fertilization during the Neogene. I think the point 2) is close to what is done here and might explain the choice of (some of the ?) cGENIE experiments. This is indeed never clear why the authors chose to test one of the mechanisms rather than the others and whether it related to full Cenozoic trend or mostly Neogene. I understand that all the technical information are provided as supplementary material (and that in this specific journal format it would end up as method sections) but I think the main text is missing part of what is necessary to really understand what is done. This feeling might also be related to the fact that the paper focus is not well specified and buried below the review aspect. I do think that this paper as I is build should be consider a review as it introduce new modeling work which purpose are not to test all the reviewed process. Some of the processes described here (those that are fully detailed but not use to support the modeling results) could therefore mostly end up as supplementary text, which would give more space to clearly describe what has been done, which processes tested and why directly in the main text.

Few examples of places where things could be improved :

- l102 - ‘Here we discuss various posited drivers of Cenozoic climate change and illustrate their potential impacts with complementary Earth System modeling approaches.’ . This is a bit different to what is done later in the paper as authors review all the potential driver of Cenozoic pCO2 changes but only test some of them in a modeling framework.

- l113 - It is stated even before describing in details the processes affected pCO2 that “It should be noted that the goal of these idealized cGENIE experiments is not to provide a quantitative estimate of Cenozoic changes in the forcings over time but rather to demonstrate the long-term impact and direction of change of individual forcing mechanisms. In reality, we expect that Cenozoic cooling resulted from the complex interplay between several forces (43, 44).” While I think this warning is mandatory it is not very clear why the focus is on some of the processes only and how did the authors decide which processes to focus on or not. Because the paper continue with a full literature review on various kind of processes I would suggest this information arrive after reviewing mechanisms and comes with an explanation on why they made such choices.

- l261-382 - while the previous paragraph incorporates at least some Cenozoic literature, the authors suddenly shift the focus much more on the Miocene. This make sense as those mechanisms are those relevant for the ‘Andean volcanism’ part that just come later but it makes the previous paragraphs on weathering and tectonics sounding to broad and a bit out of context.

- line 410 the authors briefly state they simulated particle dispersion but it is not clear that the setup is modern climate, which might have slightly different winds or ocean circulation patterns than the late Miocene. It might not be a problem as the goal is to demonstrate the effect of volcanic ashes on the productivity but this has to be stated to avoid confusion.

Relative to the point 1), the introduction mostly focused on the Miocene, Andes volcanism and the fertilization effect on ocean productivity, but the full first half of the paper then focus on reviewing the global carbon cycle components and introduce experiments testing only some processes in isolation. This is really disturbing and contribute to the focus of the paper getting lost pretty quickly. As a reader I got totally lost in that as I was convince from title to line 99 that I would read work on ‘Andes and late Miocene cooling’.

The authors suggest a link between the Andes volcanism and the LMBB / LMCIS and make the link between SiO2 volcanic input and increase in diatoms. However, figure 3 seems to show the diatom abundance and biogenic opal is increasing after the LM cooling and LMBB. Could the authors explain how they would reconcile both records ?

Minor comments :

L. 35 ‘ with a focus on the contribution of the Andes to the southern oceans over the last ~20 Myr.’ —> Late Miocene ?

L 69 ‘Columbia River Basalts and the Miocene Climatic Optimum; (18),’ —> the closing parenthesis is lacking here.

l. 218 - “The magnitude and timing of these variations are purported to explain much of the temperature decline from the notably warm early-to-middle Miocene, when global temperatures exceeded present levels by approximately 10–12°C.” What is the reference for this ?

L. 400 - “The west coast of South America has been a region of major upwelling region since the start of the Cenozoic (160) and since at least the Miocene (161)” —> Since at least the Miocene is redundant, as it is part of the Cenozoic. I suggest the authors change for ‘The west coast of South America has been a region of major upwelling region since the start of the Cenozoic (160, 161)’

l. 436 - 439. Please provide the references for HYSPLIT and CESM2.1.5

l. 465 - 467 The authors states that “This experiment indicates that most of the ash from the Puna-Altiplano region is transported to the South Atlantic by prevailing westerly winds. The resulting flux of ash deposited over the ocean extends eastward from the South Atlantic to the southern Indian Ocean (Figure 5A).” I might have missed something but how do they relates this patterns with the Pacific signal they introduce l. 419 to 429 ?

Figure 1 : The arrows direction are difficult to catch : for example the orange to pink seems to be causation arrows but the blue to maroon would be reverse (degassing to spreading rate). I suggest the authors to change the arrow direction depending on how the link goes.

Reviewer #2 (Remarks to the Author):

This paper describes mechanisms driving global cooling over million-year timescales and utilizes climate system models to explore these different mechanisms, with a focus on the role of Andes volcanism on global cooling via ocean fertilization. Understanding the mechanisms of global cooling and the interactions between carbon cycling and climate are of great interest in paleoclimate research. This study includes a comprehensive review of mechanisms driving cooling and combines a high-resolution CESM model with an intermediate complexity model to study how short-term biogeochemical processes such as the marine biological pump may influence carbon sequestration on longer timescales. The authors conclude that the Late Miocene Cooling was driven by enhanced diatom productivity in the Southern Ocean (which draws down atmospheric CO2), stimulated by nutrient input from Andean volcanic eruptions. Overall, this research provides a unique approach to

better understanding mechanisms for global cooling and should be published in Earth and Environment Communications after some clarifications and restructuring is made.

The first part of the paper describes factors controlling Earth's carbon cycling and global climate including weathering, tectonics, ocean fertilization, and the biological pump. The second part of the paper then uses modeling to provide perspective on the role of Andean volcanism on the ocean fertilization mechanism. As currently written, it is unclear what is review and what is the author's contribution (modeling component). A clear statement that this is the aim of the paper would be useful somewhere in the introduction. Additionally, the review of mechanisms for cooling could benefit from subheadings. Throughout this section, careful attention to topic sentences, and only including relevant information related to these topic sentences would make it much easier to follow. In general, the link between the information given and its relevance to atmospheric CO2 or role in the global cooling process needs to be clearer in some places (see line by line comments below). It would also be useful to include some details of the model approach and methodology in the main text as this is a major component of what the authors are presenting. Although a good job is done explaining the limitations of the model approach, information about the approach is required up front for the reader to assess the validity of the results.

Based on modeling evidence, increased Andean volcanism is argued as an important mechanism driving global cooling during the LMC. However, when compared to paleoclimate data, there is a clear lag between peak volcanism and changes in diatom productivity and extinction rates (Figure 4). However, there does seem to be more similarity between the Andean volcanism histogram and d13C timeseries, which is a primary data set indicating changes in global carbon cycling. Furthermore, it would be helpful to directly compare this alongside SST records in Figure 4. The time lag between these systems needs to be explored and explained in the text (globally complex system, perhaps other volcanic regions at play?).

