nature portfolio

Peer Review File

Unprecedented Reversal of Oceanic Stable Carbon Isotope Ratio Depth Gradient with Continued Human Carbon Emissions

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Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality.

17th Sep 21

Dear Dr Kwon,

Your manuscript titled "Unprecedented Reversal of the Oceanic d13C Depth Gradient Due to Extreme Greenhouse Gas Emissions" has now been seen by 3 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 that fully addresses these serious concerns.

We hope you will find the reviewers' comments useful as you decide how to proceed. Should additional work allow you to address these criticisms, we would be happy to look at a substantially revised manuscript.

In the following, we list our main editorial thresholds:
• Provide compelling evidence to support the conclusions regarding the rate of change in d13C-DIC in future scenario vs. during the PETM
• Provide justification for the design of model experiment and the presentation of benthic foraminiferal d13C data (main concerns raised by Reviewer #2).
• Improve the structure and clarity of the manuscript so that it is more accessible to the reader.

However, please bear in mind that we will be reluctant to approach the reviewers again in the absence of substantial revisions.

If the revision process takes significantly longer than three months, we will be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

We are committed to providing a fair and constructive peer-review process. Please do not hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees’ comments (which should be in a separate document to any cover letter) and any completed checklist:

[link redacted]

** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

Please do not hesitate to contact me 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,

Sze Ling Ho, PhD
Communications Earth & Environment
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Clare Davis
Associate Editor
Communications Earth & Environment

EDITORIAL POLICIES AND FORMAT

If you decide to resubmit your paper, please ensure that your manuscript complies with our editorial policies and complete and upload the checklist below as a Related Manuscript file type with the revised article:

Editorial Policy Policy requirements

For your information, you can find some guidance regarding format requirements summarized on the following checklist:(https://www.nature.com/documents/commsj-phys-style-formatting-checklist-article.pdf) and formatting guide (https://www.nature.com/documents/commsj-phys-style-formatting-guide-accept.pdf).

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Review of “Unprecedented Reversal of the Oceanic d13C Depth Gradient Due to Extreme Greenhouse Gas Emissions” by Kwon et al., submitted to Communications Earth and Environment, July 2021.

Overall this is a solid paper with a detailed model and the results are well-explained. The behaviour of the model is not too surprising, and I wonder if some summary statements in the conclusions along these lines are warranted: When 13C-depleted carbon release into the atmosphere is faster than deep-ocean mixing timescales (so occuring in centuries or just one or two thousand years), the Suess effect overwhelms the biological pump and you get a d13C-depth gradient reversal. When carbon release is slower than ocean mixing timescales, the light C has time to mix to depth and so depth d13C gradients are preserved.

The above seem like a simplified, general interpretation of the model results and apply to the PETM (during which C release seems to be just on the bubble of being faster than ocean mixing timescales, so you get a short-lived d13C-depth reversal or collapse). Also in the case of the PETM, it is generally thought that deepwater circulation was "sluggish" during the Eocene greenhouse, so an somewhat slower C release (a few thousand years) could cause a d13C gradient collapse.

A couple of specific comments:

Line 468: It seems to me to be a mistake to apply no d13C correction to Acaranina and Morozovella, because we know (through stable isotope ecology studies and shell size-d13C relationships) that these genera harbored photosynthetic symbionts, which increase d13C in the calcifying microenvironment. I suggest that they assume a d13C correction from a modern photosynthetic symbiont planktic foraminifer species (such as G sacculifer) as this is more likely to be closer to the truth than simply assuming no d13C offset for these ancient species.

And this becomes complicated in the case of the PETM because those very same taxa seem to (at least at some sites) lose their photosymbionts during the PETM, according to the very new Shaw et al paper that you cite (which is now fully published so you can update the citation from "in press").

Figure 3: Can you mark the North Atlantic - Southern Ocean - North Pacific transect used in column A on the maps of column B?

Reviewer #2 (Remarks to the Author):

The manuscript by Kwon et al. appears to have multiple objectives, but the presentation leaves me unclear as to what the major or novel conclusions are meant to be. I am unclear as to whether the paper is primarily meant to evaluate how spatial patterns of DIC-13C will evolve under future RCP scenarios or to provide an interpretation of PETM-carbon injection. While I understand the motivation to use the PETM as a future analog, I don't think that presentation works successfully here, and I'm left wondering whether the authors are making an argument that the PETM changes constrain the future or that the modeling of the future constrains the interpretation of the PETM.

Beyond this confusion as to the objectives of the study, my primary criticism of the paper is that it completely misinterprets the results of Ref. (8), a study that modeled changed in vertical d13C-DIC gradients across the PETM onset using the cGENIE model. That study in fact found major delays in the propagation of the d13C anomaly to the deep ocean (it rather argued that major delays were not possible from the surface ocean to the thermocline). Here, the authors summarize that paper to say that it argued against reversals in the d13C-depth gradient. As I describe below in more detail, that is incorrect, and because that study did in fact present significant durations of reversed surface-deep gradients, this then conflicts with the narrative that this study presents original results.

I see value in the extensive ensemble for preindustrial DIC-13C conducted here, though that is also not novel (see Holden et al., 2013). However, the relegation of details to the Methods and Supplementary sections is really unfortunate. Moreover, I do not understand the choice to fix the ocean circulation response across all of the simulated experiments (rather than as a subset to provide a control). In Ref. 8, that study explicitly compared different assumptions about the degree of ocean stratification and the representation of vertical mixing in the model, but also used a 'no circulation change' control, and it would be very interesting to know how those model results compare with the model used here.

Finally, in a paper entirely about the modeled representation of ocean d13C, the treatment of model description within the text is inadequate.

(Intro)
Line 44 - this is a misleading summary of the interpretation of changing d13C gradients during the PETM onset from Ref. (8). If you view Fig. 2 in Ref (8), there is very clearly an interval of reversed d13C gradients with depth - the d13C values of the mixed layer and thermocline in that model (cGENIE) are lighter than the benthic layer. The argument in Ref. (8) is rather that there cannot be a delay of 5-15,000 years before the thermocline sees excursion values, which had been argued previously based on interpretation of single foraminiferal data from ODP Site 690. Quoting from that paper, “we note that significantly larger delays are possible before the excursion reaches the deep ocean.” In the Supplementary Table 1 referenced in the text, the delays between the surface and benthic excursion reach thousands of years (sometimes greater than 5 kyr), depending on the degree of warming and hence stratification as well as the mixing parameterization used. Hence, I take issue with the author’s statement that ‘It is fair to say that the early geochemical evolution of the vertical d13C-DIC gradient remains uncertain.’ That statement effectively ignores the results presented in Ref. 8.

Line 65 - I am confused as to whether the objective of this paper is to evaluate the PETM or to evaluate the potential for d13C gradient reversals due to various RCP scenarios. The connection between the two is not well established. On the one hand, you might indicate that you are using the pre-industrial to future change to test the model behavior specifically in order to evaluate PETM records, but that is not really the way the introduction reads. I’m left struggling a bit as to the principle objective of this study. This is compounded by the title, which is ambiguous as to whether it refers to the future, the PETM, or just warming generically.

(Methods)
Line 314 - I think it is inappropriate to provide only a reference, and no summary model description of the treatment of carbon isotopes in the model used, since this is the crux of this entire study. The level of detail provided in Methods does not need to be to the level of detail in the Supplement, but it is absolutely possible to summarize in a paragraph. The ensemble alone could warrant its own manuscript, and the very short treatment in the main text feels incomplete.

Line 345 - I am trying to follow the reasoning behind the two methodologies described for estimating pre-industrial d13C-DIC but finding the detail insufficient. The authors first describe using a large ensemble to generate both a series of pre-industrial steady states as well as various trajectories from pre-industrial to modern. However, because of the degree of variation among the simulated pre-industrial steady state d13C-DIC, the authors do not use any of these simulations and instead use an observational compilation and add a spatially uniform(?) correction based on the Suess effect, hence retaining modern patterns but not absolute values for the pre-industrial DIC-13C pattern? So, does this method provide a distribution that is different from any one of 1400 ensemble members? Or significantly different from the ensemble mean?

Line 384 - when “we choose a model configuration from the Monte Carlo experiment,” what configuration was this? How did you choose? When you say you ‘fix the ocean circulation state,’ what does this mean explicitly?

Line 414 - To me, it’s really not helpful to describe these idealized sensitivity experiments as PETM-like. If nothing about the model is configured for the late Paleocene, then why is this different than just considering these as hypothetical future experiments? Also, over these long timescales, I’m even more perplexed by the choice to only evaluate experiments where the circulation is fixed throughout the simulation. Why bother simulating thousands of years of model time and ignoring the fact that the model will predict a change in the circulation state in response to warming over these timescales? Rather, I can see using ‘circulation-fixed’ experiments as a control against which to constrain the impact of changes in circulation.

Line 448 - How can you generically use all epifaunal benthic values to represent d13C values below 74 m? Do you not discriminate further by depth? Why do you even need the last 65 Myr of d13C data? (I don’t see what Fig. 6a adds to the manuscript). It is confusing to first suggest that you use data from a compilation (Ref. 30) and then indicate you selected two sites only as if those are the only two open ocean PETM sites. Moreover, if you provide both the original data sources and the references for the age models used for those two sites, what does it even mean to say you used data from the Shaw compilation?

(Results)
Line 90 - what about near regions of deepwater formation?
Line 111 - what do you mean by ‘revealing higher spatial gradients’ Is this simply to point out that a higher d13C for the preindustrial surface necessarily then means larger d13C gradients with depth in the pre-industrial compared to modern? Or are you saying something about horizontal gradients in the surface?

Line 123/Line 155 - somewhere you should formally introduce what you mean by perturbation ratio - you use this to refer to multiple different quantities.

Line 212 - again, this description of the findings of Ref. 8 is wrong and misleading.

Line 220 - what is the motivation of using different ocean ventilation states? Can you be explicit about what you are testing with these model experiments and what you expect to find?

Line 252 - the presentation of the novelty of these experiments is misleading. There is no reference of the ensemble evaluation of modeled pre-industrial to modern DIC-13C by Holden et al. (2013), and the cGENIE model has been used multiple times in evaluating d13C in response to CO2 injection (both future and past). See Norris et al., (2013), Kirtland Turner and Ridgwell, 2016. The detailed evaluation of future spatial patterns in d13C specifically under RCP scenarios is, I believe, novel, but then I think a major missing factor is any exploration of how modeled changes in ocean circulation impact these distributions.

Refs:
Holden, P.B., Edwards, N.R., Müller, S.A., Oliver, K.I.C., Death, R.M. and Ridgwell, A., 2013. Controls on the spatial distribution of oceanic δ 13 C DIC. Biogeosciences, 10(3), pp.1815-1833.

Norris, R.D., Turner, S.K., Hull, P.M. and Ridgwell, A., 2013. Marine ecosystem responses to Cenozoic global change. Science, 341(6145), pp.492-498.

Turner, S.K. and Ridgwell, A., 2016. Development of a novel empirical framework for interpreting geological carbon isotope excursions, with implications for the rate of carbon injection across the PETM. Earth and Planetary Science Letters, 435, pp.1-13.

Reviewer #3 (Remarks to the Author):

Dear Eun Young Kwon and colleagues,

Thank you for your interesting contribution to the discussion of the interpretation of marine d13C-records. Your work can be of interest to a wide audience ranging from contemporary/future carbon isotope and carbon cycle scientists, to paleoclimatologists interested in the drivers of the PETM. Your goal has been to use your projected changes in the marine d13C gradient as an analogue for the PETM d13C changes. Your results lead to your main claims that 1) the PETM elimination/reversal of the d13C gradient was driven by fast (land biosphere) release of CO2 (and its accompanying low-d13C) and 2) that the projected 21st century d13C gradient changes are likely to lead to elimination/reversal of the gradient as compared to pre-industrial times and are unprecedented over the past 65 million years. You have run your model in different setups to explore drivers/mechanisms, quantify changes and find analogues in the projected gradients for the PETM gradients. I think your results are novel and interesting, but I have several comments. Specifically, quite many conclusions are drawn and supported by quite many different methods (different model runs/datasets), which makes it difficult for the reader to find your 'take home message' or follow the main storyline. Actually, writing two separate papers may help communicate your work. I have focused on the non-PETM part of your manuscript as this is where my expertise lies. In any case I start the review document with some general comments, followed by more specific comments, then comments to the supplement and finally some technical comments. Find it attached.