Although considering the entire Cenozoic is relevant for considering the processes driving global cooling on multi-million year timescales, I am unsure what the value is of including this entire time period, especially when the majority of paleoclimate data discussed in Figures 3 and 4 only includes data from the Early Miocene to present. I wonder if the discussion could instead be focused on the past 20 million years of Earth's history?

Towards the end of the paper where the authors discuss the role of Andean volcanism on Southern Ocean productivity, it would be very useful to have a schematic including the Andes and areas of the Southern Ocean illustrating the regional processes that influence carbon cycling between different reservoirs. Where possible, label the geographic locations mentioned in the text. Having a figure like this would be a great way for readers to visualize the processes occurring.

Line-by-line comments:
1: Consider "Neogene" instead of "Cenozoic" and "impacts of Andean volcanism"
21: replace "geological" with "million-year"
30: add "which alter atmospheric CO2" at the end of the sentence to make the link between climate and atmospheric CO2
31: What is meant by "longer timescales?"
32: addition of nutrients by which processes? Weathering?
35: Rather than southern oceans throughout, refer to the whole ocean as the "Southern Ocean". If you need to refer to specific areas, you can say "Atlantic sector of the Southern Ocean".
39: Add comma after "cooling"
43: "causing" rather than "sparking"?
48: replace "global climate change" with "cooling" since this is the focus?
49-53: make one sentence?
52: Also figure 2
60: Specify when the Miocene was
62: Specify when the MCO was
66-68: Can you add a sentence to explain why these might have been important for understanding carbon cycling? Also the Monterey Excursion is shown on the figure but not in the text. This might be something you want to include to illustrate the coupling between climate/ice sheet volume/sea level during the MCO and MMCT
68: CRB associated with MCO in general- not sure if there are other instances in the Miocene of this association. Could also talk about the mid-ocean ridge spreading rate hypothesis here.
76: Increase in atmospheric CO2? Specify what is meant by "shorter timescales"
81: "nutrient-driven". Also add "(alkalinity)" at the end of the sentence? This concept may warrant more explanation
86: List the macro and micronutrients here that are supplied by volcanic ash and glacial dust
91: state that ignimbrites are silica-rich? Would be really useful to have a map/schematic here to refer to the geography and relevant carbon cycle processes
95: Reverse intensified Hadley circulation and aridity since the first contributes to the second
97: Unclear form this sentence that there is a modeling component of this study
105: First mention of calcium carbonate weathering and how this links to global carbon cycling
116: replace "forces" with "forcing mechanisms"
123: Make this a subheading, work on topic sentences in this section
125: climate refers specifically to temperature and precipitation? How are these accounted for in the models? Large drop in temp and precipitation after the MMCT
130: explain more how these things link to CO2 partitioning between different systems
162: The distinction between what is a review and what are new ideas out forth by modeling results very unclear in this section. Needs to be clear separation of the two.
167: I would expect that cooling would result in more intense latitudinal temperature gradients resulting in stronger winds, upwelling, and increased vertical mixing of nutrients. Could it also be that cooler ocean temperatures result in decreased

microbial remineralization of POC in the water column?
188: Can this proxy data be added to the paleoclimate data figure?
196: Ocean circulation mentioned here but then not elaborated on again. Important for latitudinal temp gradients which drives global atmospheric circulation?
204: cite
220: cite
226: Again, switch from review to modeling results is hard to follow
332: pCO2 not shown on the figure
348-368: Unclear how this information is relevant to carbon cycling
370: When did this change occur?
385: Again would be great to have a map and schematic
393: Add “input into the ocean” after “silica”
394: If Andean ocean fertilization is a driver, why is there a lag between peak volcanic activity and the LMC? Why is the diatom peak and increase in extinction rate after this? What is the relationship of all of this to the d13C curve?
420: Again, why the lag?
425: A lag of about 1 million years
500: Is there any sedimentary evidence from deep ocean drill cores to suggest that there was an increase in biogenic silica accumulation at this time? Maybe look just in the Southern Ocean.
511: Great discussion of the limitations. I wonder how this might time into the time lag questions raised above.
519: This overall is written well and seems like a summary of above. Concepts very clear here. Still not super clear on the main messages of the paper
554: These data need to be discussed earlier. Carbon isotopes one of the primary lines of evidence for long-term changes in global carbon cycling
569: How does whale preservation indicate productivity? Frequence of whale fossil occurrences? What about a major change in sea level which caused them all to become beached in one place?
580: This is a great point, maybe bring into the introduction to justify the approach of the study?

Figure 1
What about temperature of deep ocean controlling CO2 solubility? Large decrease in deep ocean temps across the MMCT would have changed this

Figure 2
Can remove top plot? Not really discussed in the paper
Use bars to show the duration of climate events eg. MCO, add in MMCT as well. Remove anything that isn’t discussed in text eg. Monterey Event
Add an arrow on the d18O axis to indicate which direction is ice growth/deep ocean cooling
E needs citation
Add CCD depth or carbonate preservation through time? Is there data for these or have they been modelled?

Figure 3
Difficult to understand what this is without more detail in the main text
Increased Ca weathering? Silicates? CaCo3?
Would be helpful to put % change on the x-axis for each modeling output
Specify in caption what long-term means (I think million year?)
State the boundary conditions used for the model briefly ie. the LMC period

Figure 4
Andean volcanic activity seems to follow the d13C curve closely. There is a significant lag between volcanic activity diatom silica export, and increase in extinction rate that needs to be explained. Unsure if this is strong support of the importance of the ocean fertilization mechanism for pCO2 drawdown. Global cooling records should also be shown on this plot.

Figure 5
Clearly an increase in diatom productivity after volcanism on a short timescale. But how does this translate to burial of OC on much longer timescales?

Supplementary Table
Consider including in main text as it is quite helpful to understand what was being tested with the model

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Decision Letter:

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Dear Professor Carrapa,

Your manuscript titled "Andean volcanism, ocean fertilization, marine ecosystem turnover, and global cooling in the late Miocene" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment.

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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Best regards,

Alice Drinkwater, PhD
Associate Editor
Communications Earth & Environment
Consulting Editor
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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The manuscript by Carrapa and collaborators has been now refocused on Andean volcanism and how it could have contributed to change in productivity, CO2 uptake and marine ecosystem during the Late Miocene. The overall message is thus now way clearer and the paper reads better.
The authors cleverly re-designed the simulations framework so that cGENIE simulations are now an extension of the CESM runs, both enabling to respectively understand the millennial and centennial timescale response of productivity and CO2 uptake to volcanic dust emission. The authors then used simulation results and data compilation to discuss potential contribution of Andean volcanism to the Late Miocene cooling and the coeval marine ecology shift.
The methodology and modeling results are convincing to me and the authors took into account comments from previous round of review. The introduction now gives more detailed information on the Late Miocene, volcanism and productivity, which helps understanding the story. The data compilation part of the work is also now better emphasized, and the authors also incorporated more details on the modeling methodology (that were previously in the supplementary material), which I think is great. They also conducted several sensitive experiments with the models to ensure their results are robust.