Thank you for your time considering my feedback,

Best regards,

Anne Morée

General comments

1. From the abstract and the title, I got the impression that this paper was mostly about a present/future reversal of the marine d13C gradient. However, you also want to make the connection to the PETM. You state in the abstract: a) from model projections we find that a reversal is expected (not surprising), b) this reversal is driven by reducing surface d13C-DIC due to low-d13C fossil fuel emissions and facilitated by enhanced equilibration under high DIC conditions (interesting analysis), c) such reversals are also present in the PETM (new), d) The PETM change was however much slower (this comes a bit suddenly and with little backup).

   Your statement on the unprecedented 65 million years can only be made if you have a decadal resolution of d13C gradients over these 65 million years, such that you can compare the records to the contemporary Suess-effect which occurs over the course of a few decades. Can you really do that? Is the PETM CO_2 release and its corresponding d13C signature likely comparable to the future one? Does the uncertainty of the PETM records allow for the conclusion that the gradient reversed? Also, you could make clearer that your record (Fig.6) is 65 million years such that you can make the 65-Myr statement - beyond the PETM.

2. Text structure: The Results section now has 4 very separated subsections. The PETM is not even mentioned in the first 3. This is an example of where the reader may forget your overarching storyline of the PETM gradient reversal: How do each of these sections contribute to your storyline and main points? Why is quantifying the Suess effect relevant for your PETM conclusions/analysis? At least I need some more help here to be guided through your steps.

3. I soon lost the overview over what experiments and sensitivity analysis were performed. Different circulation states were explored (10 according to the supplement?) as well as 1400 monte carlo experiments (supplement caption figure 1), as well as a ‘full model’ setup, and a fixed.CO2 and fixed.ratio setup. And then the PETM 4 permil and 5 permil simulations. These are outlined in the methods, but still I find it hard to get an overview. Could youat least rewrite/add to L 60-68, summarizing the goals and main characteristics of all your different methods/approaches?

4. Some discussion is missing: What other explanations have been given by the literature of the PETM gradient elimination/reversal? The range of d13C POC/DIC/DOC you explore in your Monte Carlo experiments is quite limited in my opinion (Supplementary table 1), variations in the abundance of C3 versus C4 plants (~-25 permil vs. -12 permil respired CO_2) must have driven a much larger range? Curiosity:

to what extent has the gradient elimination/reversal been observed yet in the contemporary ocean?

5. I think your study should be able to state how fast a release of a certain amount of CO_2 (with a certain d13C imprint, etc.) needs to be in order to reverse marine d13C gradients (this would depend on the region though). I think this result is there 'in between the lines/in the figures' especially in the results section on the PETM, but highlighting this individually could make your study useful for a broader audience.

6. Are you able to reduce the number of figures to fewer key figures to focus your storyline? Is it for example needed to have Fig 2c-f in the main text? Could you highlight e.g. only 2 or even one RCP scenario for clarity?

7. Availability model/data: Will you archive your model / model output / analysis scripts and other data to make your figures in an openly accessible repository?

Specific Comments

L35 The 10000 GtC PETM release does not really compare to the RCP scenarios, which give an anthropogenic influx of about 500-2000 GtC until 2100? Why not release the 'actual' CO_2 amount to the model?

Throughout the text: you mention 'surface' and 'subsurface' regularly, please define and repeat your definition for clarity. What living depths do your planktic and benthic forams represent, and do your definitions of 'surface' and 'subsurface' favor comparison with those? E.g. L141-142 what is exactly done here?

L84, L103-109 See the recent paper by Liu et al. (2021) who quantify an underestimation of the Eide et al. (2017) Suess effect per ocean basin.

L94-100 Another noticeable feature of Fig 3b is the weakening of the latitudinal contrasts with increasing RCP scenario. Here, another point should be discussed I think: the perturbation ratio is also affected by the d13C disequilibrium. Equilibration with a d13Catm of -6.5 permil actually increases surface d13C_DIC in low-temperature low-d13C_DIC regions such as the upwelling areas due to the negative disequilibrium, i.e. d13C_surface is lower than d13C_equilibrium (Galbraith et al., 2015; Morée et al., 2018; Schmittner et al., 2013). In your scenario where the emissions lower atmospheric d13C even further, this pattern will change. E.g. when atmospheric d13C decreases to the point that d13C_atm (e.g., -10 permil) + air-sea fractionation (e.g., +10 permil in cold waters) < d13C_DIC anywhere in the surface ocean, one gets a positive disequilibrium everywhere. The warming of surface waters will additionally decrease the thermodynamic air-sea fractionation, thereby contributing further to a global positive disequilibrium (and this is a spatially quite heterogeneous effect). Besides the aspects of disequilibrium and thermodynamic fractionation across the air-sea interface, bulk transfer of the additional low-d13C-CO_2 into the ocean will only enter the ocean in areas of net CO_2 uptake (this relates to your Revelle factor discussion). I am unsure how important dilution is here as compared to disequilibrium, bulk transfer and thermodynamic fractionation. Please discuss/clarify.

L99,L125/Fig 3b: I think it should be emphasized that the perturbation ratio is not just expected to be one and explain to the reader why (i.e. what it depends on and why it is of interest). The text under Fig. 8 in Eide et al. (2017) may be useful there.

L 119 I think it is important to refer not only to the global surface mean like in Fig 1b but also how it spatially develops (supplementary figure 6). Also in L 142-144 for example you could provide the reader with some detail about the spatial structure of these changes.

L148 I think it would help the reader to start this section with what your goals are with it and summarize its main conclusions and methods.

L 151-155 This sentence is difficult to read: I understand you want to provide the reader with a factor instead of a permil change in order to compare to DIC_ant, but maybe state the absolute change as well such that it compares easier to Fig 1b?

L 155-156 As my previous comment: The factor two is not really visible directly in Fig 3c, maybe add some absolute values for clarity?

L 158-160 Why does a stronger Suess effect response than DIC_ant mean it will penetrate less?

L 164: A reader inexperienced with d13C and its definition would not follow your sudden change to ratios here. In fact, I think the introduction of this article should start with a short intro to what d13C can be/is used for and how your study uses your analysis of the drivers of future gradient reversal to understand PETM gradient changes.

L246-256 This paragraph is more of a summary or introduction than a discussion, maybe move it to the introduction?

L 297-298 here you for the first time state that the current excursion is faster, while in L 227 you seem to conclude that the duration an magnitude is similar. Is this a comparison of the 5 kyr to the current ~150 yr anthropogenic excursion? You repeat this statement of the contemporary change being faster than the PETM change in the abstract. It undermines your comparison between the projected reversal and the PETM. More importantly, I think it needs more arguments/explanation: stress how fast and large the current CO_2 release and gradient change is, and how that is for the PETM?

L 304 Please include the name of the model here

L359 How about SST, was that fixed too? Why did you keep these aspects fixed when estimating the Suess effect in the industrial simulations? In your discussion you provide some more details on comparison with CMIP models – but they did not simulate 13C. I expect the effects of SST increase on the Suess effect are relevant for your study.

L385 Here you suddenly mention ‘The monte carlo experiment’. There are 1400 of them, rdid you pick one? Is the full model setup this fixed one monte carlo experiment? Does this mean that in all of the RCP scenario runs you have no change in air-sea gas exchange rates and SST (L 444-447 says that too)? Why not? I think to project marine d13C gradients it is important to include these.

L748 Fig 1a is not distinguishable for some color-blind readers. Style of 1a is also different from 1b,c. In 1b,c the figure would be clearer if the onset of the model experiment (2019) is indicated.

L 829 what is ‘the poorly ventilated region’, the global deep ocean below a certain depth?

L812/822 Fig. 6: I am not familiar with mbsf bins. I understand the x-axis show some sense of time through sediment depth (and you provide relative ages). The 2ky shift is not clear to me, aren’t the model data already shifted from the future to the PETM anyways, by 56 million years? What is the uncertainty of the age model? Why did you choose these planktic species?

Supplementary Text

Carbon Cycle Formulations: Do you really simulate DI12C in the model as suggested by ‘$\delta^{13}C$-DIC = [($DI^{13}C/DI^{12}C$)sample/($DI^{13}C/DI^{12}C$)standard -1]ˇ 10^3 ‘, or do you have total DIC and calculate DI12C using DIC-DI13C for sample? Could you add a few sentences on the model basics and not just refer to a supplementary of a different article (some of it can be repeated from the main text: resolution horizontal/vertical, how it compares to observations, what components are included, etc.)? Could you also shortly summarize how you set up the Monte Carlo experiments and list the exact parameters that you varied (f8, f9, etc.?). This is partly done in the main text but would be good to repeat here with additional details. From the caption of Fig S1 and the main text I get the impression that you ran 1400 experiments – this is not clear from the supplementary text?

Define ‘SGD’ at the end of page 2

‘f6 and f7 are chosen from three different values…’ what values did you choose in the end then? Do you mean randomly chosen in the monte carlo experiments?

‘The $\delta^{13}C$ endmember values for terrestrial carbon are chosen from the ranges of *f8* = -27±2‰ for DOC, *f9* = -35±2‰ for POC, and *f10* = -15±2‰ for riverine DIC [27].’ In the main text it is much clearer that the -25 is non-riverine d13C fluxes – please improve clarity here as well.

‘A suite of 10 ocean circulation fields … for each Ensemble member’ Do you mean you created 10 circulation fields which you call ensembles? They differ only slightly in nutrients/SST/salinity? It would be good to e.g. give a range of e.g. AMOC strengths/global mean SST/SSS to quantify the differences between the setups. How do these 10 circulation fields relate to the 1400 experiments?

‘A greater air-sea CO_2 piston velocity leads to a greater depletion of the δ^{13}C-DIC for the global ocean, through its influence on the oceanic uptake of anthropogenic CO_2 and enhanced exchange rates of $^{13}C/^{12}C$.’ Does it lead to a greater depletion everywhere? Or does this depend on the local disequilibrium (see e.g. the Gas Fast/Gas slow experiment in Moree et al., 2018).

P 5, last sentence ‘the other sources … (Supplementary fig. 1)’ this is a very long sentence and difficult to follow. Could you rephrase?

Supplementary Figure 5: It is confusing that you use different colors here for the RCP scenarios than in your other figures. Your reference 34 is about CMIP6 not CMIP5. Please clarify how you used CMIP5/6 (which models did you use, please provide their references to acknowledge their efforts). For benthic values, did you take the volume-weighted mean of the global bottom wet layer? In c and d 1850 would be in the upper right corner and 2100 in the lower left, right?

Supplementary Fig. 7: Here again the definition of subsurface and surface is very relevant. Do you follow the linear scaling of supplementary figure 3 here and then cap it off at 4 or 5 permil maximum change? Please provide some details of the slower model here as well (e.g AMOC/Drake passage strength as compared to the full model setup).

Minor/Technical Comments

Throughout text: I think ‘ocean circulation***s***’ even if you mean multiple states/realizations of the ocean circulation should always be ‘ocean circulation’.

L52 ‘the balance or imbalance’: the interplay

L53 ‘ocean circulations’: ‘ocean circulation

L100 ‘studies’: if only one study is cited, use ‘study’.

Use of ‘planktonic’ throughout the article and in Fig 6, please read Emiliani (1991).

L276 remove ‘paleo’

L 278-282 Long sentence which is difficult to follow.

Check your figures for color-blind suitability please

L 575 inouts -> ‘inputs’

References

Eide, M., Olsen, A., Ninnemann, U. S., & Eldevik, T. (2017). A global estimate of the full oceanic 13C Suess effect since the preindustrial. *Global Biogeochemical Cycles*, *31*(3), 492-514. https://doi.org/10.1002/2016GB005472

Emiliani, C. (1991). Planktic/planktonic, nektic/nektonic, benthic/benthonic. *Journal of Paleontology*, *65*(2), 329-329. https://doi.org/10.1017/S0022336000020576

Galbraith, E. D., Kwon, E. Y., Bianchi, D., Hain, M. P., & Sarmiento, J. L. (2015). The impact of atmospheric pCO2 on carbon isotope ratios of the atmosphere and ocean. *Global Biogeochemical Cycles*, *29*(3), 307-324. https://doi.org/10.1002/2014GB004929

Liu, B., Six, K. D., & Ilyina, T. (2021). Incorporating the stable carbon isotope 13C in the ocean biogeochemical component of the Max Planck Institute Earth System Model. *Biogeosciences*, *18*(14), 4389-4429. https://doi.org/10.5194/bg-18-4389-2021

Morée, A. L., Schwinger, J., & Heinze, C. (2018). Southern Ocean controls of the vertical marine δ13C gradient – a modelling study. *Biogeosciences*, *15*(23), 7205-7223. https://doi.org/10.5194/bg-15-7205-2018

Schmittner, A., Gruber, N., Mix, A. C., Key, R. M., Tagliabue, A., & Westberry, T. K. (2013). Biology and air-sea gas exchange controls on the distribution of carbon isotope ratios (δ13C) in the ocean. *Biogeosciences*, *10*(9), 5793-5816. https://doi.org/10.5194/bg-10-5793-2013

We would like to thank Anne Morée and two anonymous reviewers for the valuable comments and suggestions. The comments and references have been very useful in this revision. Please find our point-by-point responses to reviewers' comments, which are shown in blue fonts:

Reviewer #1 (Remarks to the Author):

Review of "Unprecedented Reversal of the Oceanic d13C Depth Gradient Due to Extreme Greenhouse Gas Emissions" by Kwon et al., submitted to Communications Earth and Environment, July 2021.

Overall this is a solid paper with a detailed model and the results are well-explained. The behaviour of the model is not too surprising, and I wonder if some summary statements in the conclusions along these lines are warranted: When 13C-depleted carbon release into the atmosphere is faster than deep-ocean mixing timescales (so occuring in centuries or just one or two thousand years), the Suess effect overwhelms the biological pump and you get a d13C-depth gradient reversal. When carbon release is slower than ocean mixing timescales, the light C has time to mix to depth and so depth d13C gradients are preserved.
The above seem like a simplified, general interpretation of the model results and apply to the PETM (during which C release seems to be just on the bubble of being faster than ocean mixing timescales, so you get a short-lived d13C-depth reversal or collapse). Also in the case of the PETM, it is generally thought that deepwater circulation was "sluggish" during the Eocene greenhouse, so an somewhat slower C release (a few thousand years) could cause a d13C gradient collapse.

=> Thank you for the compliment and suggestion. The summary statement is implicitly included in the revised abstract and main text. In particular, we have added in the main text as

"Under scenarios of slowly increasing atmospheric CO_2 over 20 kyr (ref. (Cui et al., 2011)), no reversals of the vertical δ^{13}C-DIC gradient are apparent even in poorly ventilated regions (Fig. 5c). Slow increases in atmospheric CO_2 (even slower than the vertical ocean mixing timescales) give the deep ocean sufficient time to fully equilibrate to changing atmospheric CO_2."

A couple of specific comments:

Line 468: It seems to me to be a mistake to apply no d13C correction to Acaranina and Morozovella, because we know (through stable isotope ecology studies and shell size-d13C relationships) that these genera harbored photosynthetic symbionts, which increase d13C in the calcifying microenvironment. I suggest that they assume a d13C correction from a modern photosynthetic symbiont planktic foraminifer species (such as G sacculifer) as this is more likely to be closer to the truth than simply assuming no d13C offset for these ancient species.
And this becomes complicated in the case of the PETM because those very same taxa seem to (at least at some sites) lose their photosymbionts during the PETM, according

to the very new Shaw et al paper that you cite (which is now fully published so you can update the citation from "in press").

=> Thank you for the suggestion and comments. We agree that the paleo foraminifera d13C records have large uncertainty, especially for the already extinct species like Acaranina and Morozovella. Applying any corrections to the foraminifera d13C records is however beyond our scope and expertise in the absence of any previous studies for the suggested corrections. Given the large uncertainties for the paleo d13C records associated with the benthic (concerns raised by Reviewers #2 and #3) and planktic foraminifera records, we have decided to remove the previous Fig.6a (showing the Cenozoic foraminifera d13C records) from this study. Nevertheless, we keep the previous Figs. 6b and 6c (now moved to Figs. 6a and 6b), which are based on single foraminifera species over the PETM onset. We have also added Fig. 6c, which is based on the modern 13C Suess effect taken from the previous Fig. 6a.

We have also added to Methods as *"No corrections are applied to the foraminifera $\delta^{13}C$ records because of the lack of information. Nevertheless, uncertainties are likely small in this single species comparison at the two open ocean sites where a reduced photosymbiosis has been reported for the PETM acarininids and morozovellids (therefore less offset from seawater $\delta^{13}C$-DIC) (Shaw et al., 2021)."*

We have also updated the Shaw et al. reference.

Figure 3: Can you mark the North Atlantic - Southern Ocean - North Pacific transect used in column A on the maps of column B?

=> The maps show the <u>zonally averaged</u> vertical sections from the North Atlantic to the North Pacific. Therefore, there exists no single line representing the transect. We have clarified this in the figure caption as *"The zonally averaged vertical sections of $\delta^{13}C$-DIC"*.

Reviewer #2 (Remarks to the Author):

The manuscript by Kwon et al. appears to have multiple objectives, but the presentation leaves me unclear as to what the major or novel conclusions are meant to be. I am unclear as to whether the paper is primarily meant to evaluate how spatial patterns of DIC-13C will evolve under future RCP scenarios or to provide an interpretation of PETM-carbon injection. While I understand the motivation to use the PETM as a future analog, I don't think that presentation works successfully here, and I'm left wondering whether the authors are making an argument that the PETM changes constrain the future or that the modeling of the future constrains the interpretation of the PETM.

=> We hope that our revised Abstract and Introduction provide the objectives of this study as well as our novel conclusions more clearly. For example, we have clarified our objectives in Introduction as

"Here, we use an observationally constrained global oceanic carbon cycle model (DeVries, 2014; Kwon et al., 2021) and four Representative Concentration Pathways (RCPs) (Moss et al., 2010) to estimate the temporally evolving oceanic δ^{13}C-DIC from the preindustrial era to the 21st century (Methods). We find that the 21st century greenhouse gas emissions will induce elimination or even reversal of the vertical δ^{13}C-DIC gradient by the end of the 21st century and elucidate the geochemical mechanisms underlying such radical changes in surface δ^{13}C-DIC. Our 21st century projections now allow us to compare the δ^{13}C-DIC excursions between the modern and the PETM onsets, which will benefit from the latest compilation (Shaw et al., 2021) of the PETM foraminifera δ^{13}C records."

Although we mostly agree with the reviewer's comments (see below), our manuscript remains novel in the following three aspects:

1. As the reviewer pointed out, our work provides the 21st century projections of oceanic d13C-DIC based on the well-constrained historical (preindustrial to 2018) estimates and the plausible RCP scenarios.
2. We present the convincing evidence for the early PETM d13C vertical gradient reversal, which is made possible by using the latest compilation of foraminifera d13C records.
3. We explore whether the observed d13C gradient reversal is consistent with previously suggested carbon emission rates for the PETM onset. See also our responses right below.

Beyond this confusion as to the objectives of the study, my primary criticism of the paper is that it completely misinterprets the results of Ref. (8), a study that modeled changed in vertical d13C-DIC gradients across the PETM onset using the cGENIE model. That study in fact found major delays in the propagation of the d13C anomaly to the deep ocean (it rather argued that major delays were not possible from the surface ocean to the thermocline). Here, the authors summarize that paper to say that it argued against reversals in the d13C-depth gradient. As I describe below in more detail, that is incorrect, and because that study did in fact present significant durations of reversed surface-deep gradients, this then conflicts with the narrative that this study presents original results.

=> Thank you for the corrections regarding our mis-interpretation of Turner et al. (2017). We have corrected our interpretation of the reference in Introduction as

"Foraminifera fossil δ^{13}C records also suggest that the pre-PETM depth gradients might have been eliminated or even reversed during the PETM onset (Kennett & Stott, 1991; Zachos et al., 2007). However, because the carbon emission rates for the PETM onset were estimated to be an order of magnitude slower than those during industrialization (Cui et al., 2011; Penman & Zachos, 2018; Zeebe et al., 2016), the PETM deep ocean might have had sufficient time to fully equilibrate to changing atmospheric CO_2 without necessarily invoking a vertical gradient reversal (Cui et al., 2011). Nonetheless several studies (Cui et al., 2011; Turner et al., 2017; Turner & Ridgwell, 2016) suggested that relatively rapid carbon emissions could have delayed deep ocean δ^{13}C-DIC excursions compared to the surface, causing a temporal reversal

of the vertical δ^{13}C-DIC gradient. Such sensitivity indicates that the early geochemical evolution of the vertical δ^{13}C-DIC gradient can provide an important constraint on the PETM onset duration, which has previously been suggested to be between 5 kyr and 20 kyr (Cui et al., 2011; Penman & Zachos, 2018; Zeebe et al., 2016).”

This correction however does not degrade the novelty of our work because our study is based on the two plausible carbon emission rates for the PETM onset (as suggested by Penman and Zachos (2018) and Cui et al. (2011)), whereas Turner et al. (2017) obtained the surface-deep d13C gradient reversals in a highly idealized pulse-like atmospheric forcing. For example, Turner et al. (2017) described their experiment as *“in response to injection over a single year of 2275 Pg C with δ13C of −60‰ to the atmosphere”*, with a related statement of *“We chose to employ an extreme scenario and apply a pulse of carbon released over a single year and sufficient to drive a −4‰ global δ13C excursion, following ref. 13—not because we consider such a scenario likely (or even plausible), but to create a step change in δ13C most reminiscent of the face-value interpretation of the data (Fig. 2).”*

We rather believe that Turner et al. (2017) indeed provide motivations for this study as shown in Introduction (above) and in the main text. For the latter, we have also added *“In doing so, it is important to consider different ocean ventilation states, because the duration and magnitude of the vertical δ^{13}C-DIC gradient reversals are also sensitive to ocean stratification and ventilation rates (Turner et al., 2017; Turner & Ridgwell, 2016).”*

I see value in the extensive ensemble for preindustrial DIC-13C conducted here, though that is also not novel (see Holden et al., 2013). However, the relegation of details to the Methods and Supplementary sections is really unfortunate. Moreover, I do not understand the choice to fix the ocean circulation response across all of the simulated experiments (rather than as a subset to provide a control). In Ref. 8, that study explicitly compared different assumptions about the degree of ocean stratification and the representation of vertical mixing in the model, but also used a ‘no circulation change’ control, and it would be very interesting to know how those model results compare with the model used here.

=> Thank you for pointing out the reference of Holden et al. (2013). It is encouraging to see a consistency between our study and Holden et al. (2013), despite the entirely independent approaches taken. We have added in the main text as
“A dominance of air-sea CO_2 exchange rates for the uncertainty is in line with a previous study (Holden et al., 2013) based on an Earth System Model which suggested that air-sea gas exchange rates alone explain 63% of the total variance of simulated oceanic ^{13}C Suess effects over a time period of 1858-2008.”

We fix ocean circulation in this study because our offline ocean model framework does not allow us to explore the responses and feedbacks from ocean circulation change. However, the uncertainty associated with this assumption in future d13C-DIC projection is likely to be small, as we discussed in the main text as
“Additional sources of uncertainty, including temporal changes in ocean circulation

and the biological carbon pump, also appear to be minor for the 21st century changes in DIC and ocean pH, as shown by the close agreements between our estimates and those projected from multiple climate models (Kwiatkowski et al., 2020) (Supplementary Fig. 6b), and also possibly minor for δ^{13}C-DIC (Holden et al., 2013). The modulating effects of climate change on δ^{13}C-DIC distributions may emerge on timescales that are longer than the timescales of the geochemical effects from rising atmospheric CO_2 (refs. (Schlunegger et al., 2019; Turner & Ridgwell, 2016)). Hence, we mainly focus on the effects of different CO_2 emissions scenarios applied to the full model setup (Supplementary Table 1) for the 21st century δ^{13}C-DIC projections."