I really appreciate that the conclusions are not oversold and limitation of the numerical tools are clearly stated. This paper is a nice contribution toward better understanding the driving factor of Late Miocene climate change. So congrats, and I really look forward this paper to be publish and to see further work on this topic.

Minor comments:
Authors use both « late Miocene » (eg. l. 53) and « Late Miocene » (eg. l. 52) across the paper. It would be good to homogenized and keep one or the other.

L. 67. « The Late Miocene Biogenic Bloom ». I suggest the authors put age interval for this event as they do it for LMCIS.

- Authors used both « HYSPLIT » and « Hysplit ». I suggest to keep with « HYSPLIT » across the paper.

Anta-Clarisse Sarr

Reviewer #2 (Remarks to the Author):

General comments
Overall, this version of the manuscript is improved as it now reads as an independent modeling and data comparison study than as a review and new study. The authors have addressed some of the limitations of the study clearly and include information about processes which may not be included in the model study. The restructuring has made the aim and outcome of the study more clear. There are still some minor edits that should be made to correct inconsistencies (for example, consistent use of capitalization of Late Miocene, correct labeling of panels in Fig. 2, order of figures, clear topic sentences, instances of repetition).

Line-by-line comments:
2: late should be capitalized, check for consistency throughout document and figures
25: When?
33: Clarify the link to the ocean, deposition of nutrient-rich ash in the ocean. Humboldt current should be indicated on the map
38: Put data compilation first as that is what is presented here
39: add timescales after millennial
42: increased carbon export to the sediment? Make the mechanism by which this happens clear here
46: Need heading here, Introduction? Is mountain mention relevant? Sentence is quite long consider shortening or splitting into two
49: citation?
51: New paragraph here? New topic
57: remove “in” after changing. Unclear especially to unfamiliar readers what the link is between these things listed and climate
61: briefly give some examples of limiting nutrients
64: Make this the topic sentence?
65: evidence for increased marine productivity is the LMBB? Merge these sentences for clarification
79: from rather than at
80: add jet, or spell out acronym here

101: and cooling?
101: repetition here
103: Add to investigate x we do y
104: Spell out these acronyms for the benefit of non-modelers
107: also million year timescales?
115:Maybe call this a data comparison or compilation? You could add a small paragraph about what you're doing and why (utilizing existing data) before launching into this section
117: Why is the DIC gradient relevant? Instead maybe say or define export production?
118: key producers in the Southern Ocean/regions of upwelling
124: repetition of food web from previous paragraph
126: refer to figure.
136: feedback rather than influence
147: Refer to figure
154: citations for Antarctic ice sheet growth and/or global sea level fall?
166: citation for SL change?
173: capitalize HYSPLIT
197: how was significance assessed?
209: This could also be because of carbonate dissolution as bottom water conditions changed
270: This is largely repeated from above, consider where and how best to present this information
293: Is this the summary section? Confusing as to when one section ends and another starts without subheadings

Figure comments
Figure 1
The 5 Ma mark is in the wrong position
387: add (SST) in caption since this is what is listed in figure

Figure 2
Add Humboldt Current
399: unclear what hydrologic deficit is and how to make sense of it in figure.

Figure 3
Labelling is inconsistent (B and C switched in caption).
425: Refer to isotopes in the same way as fig 1 (on axis). Can say benthic foraminifer isotopes and alkenone SST records

REVIEWER 1

I find this paper interesting and it provide a good review of processes impacting long term pCO2. Such review and results worth being publish. The results the authors obtained on the effect of Andes volcanism is particularly interesting, so is the methodology but this part would be better highlighted and have more space for discussion in a standalone paper. I think the modeling approach is appropriate. We have significantly revised the manuscript to include new modeling (cGene) and a more in-depth description of the modeling approach, results and discussion.

"I would however highlight that ***the focus of the paper is currently not really clear and the main message difficult to catch****. I think it comes from the choice to combine a long review on processes driving Cenozoic global cooling with a more focus study on the Andes. Both could be easily the focus of a single paper, as each part anyway comes with independent modeling design; and actually it really reads like two independent studies have been pasted together to make a review paper. It though does not have the style of a formal review as it introduces new material. The main drawback of this, to my opinion is that there are too many information that are not well articulated together and that the choice of performed simulations is not well justified and can seem a bit random without having to dig to the method section/supplementary material. I made some comments that could help the authors reorganize the paper so the focus is more clear and the reader does no get lost with the amount of information. Note that I am not a carbon cycle specialist and are not able to provide a thorough review on the mechanism for global Cenozoic cooling or cGENIE aspect of the paper. I will let another reviewer comment on this."*
We have largely rewritten the paper to focus on the role of Andean volcanisms in the Late Miocene on Southern Ocean fertilization, via dust-ash inputs, primary productivity, marine ecosystems and global cooling. We have run new model simulations (cGene) that now integrate the results from Hysplit and provide a more realistic ocean response to the peak in Andean volcanism associated with the Altiplano-Puna Volcanic Complex (APVC).

We have changed our entire modelling approach to simulate Andean volcanic eruptions in cGENIE. In our new simulations, we replicate the CESM simulations to the best of cGENIE's ability, using modern boundary conditions (as per CESM) and similar ash plume flux (as per HYSPLIT). First and foremost, we simulate the response of the marine carbon cycle to 75-yr pulses of ash over 20 kyr to evaluate the millennial-scale influence of ocean fertilization on pCO2. Then, we run three sensitivity simulations to test the impact of (1) increased ash pulse frequency, (2) increased ash pulse magnitude, (3) increased dust input. Each of these demonstrate that CO2 can be drawn down further when volcanic activity temporarily intensifies during the main phases of the APVC.

I think the outcome of this study would be way larger would the authors decide to focus on 1) a thorough review of Cenozoic cooling trends and processes - which they did very well , ***2) refocus on reviewing volcanism contribution to climate cooling, including the effect of Andes on the Miocene or on ocean fertilization during the Neogene. I think the point 2) is close to what is done here and might explain the choice of (some of the ?) cGENIE experiments****. This is indeed never clear why the authors chose to test one of the mechanisms*

rather than the others and whether it related to full Cenozoic trend or mostly Neogene. I understand that all the technical information are provided as supplementary material (and that in this specific journal format it would end up as method sections) but I think the main text is missing part of what is necessary to really understand what is done. This feeling might also be related to the fact that the paper focus is not well specified and buried below the review aspect. I do think that this paper as I is build should be consider a review as it introduce new modeling work which purpose are not to test all the reviewed process. Some of the processes described here (those that are fully detailed but not use to support the modeling results) could therefore mostly end up as supplementary text, which would give more space to clearly describe what has been done, which processes tested and why directly in the main text. We agree with the reviewer's concerns and have decided to follow approach 2 and refocus the paper: "Andean volcanism, ocean fertilization, marine ecosystem turnover, and global cooling in the late Miocene" as reflected by the new title. Our approach – combining proxy data correlations with climate modelling - and the goals of the paper are now clearly explained.