Unlike ocean circulation changes, potential changes in air-sea gas exchange rates (identified as an important source of uncertainty in the modern ocean 13C Suess effect by Holden et al. and this study) could be important. Therefore, we have explored the effects of future changes in air-sea CO2 transfer rates on the simulated 13C Suess effect. The results are presented in Supplementary Figs. 4 and 5 and in Supplementary Information. In the main text, we have added

"Compared to the effects of the different CO_2 emission scenarios (Fig. 1b), the effects of potential changes in air-sea CO_2 exchange rates (another primary source of uncertainty identified for the estimated oceanic ^{13}C Suess effect as of 2018) appear to be an order of magnitude smaller (Supplementary Figs. 4 and 5; Supplementary Information)."

"Projected global warming (e.g., ref. (Rodgers et al., 2021)) is expected to further lower surface δ^{13}C-DIC through increasing air-sea CO_2 exchange rates in high latitudes (mostly due to sea ice melting) and enhanced thermodynamic isotopic fractionations whose effects are most pronounced in low latitudes (Supplementary Fig. 5; Supplementary Information). These warming driven surface δ^{13}C-DIC reductions can additionally elevate the ratio of the ocean to atmosphere ^{13}C Suess effect, and also enhance the vertical δ^{13}C-DIC gradient reversal as of 2100. When globally averaged, the warming effects are small with an additional surface δ^{13}C-DIC decline of only –0.1 ‰ compared to the geochemically driven ^{13}C Suess effect of –3.7 ‰ under the RCP6.0 scenario (Supplementary Fig. 4). Yet, regionally the effects can be as large as 50% in the Weddell Sea (Supplementary Fig. 5). A more comprehensive assessment of ocean stratification and circulation change effects on δ^{13}C-DIC projections remains a subject for future studies."

Regarding the suggested comparison between our 'no circulation change' experiment with previous studies employed changes in ocean stratification mixing, we have added in the main text as
"These disparities imply that concomitant transient changes in ocean circulation, marine biology, land-derived carbon inputs, and marine sedimentary dissolutions of $CaCO_3$ might have been as important as the geochemical effects for the early PETM δ^{13}C excursion (e.g., refs. (Ilyina & Heinze, 2019; Nunes & Norris, 2006; Turner et al., 2017; Turner & Ridgwell, 2016)). In fact, Turner and Ridgwell (2016) showed that the CO_2-climate feedbacks during the PETM onset can delay the time that it takes for the surface δ^{13}C-DIC minimum to propagate to the deep ocean δ^{13}C-DIC minimum by up to 40%."

Finally, in a paper entirely about the modeled representation of ocean d13C, the treatment of model description within the text is inadequate.

=> We have employed both model and observations for this study. Because the model as well as the Monte Carlo experiment is already documented in Kwon et al. (2021), we hope that the citation of the previous study and the model descriptions in Methods and Supplementary Materials are adequate.

(Intro)
Line 44 - this is a misleading summary of the interpretation of changing d13C gradients during the PETM onset from Ref. (8). If you view Fig. 2 in Ref (8), there is very clearly an interval of reversed d13C gradients with depth - the d13C values of the mixed layer and thermocline in that model (cGENIE) are lighter than the benthic layer. The argument in Ref. (8) is rather that there cannot be a delay of 5-15,000 years before the thermocline sees excursion values, which had been argued previously based on interpretation of single foraminiferal data from ODP Site 690. Quoting from that paper, "we note that significantly larger delays are possible before the excursion reaches the deep ocean." In the Supplementary Table 1 referenced in the text, the delays between the surface and benthic excursion reach thousands of years (sometimes greater than 5 kyr), depending on the degree of warming and hence stratification as well as the mixing parameterization used. Hence, I take issue with the author's statement that 'It is fair to say that the early geochemical evolution of the vertical d13C-DIC gradient remains uncertain.' That statement effectively ignores the results presented in Ref. 8.

=> Thank you for correcting us. We have revised Introduction accordingly and corrected our mis-interpretation of Turner et al. (2017) study. See our response above.

Line 65 - I am confused as to whether the objective of this paper is to evaluate the PETM or to evaluate the potential for d13C gradient reversals due to various RCP scenarios. The connection between the two is not well established. On the one hand, you might indicate that you are using the pre-industrial to future change to test the model behavior specifically in order to evaluate PETM records, but that is not really the way the introduction reads. I'm left struggling a bit as to the principle objective of this study. This is compounded by the title, which is ambiguous as to whether it refers to the future, the PETM, or just warming generically.

=> Thank you for the comments. We have slightly changed the title as *"Unprecedented Reversal of Oceanic $\delta^{13}C$ Depth Gradient due to 21st Century Greenhouse Gas Emissions"*. We also have clarified our objectives in Introduction. See our response above.

(Methods)
Line 314 - I think it is inappropriate to provide only a reference, and no summary model description of the treatment of carbon isotopes in the model used, since this is the crux of this entire study. The level of detail provided in Methods does not need to be to the level of detail in the Supplement, but it is absolutely possible to summarize in a

paragraph. The ensemble alone could warrant its own manuscript, and the very short treatment in the main text feels incomplete.

=> We have added a summary of carbon isotope model description to Methods as

"The ocean biogeochemical processes are formulated following the OCMIP2 protocol (Najjar et al., 2007) where ocean productivity is simulated by restoring model surface PO_4 towards the observed PO_4. Simple parameterizations for the production and remineralization of organic and inorganic carbon are employed. The carbon isotope model uses two prognostic variables of $DI^{13}C$ and $DI^{12}C$ (the latter approximated as DIC), and the isotopic signature of DIC is estimated as $\delta^{13}C$-DIC = $[(DI^{13}C/DI^{12}C)_{sample}/(DI^{13}C/DI^{12}C)_{standard} - 1]$ with the Vienna Pee Dee Belemnite standard. The model employs a temperature dependent thermodynamic equilibrium fractionation from Zhang et al. (1995) and a CO_2 dependent photosynthetic fractionation following three different empirical formulations (Supplementary Table 1). The model includes riverine carbon inputs and a simple parameterization for the sedimentary burial of inorganic carbon."

"Preindustrial steady-state solutions are obtained using a time efficient Newton's method (Kwon & Primeau, 2008) whereas industrial changes are simulated by taking time steps with an atmospheric CO_2 forcing taken from observation-based estimates (Keeling et al., 2005; MacFarling Meure et al., 2006) for a time period of 1780-2018 and RCP scenarios (Moss et al., 2010) for a time period of 2019-2100. Our model is based on an annual mean climatology that is invariant over time except for the atmospheric CO_2 and $\delta^{13}C$-CO_2 forcing that change over industrial times. The model was previously shown to simulate the oceanic uptake and storage of anthropogenic carbon (DeVries, 2014), as well as the oceanic ^{13}C Suess effect (Kwon et al., 2021) (Fig. 1a), consistent with previous independent estimates. We refer readers to Kwon et al. (2021) and Supplementary Information for details of the carbon isotope model formulations. Although our primary focus is the geochemical effects from changing atmospheric CO_2, our offline model framework allows us to perform an experiment where some of biogeochemistry model inputs are taken from results of an Earth System Model (Supplementary Information)."

Line 345 - I am trying to follow the reasoning behind the two methodologies described for estimating pre-industrial d13C-DIC but finding the detail insufficient. The authors first describe using a large ensemble to generate both a series of pre-industrial steady states as well as various trajectories from pre-industrial to modern. However, because of the degree of variation among the simulated pre-industrial steady state d13C-DIC, the authors do not use any of these simulations and instead use an observational compilation and add a spatially uniform(?) correction based on the Suess effect, hence retaining modern patterns but not absolute values for the pre-industrial DIC-13C pattern? So, does this method provide a distribution that is different from any one of 1400 ensemble members? Or significantly different from the ensemble mean?

=> We have added in the main text as
"The best estimate for the preindustrial $\delta^{13}C$-DIC distributions can be derived by

combining our constrained oceanic ^{13}C Suess effect with contemporary observations (Schmittner et al., 2017) (Fig. 2b; Supplementary Fig. 2; Methods). This approach is chosen rather than using simulated preindustrial $\delta^{13}C$-DIC distributions due to the large sensitivity of the preindustrial $\delta^{13}C$-DIC estimates to a poorly constrained boundary condition of terrestrial carbon inputs (Kwon et al., 2021) (Supplementary Fig. 1a) and the unexplored sensitivity to the biological carbon pump (Morée et al., 2018).”

We have also clarified in Methods as
“To obtain a climatological mean distribution, we average the gridded data over time such that each grid cell has an averaged year of data collection and an averaged $\delta^{13}C$-DIC value (Supplementary Fig. 2a). Then, the mapped $\delta^{13}C$-DIC is corrected for the ^{13}C Suess effect with our estimate that is taken from the averaged year of data collection for each grid cell (Supplementary Fig. 2c)… This approach gives a preindustrial state of $\delta^{13}C$-DIC (Fig. 2b; Supplementary Fig. 2b) that is similar to our simulated preindustrial $\delta^{13}C$-DIC with reduced uncertainty.”

Line 384 - when “we choose a model configuration from the Monte Carlo experiment,” what configuration was this? How did you choose? When you say you ‘fix the ocean circulation state,’ what does this mean explicitly?

=> We have clarified in Methods as
“To this end, we use a model configuration, considered as a typical model setup in carbon isotope modeling (Jahn et al., 2015; Liu et al., 2021; Schmittner et al., 2013), from the Monte Carlo experiment (the “full” model setup in Supplementary Table 1). Using the full model setup, we assume that the ocean circulation, air-sea CO_2 exchange rates including sea ice effects, and sea surface temperature and salinity remain unchanged with time throughout the simulation. An uncertainty associated with this assumption is tested in Supplementary Materials where we relax the assumption of unchanged air-sea CO_2 exchange rates, and sea surface temperature and salinity using the Community Earth System Model version 2 (CESM2)-based estimates (Danabasoglu et al., 2020; Rodgers et al., 2021).”

Line 414 – To me, it’s really not helpful to describe these idealized sensitivity experiments as PETM-like. If nothing about the model is configured for the late Paleocene, then why is this different than just considering these as hypothetical future experiments? Also, over these long timescales, I’m even more perplexed by the choice to only evaluate experiments where the circulation is fixed throughout the simulation. Why bother simulating thousands of years of model time and ignoring the fact that the model will predict a change in the circulation state in response to warming over these timescales? Rather, I can see using ‘circulation-fixed’ experiments as a control against which to constrain the impact of changes in circulation.

=> Our sensitivity experiments for a PETM discussion are far from future experiments because the CO2 emission rates are at least on order of magnitude smaller. Rather, we have rephrased the experiment as *“Sensitivity Experiments with Estimated PETM Atmospheric CO_2”*

As we mentioned earlier, our offline modeling framework does not allow us to explore the CO2-climeate feedbacks. Despite this limitation, our study provides geochemically driven oceanic 13C Suess effect and is complemented by previous studies (which the reviewer provided) that explored the effects of CO2-climate feedbacks. As quoted already, we have added *"In fact, Turner and Ridgwell (2016) showed that the CO_2-climate feedbacks during the PETM onset can delay the time that it takes for the surface δ^{13}C-DIC minimum to propagate to the deep ocean δ^{13}C-DIC minimum by up to 40%."*

Line 448 - How can you generically use all epifaunal benthic values to represent d13C values below 74 m? Do you not discriminate further by depth? Why do you even need the last 65 Myr of d13C data? (I don't see what Fig. 6a adds to the manuscript). It is confusing to first suggest that you use data from a compilation (Ref. 30) and then indicate you selected two sites only as if those are the only two open ocean PETM sites. Moreover, if you provide both the original data sources and the references for the age models used for those two sites, what does it even mean to say you used data from the Shaw compilation?