- l102 - 'Here we discuss various posited drivers of Cenozoic climate change and illustrate their potential impacts with complementary Earth System modeling approaches.' . This is a bit different to what is done later in the paper as authors review all the potential driver of Cenozoic pCO2 changes but only test some of them in a modeling framework. The introduction now only briefly introduces different controls on Cenozoic global cooling, but it focusses on biological control (i.e. increased nutrient inputs on primary productivity and marine ecosystems) on global climate with a focus on Andean volcanism and the late Miocene cooling.

- l113 - It is stated even before describing in details the processes affected pCO2 that "It should be noted that the goal of these idealized cGENIE experiments is not to provide a quantitative estimate of Cenozoic changes in the forcings over time but rather to demonstrate the long-term impact and direction of change of individual forcing mechanisms. In reality, we expect that Cenozoic cooling resulted from the complex interplay between several forces (43, 44)." While I think this warning is mandatory it is not very clear why the focus is on some of the processes only and how did the authors decide which processes to focus on or not. Because the paper continue with a full literature review on various kind of processes I would suggest this information arrive after reviewing mechanisms and comes with an explanation on why they made such choices. The text has been significantly revised, and this sentence has been removed. We have now focused the paper on the effect of increased nutrient inputs from the APVC on primary productivity, CO2 and T.

- l261-382 - while the previous paragraph incorporates at least some Cenozoic literature, the authors suddenly shift the focus much more on the Miocene. This make sense as those mechanisms are those relevant for the 'Andean volcanism' part that just come later but it makes the previous paragraphs on weathering and tectonics sounding to broad and a bit out of context. The current manuscript focuses on the late Miocene and hence we now only provide a brief description of other mechanisms suggested to explain Cenozoic global cooling.

- line 410 the authors briefly state they simulated particle dispersion but it is not clear that the setup is modern climate, which might have slightly different winds or ocean circulation patterns than the late Miocene. It might not be a problem as the goal is to demonstrate the effect of volcanic ashes on the productivity but this has to be stated to avoid confusion.
We have now revised the text to clearly state that (lines 329-335): “A series of ash plumes spanning different present-day seasonal atmospheric circulation patterns were simulated with Hysplit” and on the supplementary that: “We ran particle dispersion simulations with Hysplit 5.3.0 to derive the present-day spatial distribution of ash deposition from volcanic eruptions in the Puna-Altiplano region."

Relative to the point 1), the introduction mostly focused on the Miocene, Andes volcanism and the fertilization effect on ocean productivity, but the full first half of the paper then focus on reviewing the global carbon cycle components and introduce experiments testing only some processes in isolation. This is really disturbing and contribute to the focus of the paper getting lost pretty quickly. As a reader I got totally lost in that as I was convince from title to line 99 that I would read work on ‘Andes and late Miocene cooling’. We have reorganized the paper to focus on the late Miocene cooling, and we only provide a short summary of possible mechanisms suggested for Cenozoic global cooling.

The authors suggest a link between the Andes volcanism and the LMBB / LMCIS and make the link between SiO2 volcanic input and increase in diatoms. However, figure 3 seems to show the diatom abundance and biogenic opal is increasing after the LM cooling and LMBB. Could the authors explain how they would reconcile both records? We have now revised Figure 3 to include global diatom and opal abundances (Figure 3B) and opal abundance for specific sites 1092 and 704 from the Southern Ocean. Overall, these data match well with deep ocean phosphate concentrations and with the timing of APVC ignimbrite eruptions. We have added this discussion to the text (lines 228-291): “An increase in marine productivity in the late Miocene is also supported by deep ocean phosphate concentrations, diatom and opal abundance (Figure 3B) and coincides with the LMBB suggesting that the feedback between volcanism and marine productivity may operate at the million years scale affecting the global carbon cycle and late Miocene cooling.”

L. 35 ‘ with a focus on the contribution of the Andes to the southern oceans over the last ~20 Myr.’ —> Late Miocene ? We have revised this.

L 69 ‘Columbia River Basalts and the Miocene Climatic Optimum; (18),’ —> the closing parenthesis is lacking here. We have removed this discussion since our focus is now on the late Miocene.

l. 218 - “The magnitude and timing of these variations are purported to explain much of the temperature decline from the notably warm early-to-middle Miocene, when global temperatures exceeded present levels by approximately 10–12°C.” What is the reference for this? We now focus our study and discussion on the late Miocene.

L. 400 - “The west coast of South America has been a region of major upwelling region since

the start of the Cenozoic (160) and since at least the Miocene (161)" —> Since at least the Miocene is redundant, as it is part of the Cenozoic. I suggest the authors change for 'The west coast of South America has been a region of major upwelling region since the start of the Cenozoic (160, 161)' We have revised this.

l. 436 - 439. Please provide the references for HYSPLIT and CESM2.1.5. References are now provided in the main text and more information are also provided in the method and supplementary sections.

l. 465 - 467 The authors states that "This experiment indicates that most of the ash from the Puna-Altiplano region is transported to the South Atlantic by prevailing westerly winds. The resulting flux of ash deposited over the ocean extends eastward from the South Atlantic to the southern Indian Ocean (Figure 5A)." I might have missed something but how do they relates this patterns with the Pacific signal they introduce l. 419 to 429 ? We have revised the text as following (lines 175-182): "The Hysplit simulation indicates that most of the ash from the APVC (Puna-Altiplano region) is transported to the South Atlantic (Figure 4A) by the prevailing sub-tropical high-elevation westerly winds (STJ; Figure 2). The prevailing sub-polar low-elevation westerly winds (SWW; Figure 2) transport the ash aerosols further east beyond the South Atlantic into the southern Indian Ocean (Figure 4A). Although most volcanic ash emitted from the APVC is dispersed eastward, the proximity of this volcanic source to northern Chile ensures that a substantial fraction is also deposited locally and directly offshore into the Pacific Ocean (Figure 4A)."

Figure 1: The arrows direction are difficult to catch: for example the orange to pink seems to be causation arrows but the blue to maroon would be reverse (degassing to spreading rate). I suggest the authors to change the arrow direction depending on how the link goes. We have replaced Figure 1 and our revised Figure 3 showing proxy data now has clear arrows consistent with the direction of change.