=> We have chosen to remove the previous Fig. 6a due to the high uncertainties in the long-term evolutions of d13C and concerns raised by all of the reviewers. Yet, our conclusion is still supported by the revised Fig. 6 that is based on the latest compilation of the PETM foraminifera d13C data.

Although we provide the original data sources and the references for the age models used for those two sites, we still need to give a credit to Shaw et al. for the data compilation. Without the use of the compiled dataset, our study might have taken much longer.

(Results)
Line 90 - what about near regions of deepwater formation?

=> We have revised the sentence as *"The most pronounced ^{13}C-depletion in δ^{13}C-DIC occurs in the subtropical surface waters, North Atlantic Deep Water, and global mode and intermediate waters (Fig. 2a)."*

Line 111 - what do you mean by 'revealing higher spatial gradients' Is this simply to point out that a higher d13C for the preindustrial surface necessarily then means larger d13C gradients with depth in the pre-industrial compared to modern? Or are you saying something about horizontal gradients in the surface?

=> We have revised as *"revealing higher horizontal and vertical gradients associated with water mass distributions in the upper ocean"*

Line 123/Line 155 - somewhere you should formally introduce what you mean by perturbation ratio - you use this to refer to multiple different quantities.

=> We have added *"the perturbation ratio (defined as the ratio of deviations from the respective preindustrial values)"*

Line 212 - again, this description of the findings of Ref. 8 is wrong and misleading.

=> We have deleted the sentence.

Line 220 - what is the motivation of using different ocean ventilation states? Can you be explicit about what you are testing with these model experiments and what you expect to find?

=> We have added
"To explore whether the observed δ^{13}C-DIC gradient reversal is consistent with previously suggested carbon emission rates during the PETM onset, we apply the following two emission estimates to our model: a relatively rapid increase in atmospheric CO_2 over 5 kyr (Penman & Zachos, 2018) and a slow increase over 20 kyr (Cui et al., 2011). In doing so, it is important to consider different ocean ventilation states, because the duration and magnitude of the vertical δ^{13}C-DIC gradient reversals are also sensitive to ocean stratification and ventilation rates (Turner et al., 2017; Turner & Ridgwell, 2016)."

Line 252 - the presentation of the novelty of these experiments is misleading. There is no reference of the ensemble evaluation of modeled pre-industrial to modern DIC-13C by Holden et al. (2013), and the cGENIE model has been used multiple times in evaluating d13C in response to CO2 injection (both future and past). See Norris et al., (2013), Kirtland Turner and Ridgwell, 2016. The detailed evaluation of future spatial patterns in d13C specifically under RCP scenarios is, I believe, novel, but then I think a major missing factor is any exploration of how modeled changes in ocean circulation impact these distributions.

=> Thank you for providing the references. We have found the references essential for motivating and complementing this study, and hence incorporated them to our revision. We have also strengthened our discussions of 21st projected 13C Suess effect by exploring the effects of future changes in air-sea CO2 transfer rates and sea surface temperature and salinity.

Refs:
Holden, P.B., Edwards, N.R., Müller, S.A., Oliver, K.I.C., Death, R.M. and Ridgwell, A., 2013. Controls on the spatial distribution of oceanic δ 13 C DIC. Biogeosciences, 10(3), pp.1815-1833.

Norris, R.D., Turner, S.K., Hull, P.M. and Ridgwell, A., 2013. Marine ecosystem responses to Cenozoic global change. Science, 341(6145), pp.492-498.

Turner, S.K. and Ridgwell, A., 2016. Development of a novel empirical framework for interpreting geological carbon isotope excursions, with implications for the rate of carbon injection across the PETM. Earth and Planetary Science Letters, 435, pp.1-13.

Reviewer #3 (Remarks to the Author):

Thank you for your interesting contribution to the discussion of the interpretation of marine d13C-records. Your work can be of interest to a wide audience ranging from contemporary/future carbon isotope and carbon cycle scientists, to paleoclimatologists interested in the drivers of the PETM.
Your goal has been to use your projected changes in the marine d13C gradient as an analogue for the PETM d13C changes. Your results lead to your main claims that 1) the PETM elimination/reversal of the d13C gradient was driven by fast (land biosphere) release of CO2 (and its accompanying low-d13C) and 2) that the projected 21st century d13C gradient changes are likely to lead to elimination/reversal of the gradient as compared to pre-industrial times and are unprecedented over the past 65 million years. You have run your model in different setups to explore drivers/mechanisms, quantify changes and find analogues in the projected gradients for the PETM gradients. I think your results are novel and interesting, but I have several comments. Specifically, quite many conclusions are drawn and supported by quite many different methods (different model runs/datasets), which makes it difficult for the reader to find your 'take home message' or follow the main storyline.
Actually, writing two separate papers may help communicate your work. I have focused on the non-PETM part of your manuscript as this is where my expertise lies. In any case I start the review document with some general comments, followed by more specific comments, then comments to the supplement and finally some technical comments.
Find it attached.

=> Thank you for the general assessment of our work.

General comments
1. From the abstract and the title, I got the impression that this paper was mostly about a present/future reversal of the marine d13C gradient. However, you also want to make the connection to the PETM. You state in the abstract: a) from model projections we find that a reversal is expected (not surprising), b) this reversal is driven by reducing surface d13C-DIC due to low-d13C fossil fuel emissions and facilitated by enhanced equilibration under high DIC conditions (interesting analysis), c) such reversals are also present in the PETM (new), d) The PETM change was however much slower (this comes a bit suddenly and with little backup). Your statement on the unprecedented 65 million years can only be made if you have a decadal resolution of d13C gradients over these 65 million years, such that you can compare the records to the contemporary Suess-effect which occurs over the course of a few decades. Can you really do that? Is the PETM CO2 release and its corresponding d13C signature likely comparable to the future one? Does the uncertainty of the PETM records allow for the conclusion that the gradient reversed? Also, you could make clearer that your record (Fig.6) is 65 million years such that you can make the 65-Myr statement - beyond the PETM.

=> Our revised Fig. 6c has the upper X-label with relative time since 1800 CE, which can be compared with the relative time since the base of PETM onset in Figs. 6a and 6b. We have also clarified the comparison between the future and PETM d13C excursion in the last paragraph of the manuscript as

"Despite the analogy between the modern and PETM onset in terms of the large δ^{13}C-DIC excursion and the existence of vertical gradient reversal, the rate at which the 21st century anthropogenic carbon isotope excursion occurs is at least one order of magnitude faster than PETM excursion rates (Fig. 6c). A precise comparison of the time rate of change is hampered due to the difficulty in reconstructing high temporal resolution PETM δ^{13}C records (e.g., refs. (Shaw et al., 2021; Westerhold et al., 2018)) and large uncertainties in age models (e.g., refs. (Bains et al., 1999; Thomas & Shackleton, 1996)). Nevertheless, the 21st century δ^{13}C-DIC gradient reversal rates (taking only ~3 centuries from the preindustrial era to the projected gradient reversals) appear to be much faster than those of the PETM (taking at least 5 kyr from the pre-PETM to the maximum gradient reversal). Given the fact that the PETM is the best known ancient time when the most rapid carbon emission has been reported over the Cenozoic (e.g., refs. (Zachos et al., 2001; Zeebe et al., 2016)), our comparison suggests that the time rates of 21st century δ^{13}C-DIC excursion and associated gradient reversal may be unprecedented over the Cenozoic. "

Although uncertainty is large for the PETM records, we believe that our Fig. 6 is the best comparison (so far) between the modern and early PETM evolutions of oceanic d13C-DIC. We have decided to remove the previous Fig. 6a showing the 65 Myr long records due to the concerns raised by all of the three reviewers. Even without it, our conclusion remains robust given the currently available PETM d13C records and literature.

2. Text structure: The Results section now has 4 very separated subsections. The PETM is not even mentioned in the first 3. This is an example of where the reader may forget your overarching storyline of the PETM gradient reversal: How do each of these sections contribute to your storyline and main points? Why is quantifying the Suess effect relevant for your PETM conclusions/analysis? At least I need some more help here to be guided through your steps.

=> We have restructured the manuscript by replacing separate "Results" and "Discussions" sections with a combined section of "Results and Discussions" that has 4 subsections. The restructuring and revision have helped us improve the flow and clarity of this manuscript.

The subsections under "Results and Discussions" are still separate due to different approaches taken and different timescales involved. Yet our revised subsections all support our storyline outlined in Introduction (i.e., the last paragraph of Introduction).

We have also revised the abstract to better reflect our objectives and results as

"The stable carbon isotope compositions (δ^{13}C) of dissolved inorganic carbon (DIC) recorded in marine carbonate fossils are one of the most commonly used proxies for the reconstruction of past changes in the global carbon cycle. However, the preindustrial state of δ^{13}C-DIC and its industrial changes are poorly understood, hampering a comparison with the past. Here we use an observationally constrained ocean model and various greenhouse gas emissions scenarios to show that the globally averaged surface

ocean is projected to decrease in δ^{13}C-DIC by –(1.8 to 6.3) ‰ as of a year of 2100. This reduction, driven by the oceanic absorption of ^{13}C-depleted anthropogenic CO_2 and facilitated by enhanced air-sea carbon isotopic equilibrium under higher DIC conditions, may eliminate or reverse the naturally formed vertical gradients towards the end of the 21st century. Such gradient reversals were apparent in the early Paleocene-Eocene Thermal Maximum (PETM) foraminifera fossil records, supporting rapid carbon emission rates over ~5 thousand years and stagnant deep waters during the PETM onset."

We have also revised Introduction to clarify why quantifying future 13C Suess effect is relevant to our PETM analyses, as

"The best known geologic analog to the future perturbation of the global carbon cycle is the PETM (approximately 56 million years ago) (Norris et al., 2013), which was characterized by carbon isotope excursions of –(3 to 5) ‰ (Kennett & Stott, 1991; McInerney & Stott, 2011; Zachos et al., 2007). These anomalies were accompanied by rising temperatures and widespread ocean acidification, and have been attributed to a rapid carbon release of ~10,000 GtC to the climate system (Kennett & Stott, 1991; McInerney & Stott, 2011)."

3. I soon lost the overview over what experiments and sensitivity analysis were performed. Different circulation states were explored (10 according to the supplement?) as well as 1400 monte carlo experiments (supplement caption figure 1), as well as a 'full model' setup, and a fixed.CO2 and fixed.ratio setup. And then the PETM 4 permil and 5 permil simulations. These are outlined in the methods, but still I find it hard to get an overview. Could you at least rewrite/add to L 60-68, summarizing the goals and main characteristics of all your different methods/approaches?

=> Instead of providing a summary statement about the different approaches in Introduction, we have added an appropriate summary to the beginning of each subsection as follows.

"Modern Ocean ^{13}C Suess Effect

The oceanic δ^{13}C-DIC changes from the preindustrial era to 2018 are assessed based on a Monte Carlo experiment (Methods) where uncertainties in various model parameters (Supplementary Table 1) are allowed to propagate in our model into the δ^{13}C-DIC simulations."

"Projected 21st Century Changes in δ^{13}C-DIC

We use four CO_2 emission scenarios (Moss et al., 2010) and the linear relationship between the atmospheric CO_2 and the δ^{13}C of CO_2 estimated based on last decades of observations (Keeling et al., 2005) (Supplementary Fig. 3) for the 21st century projections (Methods)… Hence, we mainly focus on the effects of different CO_2 emissions scenarios applied to the full model setup (Supplementary Table 1) for the 21st century δ^{13}C-DIC projections."

"Geochemical Mechanisms for the Ocean ^{13}C Suess Effect

We elucidate the geochemical mechanisms by which the 21st century CO_2 emissions can drive such radical changes in surface δ^{13}C-DIC."

"Comparison with PETM Depth Gradient Reversal

... To explore whether the observed δ^{13}C-DIC gradient reversal is consistent with previously suggested carbon emission rates during the PETM onset, we apply the following two emission estimates to our model: a relatively rapid increase in atmospheric CO_2 over 5 kyr (Penman & Zachos, 2018) and a slow increase over 20 kyr (Cui et al., 2011)."

We hope that this introductory paragraphs guide readers regarding how we arrive at the results of each subsection.

4. Some discussion is missing: What other explanations have been given by the literature of the PETM gradient elimination/reversal?

=> To our best understanding, no previous studies have assessed the early PETM reversal of the vertical d13C gradient to the extent that we have done here, presumably due to the lack of observations in earlier times. For example, a study of Zachos et al., (2007) seems to be the best work presenting the reversal of surface-deep ocean d13C contrasts during the PETM onset. Due partly to the lack of convincing observational evidence, the PETM gradient reversal/elimination have been discussed only in context of highly idealized pulse-like carbon emission scenarios in previous studies (cited in our manuscript and provided by Reviewer #2). In this regards, our study is novel and important. We are not aware of any other explanations except the CO2-climate feedbacks extending the reversal duration as discussed in our revision.

The range of d13C POC/DIC/DOC you explore in your Monte Carlo experiments is quite limited in my opinion (Supplementary table 1), variations in the abundance of C3 versus C4 plants (~-25 permil vs. -12 permil respired CO2) must have driven a much larger range?

=> The d13C POC/DIC/DOC explored in this study is the globally averaged d13C values for riverine carbon fluxes. Those can vary regionally depending on the dominance of C3 or C4 plants. However due to the lack of data and information, we do not consider regional variations of d13C POC/DIC/DOC. The values we used are based on observations averaged globally (Marwick et al., 2015; Peterson & Fry, 1987). Nevertheless, an even larger uncertainty lies in the non-riverine carbon fluxes, which we have varied from 0 to 1.4 GtC/yr. Because the effects on oceanic d13C-DIC are the same between riverine and non-riverine carbon fluxes, the limited range in d13C POC/DIC/DOC explored in this study may have been covered by the large variation of 0 to 1.4 GtC/yr.

We have revised the Supplementary Information as
"The δ^{13}C endmember values for riverine carbon are chosen from the ranges of f8 = -

27±2‰ for DOC, f9 = -30±2‰ for POC, and f10 = -15±2‰ for DIC (Marwick et al., 2015; Peterson & Fry, 1987). For the coastal margin carbon flux, the δ^{13}C endmember value is fixed at -26‰ (Abril et al., 2013; Maher et al., 2013). Although the δ^{13}C values for the riverine and coastal margin inputs are highly uncertain spanning -(14-30)‰ (Abril et al., 2013; Maher et al., 2013), its uncertainty is implicitly included in our Monte Carlo experiment because the effects on the δ^{13}C-DIC are identical between the magnitude of the coastal margin inputs (f2) and the δ^{13}C values of the riverine or coastal margin inputs."

Curiosity: to what extent has the gradient elimination/reversal been observed yet in the contemporary ocean?

=> A previous study based on observations reported that the gradient has been nearly eliminated as we cite *"Nevertheless, our results support a previous finding that surface water δ^{13}C-DIC was more positive during preindustrial times than the present-day observations, revealing higher horizontal and vertical gradients associated with water mass distributions in the upper ocean (Eide, Olsen, Ninnemann, & Johannessen, 2017; Olsen & Ninnemann, 2010)."*

5. I think your study should be able to state how fast a release of a certain amount of CO2 (with a certain d13C imprint, etc.) needs to be in order to reverse marine d13C gradients (this would depend on the region though). I think this result is there 'in between the lines/in the figures' especially in the results section on the PETM, but highlighting this individually could make your study useful for a broader audience.

=> We have revised in the main text as
"As a result, the naturally formed vertical gradients of δ^{13}C-DIC are eliminated or reversed towards the end of the 21st century. For example, the difference between the globally averaged surface and subsurface δ^{13}C-DIC becomes 0.2‰ for RCP2.6, –0.7 ‰ for RCP4.5, –1.6 ‰ for RCP6.0, and –3.8 ‰ for RCP8.5 as of 2100, compared to the preindustrial value of 1.6±0.2 ‰ (Figs. 1c and 3a). Such varying degrees of vertical gradient reversals indicate a large sensitivity of the gradient disruption to the rapidity of CO_2 emissions, given the present-day ocean circulation rates. Furthermore, the regional magnitude of gradient reversal is larger in strongly stratified low latitude ocean than convective high latitudes, suggesting a sensitivity to ocean ventilation state as well (Fig. 3a)."

6. Are you able to reduce the number of figures to fewer key figures to focus your storyline? Is it for example needed to have Fig 2c-f in the main text? Could you highlight e.g. only 2 or even one RCP scenario for clarity?

=> If necessary, we can move some figures (Fig 2c-f) to Supplementary Materials. As long as the length is okay, we would rather like to keep them as they are.
Because the four different RCP scenarios provide a primary source for uncertainty in future projection, it is important to show the results of all 4 scenarios. For an additional experiment using the CESM2 estimates in this revision, however, we have focused only on the RCP 6.0 scenario as the CESM2 estimates are based on a similar SSP3-7.0

scenario.

7. Availability model/data: Will you archive your model / model output / analysis scripts and other data to make your figures in an openly accessible repository?

=> We have added in acknowledgement as *"All of the model results presented here will be made available at https://climatedata.ibs.re.kr/data/papers/kwon-et-al-2021-commsenv and a public repository. The model code will be made available from the corresponding author on reasonable request."*

Specific Comments

L35 The 10000 GtC PETM release does not really compare to the RCP scenarios, which give an anthropogenic influx of about 500-2000 GtC until 2100? Why not release the 'actual' CO2 amount to the model?

=> The model used here is an ocean only model with atmospheric CO2 as a boundary condition. Hence, we can only prescribe atmospheric CO2 in the model rather than imposing CO2 release. This point is described in Methods.

Throughout the text: you mention 'surface' and 'subsurface' regularly, please define and repeat your definition for clarity. What living depths do your planktic and benthic forams represent, and do your definitions of 'surface' and 'subsurface' favor comparison with those?

E.g. L141-142 what is exactly done here?

=> We have added in the main text as *"below a depth of 74 m of the water column (referred to as "subsurface" hereinafter)".*

We have also added in Methods as

"We assume that the δ^{13}C values of mixed layer planktic foraminifera represent the surface δ^{13}C-DIC whereas the δ^{13}C values of epifaunal benthic foraminifera represent the δ^{13}C-DIC at the depths of the ODP sites. Those inferred δ^{13}C-DIC values are then compared with our model-based estimates averaged over 0-74 m depths for planktic and over 2-3 km depths for benthic."

We have also added the depths of the two ODP sites as *"For the ODP Site 690 (South Atlantic, 65°S, 1°E, 2100m deep)"* and *"For the ODP Site 1209 (North Pacific, 33°N, 159°E, 2387m deep)"* where the depths of the ODP sites represent the living depths of benthic foraminifera.

L84, L103-109 See the recent paper by Liu et al. (2021) who quantify an underestimation of the Eide et al. (2017) Suess effect per ocean basin.

=> We have added *"yet would be consistent if the previous estimate is corrected for the uncertainty of 0.15-0.24 ‰ suggested by the same study (Eide, Olsen, Ninnemann, & Eldevik, 2017) and a later independent study (Liu et al., 2021)."*

L94-100 Another noticeable feature of Fig 3b is the weakening of the latitudinal contrasts with increasing RCP scenario. Here, another point should be discussed I think: the perturbation ratio is also affected by the d13C disequilibrium. Equilibration with a d13Catm of -6.5 permil actually increases surface d13C_DIC in low-temperature low-d13C_DIC regions such as the upwelling areas due to the negative disequilibrium, i.e. d13C_surface is lower than d13C_equilibrium (Galbraith et al., 2015; Mor.e et al., 2018; Schmittner et al., 2013). In your scenario where the emissions lower atmospheric d13C even further, this pattern will change.
E.g. when atmospheric d13C decreases to the point that d13C_atm (e.g., -10 permil) + air-sea fractionation (e.g., +10 permil in cold waters) < d13C_DIC anywhere in the surface ocean, one gets a positive disequilibrium everywhere. The warming of surface waters will additionally decrease the thermodynamic air-sea fractionation, thereby contributing further to a global positive disequilibrium (and this is a spatially quite heterogeneous effect). Besides the aspects of disequilibrium and thermodynamic fractionation across the air-sea interface, bulk transfer of the additional low-d13C-CO2 into the ocean will only enter the ocean in areas of net CO2 uptake (this relates to your Revelle factor discussion). I am unsure how important dilution is here as compared to disequilibrium, bulk transfer and thermodynamic fractionation. Please discuss/clarify.

=> We have performed an additional experiment where we explored the effects of future changes in air-sea gas exchange rates and thermodynamic equilibrium fractionations, which now complement our previous discussions focusing on the effects of increasing atmospheric CO2. The results are discussed in the main text as

"Projected global warming (e.g., ref. (Rodgers et al., 2021)) is expected to further lower surface δ^{13}C-DIC through increasing air-sea CO_2 exchange rates in high latitudes (mostly due to sea ice melting) and enhanced thermodynamic isotopic fractionations whose effects are most pronounced in low latitudes (Supplementary Fig. 5; Supplementary Information). These warming driven surface δ^{13}C-DIC reductions can additionally elevate the ratio of the ocean to atmosphere ^{13}C Suess effect, and also enhance the vertical δ^{13}C-DIC gradient reversal as of 2100. When globally averaged, the warming effects are small with an additional surface δ^{13}C-DIC decline of only –0.1 ‰ compared to the geochemically driven ^{13}C Suess effect of –3.7 ‰ under the RCP6.0 scenario (Supplementary Fig. 4). Yet, regionally the effects can be as large as 50% in the Weddell Sea (Supplementary Fig. 5). A more comprehensive assessment of ocean stratification and circulation change effects on δ^{13}C-DIC projections remains a subject for future studies."

In Supplementary Materials, we have also added
"The combined effects from changing air-sea CO_2 exchange rates, SST, and SSS on the 21st century oceanic ^{13}C Suess effect are shown in Supplementary Figs. 4 and 5. Overall, the changing air-sea CO_2 exchange rates, SST, and SSS in a warming climate, has an impact on further decreasing surface δ^{13}C-DIC. The most pronounced differences of up to –0.4 ‰ as of 2100 are found at high latitudes where sea ice melting increases air-sea CO_2 transfer rates (Supplementary Figs. 5c and 5d). Increasing SST in a major fraction of the global ocean surface also enhances the thermodynamic equilibrium fractionation, whose effects are mostly pronounced in relatively well equilibrated subtropical gyres

(Supplementary Figs. 5e and 5f). The perturbation ratio of surface ocean δ^{13}C-DIC to atmospheric δ^{13}C-CO_2 also increases accordingly by up to 0.1. Nevertheless, the climate driven modulations of the oceanic ^{13}C Suess effect are an order of magnitude smaller than the geochemically driven ^{13}C Suess effect that ranges from –(1 to 5) ‰ spatially (Supplementary Figs. 5a and 5b). "

In the main text, we have also strengthened our discussion of Fig. 3b as *"As a result, the perturbation ratio (defined as the ratio of deviations from the respective preindustrial values) of surface δ^{13}C-DIC to atmospheric δ^{13}C-CO_2 exhibits a large latitudinal contrast depending on the oceanic uptake of CO_2 and the air-sea equilibrium states relative to vertical mixing rates (Kortzinger et al., 2003; McNeil et al., 2001). For example, as of 2000 the surface to atmospheric δ^{13}C perturbation ratio ranges from 0.1 in highly convective Southern Ocean to 0.7±0.1 in relatively stable Northern Hemisphere subtropical gyres (Fig. 3b), in agreement with a previous study (Eide, Olsen, Ninnemann, & Eldevik, 2017).*"

L99,L125/Fig 3b: I think it should be emphasized that the perturbation ratio is not just expected to be one and explain to the reader why (i.e. what it depends on and why it is of interest). The text under Fig. 8 in Eide et al. (2017) may be useful there.

=> Please see our response to the above comment.

L 119 I think it is important to refer not only to the global surface mean like in Fig 1b but also how it spatially develops (supplementary figure 6). Also in L 142-144 for example you could provide the reader with some detail about the spatial structure of these changes.