REVIEWER 2

This paper describes mechanisms driving global cooling over million-year timescales and utilizes climate system models to explore these different mechanisms, with a focus on the role of Andes volcanism on global cooling via ocean fertilization. Understanding the mechanisms of global cooling and the interactions between carbon cycling and climate are of great interest in paleoclimate research. This study includes a comprehensive review of mechanisms driving cooling and combines a high-resolution CESM model with an intermediate complexity model to study how short-term biogeochemical processes such as the marine biological pump may influence carbon sequestration on longer timescales. The authors conclude that the Late Miocene Cooling was driven by enhanced diatom productivity in the Southern Ocean (which draws down atmospheric CO2), stimulated by nutrient input from Andean volcanic eruptions. Overall, this research provides a unique approach to better understanding mechanisms for global cooling and should be published in Earth and Environment Communications after some clarifications and restructuring is made.

The first part of the paper describes factors controlling Earth's carbon cycling and global climate including weathering, tectonics, ocean fertilization, and the biological pump. The second part of the paper then uses modeling to provide perspective on the role of Andean volcanism on the ocean fertilization mechanism. As currently written, it is unclear what is review and what is the author's contribution (modeling component). A clear statement that this is the aim of the paper would be useful somewhere in the introduction. We have revised the paper to remove the review of drivers of Cenozoic global cooling, and we have focused the paper on the role of Andean volcanisms on ocean fertilization and the late Miocene cooling. The aim of the paper is now clear.

Additionally, the review of mechanisms for cooling could benefit from subheadings. Throughout this section, careful attention to topic sentences, and only including relevant information related to these topic sentences would make it much easier to follow. In general, the link between the information given and its relevance to atmospheric CO2 or role in the global cooling process needs to be clearer in some places (see line by line comments below).
It would also be useful to include some details of the model approach and methodology in the main text as this is a major component of what the authors are presenting. Although a good job is done explaining the limitations of the model approach, information about the approach is required up front for the reader to assess the validity of the results. See comment above. We have also run new modeling simulations with cGene and discussion about the role of Andean dust-ash inputs on ocean fertilization and CO2 drawdown.

Based on modeling evidence, increased Andean volcanism is argued as an important mechanism driving global cooling during the LMC. However, when compared to paleoclimate data, there is a clear lag between peak volcanism and changes in diatom productivity and extinction rates (Figure 4). However, there does seem to be more similarity between the Andean volcanism histogram and d13C timeseries, which is a primary data set indicating changes in global carbon cycling. Furthermore, it would be helpful to directly compare this alongside SST records in Figure 4. The time lag between these systems needs to be explored and explained in the text (globally complex system, perhaps other volcanic regions at play?). We appreciate the reviewer's thoughtful observations regarding the temporal relationships among Andean volcanism, biotic responses, and global climate proxies. In the revised manuscript, we have expanded our discussion on temporal and causal relationships to clarify how they fit within expectations for a globally integrated climate system. The reviewer notes that the peak in Andean volcanism precedes the major increase in diatom occurrences and the marked rise in extinction rates. However, we point out that a significant increase in diatom abundance matches the peak in Andean volcanism. Offsets between increases in diatom abundance and Andean volcanism likely reflects the time required for volcanic forcing to propagate through the coupled climate–ocean system. It may also relate to ecological thresholds that must be crossed before environmental perturbations affect higher trophic-level consumers (e.g., whales, sharks, seals). Although extinction rates show a modest increase near the peak in volcanism, they continue to rise throughout the sustained interval of elevated volcanic activity, reaching their maximum in the Pliocene before declining in the Pleistocene. These patterns suggest that impacts on higher trophic-level consumers were cumulative and tied largely to indirect consequences of

volcanism, including progressive loss of coastal habitats as global cooling and ice-sheet expansion drove sea-level fall. Thus, while the immediate ecological effects of volcanism appear limited, the combined influence of repeated climatic and oceanographic perturbations ultimately exerted substantial pressure on marine species throughout this interval.
We have revised the text as following (lines 144-156) to include data from the Southern Ocean: "Diatom abundance fluctuates through the Miocene but rises sharply in the late Miocene (Figure 3B). Notably, records from IODP Sites 1092 and 702 in the Southern Ocean show pronounced productivity spikes at ~6 Ma and ~4.5 Ma that coincide with major ignimbrite eruptions of the APVC (17). Additional indicators of heightened marine productivity include increased opal accumulation and elevated deep-ocean phosphate concentrations, which also intensify during this interval (Figure 3B). Baleen whale body size begins to increase around ~7 Ma (Figure 3C), while extinction rates show a modest rise during the peak in volcanism and continue to climb throughout the protracted phase of elevated activity, reaching a maximum in the Pliocene before declining in the Pleistocene (Figure 3C). These patterns suggest cumulative marine ecosystem impacts driven largely by indirect consequences of volcanism—most notably the progressive loss of coastal habitats as global cooling and ice-sheet expansion lowered sea level (43). Such repeated oceanographic and environmental perturbations ultimately imposed substantial pressure on marine species."
We have also revised Figure 3 to add SST data (Figure 3D).

Although considering the entire Cenozoic is relevant for considering the processes driving global cooling on multi-million year timescales, I am unsure what the value is of including this entire time period, especially when the majority of paleoclimate data discussed in Figures 3 and 4 only includes data from the Early Miocene to present. I wonder if the discussion could instead be focused on the past 20 million years of Earth's history? We now focus on the Late Miocene.

Towards the end of the paper where the authors discuss the role of Andean volcanism on Southern Ocean productivity, it would be very useful to have a schematic including the Andes and areas of the Southern Ocean illustrating the regional processes that influence carbon cycling between different reservoirs. Where possible, label the geographic locations mentioned in the text. Having a figure like this would be a great way for readers to visualize the processes occurring. Since we are not providing a review of all other possible drivers of Cenozoic cooling, we have decided that a figure showing different carbon reservoirs is no longer relevant. However, we have now included a new Figure 2 showing the wind patterns over the Andes, the location of the APVC, dust activity and the wind directions. Figure 4 now shows how ash/dust from the Andes into the Southern Ocean.

Line-by-line comments:
1: Consider "Neogene" instead of "Cenozoic" and "impacts of Andean volcanism" *We have revised this to focus on the late Miocene.*
21: replace "geological" with "million-year" Revised.
30: add "which alter atmospheric CO2" at the end of the sentence to make the link between climate and atmospheric CO2. Revised.
31: What is meant by "longer timescales?" Revised – we now specify "millions of years".

32: addition of nutrients by which processes? Weathering? Revised (lines 46-51): "South America is the site of some of the highest mountains and largest volcanoes on Earth, the Andes, and one of the major sources of nutrients, via rivers and dust, to the world's oceans (1-3)fueling both the Humboldt Current and the Southern Ocean—key regions for marine productivity, nutrient cycling, and CO drawdown. Andean volcanoes also contribute to 1 % of global CO uptake via silicate weathering (4), but their impacts on ocean fertilization, marine ecosystems, and global cooling remain largely unexplored."

35: Rather than southern oceans throughout, refer to the whole ocean as the "Southern Ocean". If you need to refer to specific areas, you can say "Atlantic sector of the Southern Ocean". Revised.

39: Add comma after "cooling" Revised.

43: "causing" rather than "sparking"? Revised

48: replace "global climate change" with "cooling" since this is the focus? Revised.

49-53: make one sentence? Revised.

52: Also figure 2. We have replaced the original Figure 2 and updated all references in the text.

60: Specify when the Miocene was. Revised

62: Specify when the MCO was. We have removed reference to the MCO since the focus now is on the LMC.

66-68: Can you add a sentence to explain why these might have been important for understanding carbon cycling? Also the Monterey Excursion is shown on the figure but not in the text. This might be something you want to include to illustrate the coupling between climate/ice sheet volume/sea level during the MCO and MMCT. We now refer only briefly to the Late Miocene Biogenic Bloom and the associated carbon-isotope shift in the introduction, but we do not discuss them further because the LMBB spans a long interval (~9–3.5 Ma) and reflects a complex interplay of processes that likely contributed to these biogeochemical changes.

68: CRB associated with MCO in general- not sure if there are other instances in the Miocene of this association. Could also talk about the mid-ocean ridge spreading rate hypothesis here. We have revised the text and we include reference to Herbert et al. (2022) hypothesis.

76: Increase in atmospheric CO2? Specify what is meant by "shorter timescales". Revised.

81: "nutrient-driven". Also add "(alkalinity)" at the end of the sentence? This concept may warrant more explanation. Revised. We have also removed the discussion of carbonates in this version since we mostly focus on diatoms.

86: List the macro and micronutrients here that are supplied by volcanic ash and glacial dust. Revised.

91: state that ignimbrites are silica-rich? Would be really useful to have a map/schematic here to refer to the geography and relevant carbon cycle processes. We have added a new Figure 2.

95: Reverse intensified Hadley circulation and aridity since the first contributes to the second. Revised.

97: Unclear form this sentence that there is a modeling component of this study. Revised.

105: First mention of calcium carbonate weathering and how this links to global carbon cycling. Revised and removed.

116: replace "forces" with "forcing mechanisms". Revised.

123: Make this a subheading, work on topic sentences in this section. The paper has been

majorly revised.
125: climate refers specifically to temperature and precipitation? How are these accounted for in the models? Large drop in temp and precipitation after the MMCT. We have removed discussion of the MMCT and we now better explain our modeling approach with CESM, Hysplit and CGene.
130: explain more how these things link to CO2 partitioning between different systems. We now focus the paper on ocean fertilization by dust-ash inputs and the response of the biological pump, CO_2 and T.
162: The distinction between what is a review and what are new ideas out forth by modeling results very unclear in this section. Needs to be clear separation of the two. The paper has been largely revised to remove the review component and focus on the role of Andean volcanism on ocean fertilization, biological pump, carbon cycle and the late Miocene cooling.
167: I would expect that cooling would result in more intense latitudinal temperature gradients resulting in stronger winds, upwelling, and increased vertical mixing of nutrients. Could it also be that cooler ocean temperatures result in decreased microbial remineralization of POC in the water column? We have removed discussion of carbonates since this is no longer a review paper.
188: Can this proxy data be added to the paleoclimate data figure? We have removed discussion of weathering since this is no longer a review paper.
196: Ocean circulation mentioned here but then not elaborated on again. Important for latitudinal temp gradients which drives global atmospheric circulation? We have removed this discussion since this is no longer a review paper.
204: cite. Revised.
220: cite. We have removed discussion of the early to middle Miocene
226: Again, switch from review to modeling results is hard to follow. We have largely revised the manuscript.
332: pCO2 not shown on the figure. We have now revised Figure 1 to include pCO2.
348-368: Unclear how this information is relevant to carbon cycling. We now provide a better discussion of the links between primary productivity, whale evolution and carbon cycle (lines 129-137): “Whales shape marine ecosystems today through nutrient recycling, including iron-rich feces that stimulate phytoplankton blooms (37), as well as through prey consumption and other trophic interactions (38). Upon death, their carcasses serve as significant carbon sinks, potentially impacting climate regulation through CO2 sequestration (39). These bottom-up and top-down connections between whales and diatoms have been present for much of the Cenozoic but they may have been intensified by enhanced ocean fertilization during the late Miocene. The increase in whale size enhances their ability to transport carbon and energy across ocean basins through long-distance migrations, thereby strengthening their positive influence on the global carbon cycle (39). Lines 271-277: “Whales play a significant role in the global carbon cycle because of their size, which enables long-distance migration and thereby the export of carbon and energy from high-latitude nutrient-rich regions in the Southern and Arctic Oceans to subtropical regions (39). Whale death assemblages along the coast of Chile are preserved in Upper Miocene marginal-marine deposits interbedded with ash layers derived from the Central Andes, supporting a link between whale mortality and ash-driven fertilization that stimulated primary productivity and triggered harmful algal blooms (24, 51).”

70: When did this change occur? We have now added this information to Figure 3C and discuss temporal relationships in the text.

385: Again would be great to have a map and schematic. We have now added a revised Figure 2 indicating the location of the APVC.

393: Add “input into the ocean” after “silica”. Revised.

394: If Andean ocean fertilization is a driver, why is there a lag between peak volcanic activity and the LMC? Why is the diatom peak and increase in extinction rate after this?
The peak in Andean volcanism coincides with the LMC. We address this above.

What is the relationship of all of this to the d13C curve? We have added the following sentences: lines 64-68: “The Late Miocene Carbon Isotope Shift (LMCIS) (7.6 - 6.6 Ma) has been proposed to be a result of increased marine productivity associated with ocean fertilization (10) and overlaps with the late Miocene cooling (Figure 3A). The Late Miocene Biogenic Bloom (LMBB) has been associated with increased aeolian dust delivery to the oceans, leading to elevated nutrient supply and enhanced marine productivity (8).”

420: Again, why the lag? We now discuss these temporal relationships in the text (see above).

425: A lag of about 1 million years. See above.

500: Is there any sedimentary evidence from deep ocean drill cores to suggest that there was an increase in biogenic silica accumulation at this time? Maybe look just in the Southern Ocean. We have added diatom data from the Southern Ocean and revised the text as follow (lines 204-209): “This enhanced soft tissue export is dominated by diatom productivity (as supported by sites IODP 704 and 1092; Figure 3B), as is evident from enhanced 100-m-depth biological opal (SiO_2) export, which closely follows the cycles of volcanic ash-derived major and micronutrient inputs (e.g. N, P, Si and Fe) (Figure 5A-D). Diatoms clearly outcompete calcifiers at centennial scales as shown by the marked cumulative reduction in carbonate ($CaCO_3$) export (Figure 5D).”