=> We have added *"Furthermore, the regional magnitude of gradient reversal is larger in strongly stratified low latitude ocean than convective high latitudes, suggesting a sensitivity to ocean ventilation state as well (Fig. 3a)."*

L148 I think it would help the reader to start this section with what your goals are with it and summarize its main conclusions and methods.

=> We have added to the beginning of the section as *"We elucidate the geochemical mechanisms by which the 21st century CO_2 emissions can drive such radical changes in surface δ^{13}C-DIC."* We have also made this section more succinct.

L 151-155 This sentence is difficult to read: I understand you want to provide the reader with a factor instead of a permil change in order to compare to DIC_ant, but maybe state the absolute change as well such that it compares easier to Fig 1b?

=> We have revised as *"For example, under the RCP8.5 the surface-averaged ^{13}C Suess effect increases 8 times from –0.8 ‰ in 2000 to –6.2 ‰ in 2100 (Fig. 1b), whereas the surface-averaged anthropogenic DIC increases 4 times from 40 μmol/kg to 201 μmol/kg over the same time period (Supplementary Fig. 8b)."*

L 155-156 As my previous comment: The factor two is not really visible directly in Fig 3c, maybe add some absolute values for clarity?

=> We have revised as *"The perturbation ratios of the surface δ^{13}C-DIC to DIC also increase regionally by up to a factor of two from –15 ‰/(mmol kg^{-1}) in 2000 to –30 ‰/(mmol kg^{-1}) in 2100 under the RCP8.5 (Fig. 3c)."*

L 158-160 Why does a stronger Suess effect response than DIC_ant mean it will penetrate less?

=> We have rephrased as *"The progressively amplified response of surface δ^{13}C-DIC, relative to surface DIC or pH, manifests as sharper vertical gradients for the ^{13}C Suess effect than anthropogenic DIC (McNeil et al., 2001)."*

L 164: A reader inexperienced with d13C and its definition would not follow your sudden change to ratios here. In fact, I think the introduction of this article should start with a short intro to what d13C can be/is used for and how your study uses your analysis of the drivers of future gradient reversal to understand PETM gradient changes.

=> We have added to Introduction as *"Anthropogenic CO_2 emissions have led to an accumulation of ^{13}C-depleted carbon in the atmosphere and the upper ocean (referred to as the ^{13}C Suess effect), disturbing naturally formed vertical gradients of δ^{13}C-DIC (where δ^{13}C is defined as $[(^{13}C/^{12}C)_{sample}/(^{13}C/^{12}C)_{standard} - 1]$ with the Vienna Pee Dee Belemnite standard) (Eide, Olsen, Ninnemann, & Eldevik, 2017; Keeling, 1979)."*

We have also revised Abstract and Introduction accordingly.

L246-256 This paragraph is more of a summary or introduction than a discussion, maybe move it to the introduction?

=> We have moved part of the paragraph to Abstract and Introduction.

L 297-298 here you for the first time state that the current excursion is faster, while in L 227 you seem to conclude that the duration an magnitude is similar. Is this a comparison of the 5 kyr to the current ~150 yr anthropogenic excursion? You repeat this statement of the contemporary change being faster than the PETM change in the abstract. It undermines your comparison between the projected reversal and the PETM. More importantly, I think it needs more arguments/explanation: stress how fast and large the current CO2 release and gradient change is, and how that is for the PETM?

=> We have clarified our comparison of d13C-DIC changes between the future and PETM by revising the last paragraph of the main text as

"Despite the analogy between the modern and PETM onset in terms of the large δ^{13}C-DIC excursion and the existence of vertical gradient reversal, the rate at which the 21st century anthropogenic carbon isotope excursion occurs is at least one order of magnitude faster than PETM excursion rates (Fig. 6c). A precise comparison of the time

rate of change is hampered due to the difficulty in reconstructing high temporal resolution PETM $\delta^{13}C$ records (e.g., refs. (Shaw et al., 2021; Westerhold et al., 2018)) and large uncertainties in age models (e.g., refs. (Bains et al., 1999; Thomas & Shackleton, 1996)). Nevertheless, the 21st century $\delta^{13}C$-DIC gradient reversal rates (taking only ~3 centuries from the preindustrial era to the projected gradient reversals) appear to be much faster than those of the PETM (taking at least 5 kyr from the pre-PETM to the maximum gradient reversal). Given the fact that the PETM is the best known ancient time when the most rapid carbon emission has been reported over the Cenozoic (e.g., refs. (Zachos et al., 2001; Zeebe et al., 2016)), our comparison suggests that the time rates of 21st century $\delta^{13}C$-DIC excursion and associated gradient reversal may be unprecedented over the Cenozoic. "

L 304 Please include the name of the model here

=> We have revised as *"an observationally constrained ocean circulation inverse model (OCIM) (DeVries, 2014)"*

L359 How about SST, was that fixed too? Why did you keep these aspects fixed when estimating the Suess effect in the industrial simulations? In your discussion you provide some more details on comparison with CMIP models – but they did not simulate 13C. I expect the effects of SST increase on the Suess effect are relevant for your study.

=> We have explored the effects of changing SST along with the effects of changing SSS, sea ice, and wind speed (See our response to a similar comment above).

We have also added to Methods as
"Using the full model setup, we assume that the ocean circulation, air-sea CO_2 exchange rates including sea ice effects, and sea surface temperature and salinity remain unchanged with time throughout the simulation. An uncertainty associated with this assumption is tested in Supplementary Materials where we relax the assumption of unchanged air-sea CO_2 exchange rates, and sea surface temperature and salinity using the Community Earth System Model version 2 (CESM2)-based estimates (Danabasoglu et al., 2020; Rodgers et al., 2021)."

L385 Here you suddenly mention 'The monte carlo experiment'. There are 1400 of them, rdid you pick one? Is the full model setup this fixed one monte carlo experiment? Does this mean that in all of the RCP scenario runs you have no change in air-sea gas exchange rates and SST (L 444-447 says that too)? Why not? I think to project marine d13C gradients it is important to include these.

=> Please see our response to the above comment.

L748 Fig 1a is not distinguishable for some color-blind readers. Style of 1a is also different from 1b,c. In 1b,c the figure would be clearer if the onset of the model experiment (2019) is indicated.

=> We have revised Fig. 1. The style difference is inevitable due to different approaches taken for the historical and future estimates, described in the main text and methods. We have also added in the figure caption as *"(b) Estimated oceanic and atmospheric ^{13}C Suess effects from 1950 to 2018 (gray-shaded background) are combined with the projected ^{13}C Suess effects from 2019 to 2100."*

L 829 what is 'the poorly ventilated region', the global deep ocean below a certain depth?

=> We have made them more specific as *"stagnant deep waters (i.e., the North Pacific in the present-day configuration).", "relatively well-ventilated regions (i.e., the North Atlantic in the present-day configuration)",* and *"deep (average over 2-3km depth)"*

L812/822 Fig. 6: I am not familiar with mbsf bins. I understand the x-axis show some sense of time through sediment depth (and you provide relative ages). The 2ky shift is not clear to me, aren't the model data already shifted from the future to the PETM anyways, by 56 million years? What is the uncertainty of the age model? Why did you choose these planktic species?

=> We have revised the figure and caption.
The X label is now named as *"Sediment depth (meters below the sea floor; msbf)"*. We have revised the figure caption as *"The positions of both dashed lines are shifted such that the positions for initial declining match between the foraminifera records and the simulation."*.

Regarding the uncertainty of age model, we noted as *"The relative age outside parentheses is based on Thomas and Shackleton (1996) while the relative age based on Bains et al. (1999) is shown inside parentheses."*

We did not choose any particular planktic species, they are simply the ones whose high resolution records are currently available for the PETM onset.

Supplementary Text
Carbon Cycle Formulations: Do you really simulate DI12C in the model as suggested by 'δ13C-DIC = [(DI13C/DI12C)sample/(DI13C/DI12C)standard -1]´103 ', or do you have total DIC and calculate DI12C using DIC-DI13C for sample? Could you add a few sentences on the model basics and not just refer to a supplementary of a different article (some of it can be repeated from the main text: resolution horizontal/vertical, how it compares to observations, what components are included, etc.)? Could you also shortly summarize how you set up the Monte Carlo experiments and list the exact parameters that you varied (f8, f9, etc.?). This is partly done in the main text but would be good to repeat here with additional details. From the caption of Fig S1 and the main text I get the impression that you ran 1400 experiments – this is not clear from the supplementary text?

=> We have revised in Methods *as " The carbon isotope model uses two prognostic variables of $DI^{13}C$ and $DI^{12}C$ (the latter approximated as DIC)".*

We have also added in Supplementary Information as

"Here, we present the details of model formulations and identify the model parameters that are varied in our Monte Carlo experiment. Specifically, we consider the following sources of uncertainty: (A) preindustrial $\delta^{13}C$ values for atmospheric CO_2, (B) thermodynamic equilibrium fractionation factors for air-sea CO_2 exchange and the historical changes in sea surface temperature, (C) the globally uniform $\delta^{13}C$ values of riverine carbon inputs of dissolved organic carbon (DOC), particulate organic carbon (POC), dissolved inorganic carbon (DIC), (D) the magnitude of non-riverine terrestrial carbon inputs, (E) the air-sea CO_2 exchange rates, (F) fractionation factors for the photosynthetic uptake of carbon, (G) ocean mixing and circulation states, and (H) the magnitude of inorganic carbon buried in marine sediments. Those are varied over the ranges summarized in Supplementary Table 1, and represented by the parameters f1-f10 as shown below. We note that the same model formulations are also presented in the Supplementary Materials of Kwon et al. (2021)."

For the model description such as the horizontal and vertical resolutions and the design of Monte Carlo experiment, we would like to refer readers to Methods.

Define 'SGD' at the end of page 2

=> We have changed it to *"submarine groundwater discharge"*

'f6 and f7 are chosen from three different values…' what values did you choose in the end then? Do you mean randomly chosen in the monte carlo experiments?

=> We have revised as *"the coefficients f6 and f7 are randomly chosen from three different sets suggested by Goericke and Fry (1994), Popp et al. (1989), and Freeman and Hayes (1992) (Supplementary Table 1)."*

'The δ13C endmember values for terrestrial carbon are chosen from the ranges of f8 = -27Å}2‰ for DOC, f9 = -35Å}2‰ for POC, and f10 = -15Å}2‰ for riverine DIC 27.' In the main text it is much clearer that the -25 is non-riverine d13C fluxes – please improve clarity here as well.

=> We have revised in Methods as *"the globally uniform $\delta^{13}C$ values of riverine carbon inputs of –27±2 ‰ for dissolved organic carbon (DOC), –30±2 ‰ for particulate organic carbon (POC), and –15±2 ‰ for DIC (Peterson & Fry, 1987), (D) the magnitude of non-riverine terrestrial carbon inputs with a $\delta^{13}C$ value of –26 ‰ (Maher et al., 2013), including uncertainties in groundwater driven fluxes (Szymczycha et al., 2014) and the carbon export from coastal vegetation (Duarte, 2017), that are assumed to be uniformly distributed along the coastal margins except around the Antarctica"*

We have also revised in Supplementary Information as *"The $\delta^{13}C$ endmember values for riverine carbon are chosen from the ranges of f8 = -27±2‰ for DOC, f9 = -30±2‰ for POC, and f10 = -15±2‰ for DIC (Marwick et al., 2015; Peterson & Fry, 1987). For the coastal margin carbon flux, the $\delta^{13}C$ endmember value is fixed at -26‰ (Abril* *et al., 2013; Maher et al., 2013). Although the $\delta^{13}C$ values for the riverine and coastal margin inputs are highly uncertain spanning -(14-30)‰ (Abril et al., 2013; Maher et al., 2013), its uncertainty is implicitly included in our Monte Carlo experiment because the effects on the $\delta^{13}C$-DIC are identical between the magnitude of the coastal margin inputs (f2) and the $\delta^{13}C$ values of the riverine or coastal margin inputs."*

'A suite of 10 ocean circulation fields … for each Ensemble member' Do you mean you created 10 circulation fields which you call ensembles? They differ only slightly in nutrients/SST/salinity? It would be good to e.g. give a range of e.g. AMOC strengths/global mean SST/SSS to quantify the differences between the setups. How do these 10 circulation fields relate to the 1400 experiments?