511: Great discussion of the limitations. I wonder how this might time into the time lag questions raised above. We now better discuss the issue related to the time resolution of the models compared to millions of years time scale we are proposing for Andean volcanism.

519: This overall is written well and seems like a summary of above. Concepts very clear here. Still not super clear on the main messages of the paper. The paper has been largely rewritten and refocused to take care of this.

554: These data need to be discussed earlier. Carbon isotopes one of the primary lines of evidence for long-term changes in global carbon cycling. Revised.

569: How does whale preservation indicate productivity? Frequence of whale fossil occurrences? What about a major change in sea level which caused them all to become beached in one place? We have now revised the text to include the following (lines 275-281): "Whale death assemblages along the coast of Chile are preserved in Upper Miocene marginal-marine deposits interbedded with ash layers derived from the Central Andes, supporting a link between whale mortality and ash-driven fertilization that stimulated primary productivity and triggered harmful algal blooms (24, 51). This hypothesis is supported by multiproxy data which show a strong link between intensified APVC volcanism at ~7 Ma and ~4 Ma, marine mammal reorganizations (e.g., increase in whale size; (41)), enhanced marine productivity (9), and changes in Southern Ocean biogeochemistry (52) during the late Miocene (Figure 3)."

580: This is a great point, maybe bring into the introduction to justify the approach of the study?

Figure 1

What about temperature of deep ocean controlling CO2 solubility? Large decrease in deep ocean temps across the MMCT would have changed this. We have revised Figure 1 to focus on the late Miocene.

Figure 2

Can remove top plot? We have revised Figure 2 and included the δO and δC records in Figure 1 and focused on the late Miocene.

Use bars to show the duration of climate events eg. MCO, add in MMCT as well. Remove anything that isn't discussed in text eg. Monterey Event
Add an arrow on the d18O axis to indicate which direction is ice growth/deep ocean cooling
E needs citation
Add CCD depth or carbonate preservation through time? Is there data for these or have they been modelled?
We have now revised all figures. Figures 1 and 2 now include the relevant data and focus on the late Miocene. We have included bars to show temporal extent of specific events and use arrows in Figure 3 to match the direction of specific changes.

Figure 3
Difficult to understand what this is without more detail in the main text
Increased Ca weathering? Silicates? CaCo3?
Would be helpful to put % change on the x-axis for each modeling output
Specify in caption what long-term means (I think million year?)
State the boundary conditions used for the model briefly ie. the LMC period
This Figure has been replaced by new Figure 6.

Figure 4

Andean volcanic activity seems to follow the d13C curve closely. There is a significant lag between volcanic activity diatom silica export, and increase in extinction rate that needs to be

explained. Unsure if this is strong support of the importance of the ocean fertilization mechanism for pCO2 drawdown. Global cooling records should also be shown on this plot.
We have revised Figure 4 and provided a better discussion of the temporal relationships between proxy records and global cooling. The new section (lines 264-324): “Andean volcanism, Marine ecosystem turnover, Global Carbon cycle and the Late Miocene cooling”.

Figure 5
Clearly an increase in diatom productivity after volcanism on a short timescale. But how does this translate to burial of OC on much longer timescales? See above.

Supplementary Table
Consider including in main text as it is quite helpful to understand what was being tested with the model
Because we have changed out modeling approach and focus this table is no longer relevant.

Please find below detailed responses to reviewers' comments in blue.

REVIEWERS' COMMENTS:
Reviewer #1 (Remarks to the Author):

The manuscript by Carrapa and collaborators has been now refocused on Andean volcanism and how it could have contributed to change in productivity, CO2 uptake and marine ecosystem during the Late Miocene. The overall message is thus now way clearer and the paper reads better.
The authors cleverly re-designed the simulations framework so that cGENIE simulations are now an extension of the CESM runs, both enabling to respectively understand the millennial and centennial timescale response of productivity and CO2 uptake to volcanic dust emission. The authors then used simulation results and data compilation to discuss potential contribution of Andean volcanism to the Late Miocene cooling and the coeval marine ecology shift.
The methodology and modeling results are convincing to me and the authors took into account comments from previous round of review. The introduction now gives more detailed information on the Late Miocene, volcanism and productivity, which helps understanding the story. The data compilation part of the work is also now better emphasized, and the authors also incorporated more details on the modeling methodology (that were previously in the supplementary material), which I think is great. They also conducted several sensitive experiments with the models to ensure their results are robust.

I really appreciate that the conclusions are not oversold and limitation of the numerical tools are clearly stated. This paper is a nice contribution toward better understanding the driving factor of Late Miocene climate change. So congrats, and I really look forward this paper to be publish and to see further work on this topic.

Minor comments:
Authors use both « late Miocene » (eg. l. 53) and « Late Miocene » (eg. l. 52) across the paper. It would be good to homogenized and keep one or the other.

We have revised to "Late Miocene" everywhere.

L. 67. « The Late Miocene Biogenic Bloom ». I suggest the authors put age interval for this event as they do it for LMCIS. We have now specified time interval in line 73.

- Authors used both « HYSPLIT » and « Hysplit ». I suggest to keep with « HYSPLIT » across the paper. We have revised to HYSPLIT everywhere.

Anta-Clarisse Sarr

Reviewer #2 (Remarks to the Author):

General comments
Overall, this version of the manuscript is improved as it now reads as an independent modeling and data comparison study than as a review and new study. The authors have addressed some of the limitations of the study clearly and include information about processes which may not be included in the model study. The restructuring has made the aim and outcome of the study more clear. There are still some minor edits that should be made to correct inconsistencies (for example, consistent use of capitalization of Late Miocene, correct labeling of panels in Fig. 2, order of figures, clear topic sentences, instances of repetition).

Line-by-line comments:
2: late should be capitalized, check for consistency throughout document and figures. Revised
25: When? Revised.

33: Clarify the link to the ocean, deposition of nutrient-rich ash in the ocean. Humboldt current should be indicated on the map. The Humboldt current has been added to Figure 2.
38: Put data compilation first as that is what is presented here. Revised
39: add timescales after millennial. Revised
42: increased carbon export to the sediment? Make the mechanism by which this happens clear here. We revised the text as following (lines 41-43): “Our results indicate that enhanced nutrient supply from episodic Andean volcanic ash to the Southern Ocean enhanced primary productivity and CO2 sequestration in the deep ocean”.
46: Need heading here, Introduction? Is mountain mention relevant? Sentence is quite long consider shortening or splitting into two. We followed Nat Comm Earth and Environment formatting but we have split the sentence into two.
49: citation? We added citation.
51: New paragraph here? New topic – we started a new paragraph and slightly revised the opening sentence to (lines 48-50): “South America is home to one of the highest mountain ranges and largest volcanic systems on Earth, the Andes (1), and represents a major sources of nutrients to the world’s oceans via both riverine and atmospheric transport (2-4).”
57: remove “in” after changing. Unclear especially to unfamiliar readers what the link is between these things listed and climate. Revised to (lines 59-62): “Mechanisms of global cooling include enhanced silicate weathering linked to mountain building,

changes in ocean circulation, upwelling, and seafloor spreading rates, intensification of monsoonal winds, and greater aeolian nutrient fluxes from expanding continental deserts (8, 11-15).”
61: briefly give some examples of limiting nutrients. We have added: “(e.g., Fe, P)” (now line 65).
64: Make this the topic sentence? We have revised this sentence as following (lines 65-68): “Primary production in marine ecosystems can be stimulated by increasing the supply of limiting nutrients (e.g., phosphorus and iron), for example through dust and ash deposition, potentially lowering atmospheric CO□ over a range of timescales (16), provided that the resulting increase in organic carbon is ultimately sequestered in the deep ocean and sediments (3).”.
65: evidence for increased marine productivity is the LMBB? Merge these sentences for clarification. The text has been revised as following (lines 70-73): “Increased aeolian dust delivery to the oceans has been proposed to enhance nutrient supply and stimulate marine productivity, as evidenced by the Late Miocene Biogenic Bloom (LMBB; ~7.5–3.5 Ma), which overlaps with the Late Miocene Cooling (LMC) (Figure 3A) (9) (Figure 1).”

79: from rather than at. Revised
80: add jet, or spell out acronym here. Revised.
101: and cooling? Revised to add “cooling”.
101: repetition here. We removed the following sentence (Multiproxy correlations support a link between increased Andean volcanism, elevated marine productivity, and changes in Southern Ocean biogeochemistry during the Late Miocene) and integrated this later in this sentence (117-121): “Combined with proxy records, our results suggest that sustained nutrient inputs from Andean volcanic eruptions enhanced marine productivity and altered Southern Ocean biogeochemistry, potentially contributing to longer-term atmospheric CO□ decline and, in turn, to Late Miocene cooling.”
103: Add to investigate x we do y. Revised to (lines 108-110): “To investigate the role of fertilization from Andean volcanic ash on ocean biogeochemistry, carbon budget and global cooling we apply a hierarchical modeling approach using…”

104: Spell out these acronyms for the benefit of non-modelers. We revised text accordingly

107: also million year timescales? Not in our simulations.
115: Maybe call this a data comparison or compilation? Revised. You could add a small paragraph about what you’re doing and why (utilizing existing data) before launching into this section. We have revised the following text to better introduce our approach (104-108): “This study integrates geochronological records of Andean volcanism, proxies for marine productivity and marine vertebrates (Figure 3), ash-transport modeling, and biogeochemical Earth system simulations to quantify how Late Miocene

eruptions from the Central Andes enhanced nutrient delivery, stimulated ocean productivity and carbon export, and contributed to global cooling".

117: Why is the DIC gradient relevant? We have revised the following text (lines 125-127): The ocean's capacity to sequester carbon depends largely on the biological pump, which governs about 90% of the vertical dissolved inorganic carbon (DIC) gradient with steeper gradients reflecting stronger export production and increased deep ocean carbon storage (34). Instead maybe say or define export production? We have added "storage".
118: key producers in the Southern Ocean/regions of upwelling. Revised to (line 128): "Diatoms, key primary producers in the Southern Ocean and regions of upwelling"
124: repetition of food web from previous paragraph. We have removed repetitive text.
126: refer to figure. Revised to refer to Figure 3.
136: feedback rather than influence. Revised
147: Refer to figure. Revised.
154: citations for Antarctic ice sheet growth and/or global sea level fall? We do appreciate the reviewer's suggestion but because of limited space and the fact that this is not a review paper, we have to limit our reference to Pimiento et al., and references therein.
166: citation for SL change? See comment above.
173: capitalize HYSPLIT. Revised.
197: how was significance assessed? We revised text as following (lines 206-207): "…the model simulates a significant increase in surface diatom chlorophyll concentrations (>two-fold increase) and associated CO2 uptake (Figure 4D-E)".
209: This could also be because of carbonate dissolution as bottom water conditions changed. This is incorrect. In Figure 5, the curves we plot are anomalies, that is, we plot the results (for each variable shown) of the simulation with ash injection minus the results of the control simulation. The control simulation is configured exactly the same as the main simulation, except that for the main simulation we add ash-derived nutrient injections. In particular, the atmosphere and ocean configuration is the same, so there are no changes in the ocean temperature distribution among main and control simulations. So, the changes in opal and carbonate productivities cannot be explained in terms of changes in ocean temperatures.
270: This is largely repeated from above, consider where and how best to present this information. We have revised this whole section to remove redundances.

293: Is this the summary section? Confusing as to when one section ends and another starts without subheadings. This section is meant to discuss the role of Andean volcanism on ocean geochemistry and marine ecosystems and ultimately global carbon and climate on geological time scales. We have revised this section to remove redundances.

Figure comments
Figure 1. The 5 Ma mark is in the wrong position. Revised.
387: add (SST) in caption since this is what is listed in figure. Revised.

Figure 2
Add Humboldt Current. Revised.
399: unclear what hydrologic deficit is and how to make sense of it in figure. We explicitly define "hydrologic deficit" in the caption: "Hydrologic deficit is calculated as total evaporation minus total precipitation, which were derived from monthly ERA5-Land reanalysis data between 2011-2020". We have improved the caption of Figure 2 to clarify this: "Dust activity in South America. Dust Optical Depth (DOD, 550 nm) monthly climatology (November 2011-October 2020) is based on thermal infrared IASI-METOPA measurements and the MAPIRv4.1 algorithm. The dust extinction coefficient monthly climatology (January 2011-December 2020) is based on surface visibility data (Xi, 2021). AERONET coarse-mode, Aerosol Optical Depth (AOD, 550 nm) monthly climatology (January 2011-December 2020) is based on level 2.0 data, obtained using the spectral deconvolution algorithm. Only stations with at least 60 monthly values are included. Hydrologic deficit is calculated as total evaporation minus total precipitation, which were derived from monthly ERA5-Land reanalysis data between 2011-2020. Regions with high hydrologic deficit correspond to arid to semi-arid terrains, from which dust emissions are most probable. Zonal mean 850-hPa wind speed and direction (light blue arrows) represent the sub-polar low-altitude jet (i.e., the westerlies, SWW), while the zonal (only for longitudes within the Altiplano-Puna Volcanic Zone (APVC)) mean 200-hPa wind speed and direction (light brown arrows) represent the sub-tropical high-altitude jet (STJ). Both were calculated from a monthly ERA5 climatology (January 2011-December 2020)."

Figure 3
Labelling is inconsistent (B and C switched in caption). Revised.
425: Refer to isotopes in the same way as fig 1 (on axis). Can say benthic foraminifer isotopes and alkenone SST records. Revised.