=> We have revised as
"A circulation field is also randomly selected from a suite of 10 ocean circulation fields (DeVries, 2014). With slightly different ocean mixing parameterizations and data-assimilation methods, the suite of circulation models is designed to cover the uncertainty of the present-day climatological mean ocean circulation within the inverse modeling framework (DeVries, 2014; DeVries & Primeau, 2011). The model density structure and circulation are very close across the circulation fields with Atlantic overturning rates of 20±1 Sv (1 Sv = 10^6 m^3/s), Southern Ocean overturning rates of 16±1 Sv, and Drake Passage transport of 151±3 Sv (DeVries & Holzer, 2019). Nonetheless, the deep ocean ventilation age (defined as the time that has elapsed since the water was last in contact with the atmosphere) averaged at 2-3 km depths ranges from 687 years to 777 years, which is roughly scaled with three different horizontal maxing coefficients (600 m^2/s, 1000 m^2/s, and 2000 m^2/s) imposed in the model. The slight differences in ocean circulation result in slight differences in ocean surface productivity and the subsequent remineralization, due to different supply rates of PO_4 and nutrient availability in the euphotic zone."

'A greater air-sea CO2 piston velocity leads to a greater depletion of the δ13C-DIC for the global ocean, through its influence on the oceanic uptake of anthropogenic CO2 and enhanced exchange rates of 13C/12C.' Does it lead to a greater depletion everywhere? Or does this depend on the local disequilibrium (see e.g. the Gas Fast/Gas slow experiment in Moree et al., 2018).

=> The sensitivity of 13C Suess effect to an increase in air-sea CO2 piston velocity gives decreases in δ13C-DIC everywhere. However, the sensitivity of preindustrial δ13C-DIC or industrial δ13C-DIC to an increase in air-sea CO2 piston velocity gives decreases in δ13C-DIC in low latitudes surface and increases in δ13C-DIC in high latitude surface, the pattern similar to Fig. 3 of Moree et al. Because the section focuses on the d13C-DIC changes (i.e., the 13C Suess effect) rather than d13C-DIC itself, we hope that our description is clear as we write *"A greater air-sea CO_2 piston velocity leads to a greater depletion of the $\delta^{13}C$-DIC for the global ocean, through its influence on the oceanic uptake of anthropogenic CO_2 and enhanced exchange rates of $^{13}C/^{12}C$."*

P 5, last sentence 'the other sources … (Supplementary fig. 1)' this is a very long sentence and difficult to follow. Could you rephrase?

=> We have rephrased as *"The other sources of uncertainties, including the terrestrial carbon inputs, ocean circulations, fractionation factors during photosynthesis and air-sea CO_2 exchange, and the fraction of terrestrial carbon buried into marine sediments, also contribute little to the estimated uncertainty in the oceanic ^{13}C Suess effect (Supplementary Fig. 1). On the other hand, ocean circulations are of the first order importance for the oceanic inventory of anthropogenic DIC."*

Supplementary Figure 5: It is confusing that you use different colors here for the RCP scenarios than in your other figures. Your reference 34 is about CMIP6 not CMIP5. Please clarify how you used CMIP5/6 (which models did you use, please provide their references to acknowledge their efforts). For benthic values, did you take the volume-weighted mean of the global bottom wet layer? In c and d 1850 would be in the upper right corner and 2100 in the lower left, right?

=> The figure is now moved to Supplementary Fig. 6. Although the figure has different colors from other figures, we have legends to guide the readers. We have also made the colors consistent within the Supplementary Fig. 6.

Our reference 34 presents results from both CMIP5 and CMIP6. Specifically, we took the multi-model averaged surface pH changes from the CMIP5 results of Table 4, and the multi-model averaged benthic pH changes from Table 5 of the reference. Accordingly, we have changed the legend of Supplementary Fig. 6b as e.g., "RCP2.6/SSP1-2.6 benthic".

We have also revised the figure caption as
"(b) Globally averaged simulated pH changes (anomalies of the 2080-2099 average relative to the 1870-1899 average) from the full model setup are compared with those from CMIP5 models (Bopp et al., 2013; Kwiatkowski et al., 2020) for the top 100m values and from CMIP6 models (Kwiatkowski et al., 2020) for the benthic averaged values. Open circles show the averages for the top 100m and crosses show the averages for the benthic grid cells (the bottom layer of the ocean model). The inter-model spreads are shown in error bars for the CMIP5/CMIP6 model estimates, although they are small and hence not visible. (c) Simulated surface-averaged ^{13}C Suess effects are compared with simulated surface-averaged pH changes from a model year of 1850 (dots in the upper right) to 2100 (dots in the lower left)."

For benthic values from our model, we took the volume weighted mean of the global bottom wet layer.

All averages in our work are based on volume-weighting. We believe that this is a common practice.

Supplementary Fig. 7: Here again the definition of subsurface and surface is very relevant. Do you follow the linear scaling of supplementary figure 3 here and then cap it off at 4 or 5 permil maximum change? Please provide some details of the slower model here as well (e.g AMOC/Drake passage strength as compared to the full model setup).

=> The figure is now moved to Supplementary Fig. 9. We have specified the depth of 2-3 km in the figure caption and the text. E.g., *"Color shading shows the temporal duration over which the averaged δ^{13}C-DIC at 2-3 km depths lies above the local surface δ^{13}C-DIC by more than 0.5 ‰. Contour lines show the averaged ventilation ages at 2-3 km depths."*

We have revised in Methods as

"Atmospheric δ^{13}C-CO_2 is assumed to linearly decrease from –6.5 ‰ at a model year of zero to either –10.5 ‰ and –11.5 ‰ at a model year of 5 kyr, which corresponds to the atmospheric CO_2 perturbation period."

"In both "rapid" and "slow" setups, we use two ocean circulation states: the present-day ocean circulation as in the full model setup and a circulation where an averaged deep ocean (2-3 km depth) ventilation age is 3 times larger at 2237 years compared to 758 years for the present-day ocean (See Supplementary Fig. 9 for the distribution of deep ocean ventilation age). The slow ocean ventilation state was previously named as "KL" model in Kwon et al. (2011), and has slower meridional overturning rates of 12 Sv (1 Sv = 10^6 m^3/s) for the North Atlantic Deep Water and 5 Sv for the Antarctic Bottom Water, compared to the present-day circulation model (DeVries & Holzer, 2019) of 20 Sv and 16 Sv, respectively."

Minor/Technical Comments
Throughout text: I think 'ocean circulations' even if you mean multiple states/realizations of the ocean circulation should always be 'ocean circulation'.

=> corrected.

L52 'the balance or imbalance': the interplay

=> done.

L53 'ocean circulations': 'ocean circulation

=> done.

L100 'studies': if only one study is cited, use 'study'.

=> done.

Use of 'planktonic' throughout the article and in Fig 6, please read Emiliani (1991).

=> done.

L276 remove 'paleo'

=> done.

L 278-282 Long sentence which is difficult to follow.

=> We have removed the sentence.

Check your figures for color-blind suitability please

=> We hope that all revised figures are color-blind suitable.

L 575 inouts -> ‘inputs’

=> done.

Thank you for such detailed reading and suggestions.

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22nd Dec 21

Dear Dr Kwon,

Your manuscript titled "Unprecedented Reversal of Oceanic d13C Depth Gradient due to 21st Century Greenhouse Gas Emissions" has now been seen by 3 of the original reviewers, and I include their comments at the end of this message. They continue to find your work of interest, two of the reviewers are satisfied with the revision. However, Reviewer #2 raises some concerns about the timing of the PETM and the duration of d13C gradient suppression in proxy data vs. model simulation.

We are very interested in the possibility of publishing your study in Communications Earth & Environment, but would first like to consider your responses to these concerns raised by Reviewer #2 and assess a revised manuscript before making a final decision on publication. We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter) and the completed checklist:
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We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Dr Sze Ling Ho
Editorial Board Member

Communications Earth & Environment

Dr Clare Davis
Associate Editor
Communications Earth & Environment

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Please refer to our data policies at http://www.nature.com/authors/policies/availability.html.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

I have read the authors rebuttals to my and two other reviewers comments, and the relevant portions of the revised manuscript. My concerns have been addressed satisfactorily and I suggest the authors and editor devote their attention to the satisfactory response to reviewer #2's comments.

Reviewer #2 (Remarks to the Author):

I reviewed a previous version of this manuscript, and I think the authors have done a good job with revisions based on feedback. The overall objectives are clearer and I found it easier to follow the descriptions of the model and experimental framework.

I have just a few remaining comments. The authors set up the possible PETM onset duration as ranging between 5 to 20 kyr, but this is not the most accurate summary of the published literature. Some of the cited references suggest a more rapid onset (around 3 kyr). Also, see the review: Turner, S.K., 2018. Constraints on the onset duration of the Paleocene–Eocene Thermal Maximum. Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 376(2130), p.20170082. I just suggest a slight rephrasing of the text where appropriate - I think the choice of experiment onset durations adequately demonstrates the authors’ objectives.

My second comment is that the authors could more directly address the contrast between model results for changes in d13C gradients from the preindustrial - year 2100 period versus across the PETM. It would be very useful to know how long the suppressed gradients persist in response to the RCP forcings. In the long-PETM style simulations displayed in Fig. 5, the suppressed gradients in panels (a) and (b) reestablish prior to the end of the experiment at 10 kyr. But, the few thousand year persistence of these suppressed gradients is feasible to imagine detecting in proxy records. In contrast, a suppression of a few centuries, if that is in fact the duration of the RCP effects, would be much less likely to be detected in a typical pelagic deep ocean sediment core. The authors conclude that the similarity between the PETM data and the modeled RCP response suggests a rapid onset for the PETM, but it is also interesting if very rapid carbon emissions, comparable to the RCPs, allows vertical gradients to reestablish on timescales shorter than what pelagic deep sea cores can capture.

Also, the authors should be aware that there is particular reason to believe that the Site 1209 record may be lacking thousands of years of the PETM onset (see Haynes, L.L. and Hönisch, B., 2020. The seawater carbon inventory at the Paleocene–Eocene Thermal Maximum. Proceedings of the National Academy of Sciences, 117(39), pp.24088-24095).

Finally, another model-data difference stems from the fact that the PETM drove a benthic foraminiferal extinction - a major factor in why there should be “a delayed…decline in benthic d13C” as the authors note on Line 308.

Line 70 - I suggest change to ‘a recent compilation’

Figure 6c - I’m not sure I understand the purpose of this panel. It’s really challenging to see the difference between the surface and deep ocean values because of the overlapping of lines so the takeaway about reduced gradients is not obvious.

Reviewer #3 (Remarks to the Author):

Dear Eun Young Kwon and colleagues,

Thank you for your revised manuscript and the clear investment made to improve it in response to the reviewers. I think the current version of the manuscript is of high quality and of interest to different research fields. A few final small comments are listed below.

Best wishes,

Anne Morée

P2, l49: excursions in the surface ocean/atmosphere/mean d13C-DIC? Please specify.
P6, l178: ‘remain relatively unchanged’? A one permil deep water change is definitely large for many d13C records (eg Ziegler et al their nice Fig. 2 or of course your own Fig 6). Please also refer to a figure here or in the next sentence. For which scenario is your -1 permil statement meant?
Fig 6 replace planktonic with planktic
Fig 6c nicely done although it would help the comparison to a and b if the figure size and design was more similar.

Ziegler, M., Diz, P., Hall, I. R., and Zahn, R.: Millennial-scale changes in atmospheric CO2 levels linked to the Southern Ocean carbon isotope gradient and dust flux, Nature Geoscience, 6, 457, 10.1038/ngeo1782, 2013.

Thank you for the constructive reviews. Please find our point-by-point responses below:

Reviewer #1 (Remarks to the Author):

I have read the authors rebuttals to my and two other reviewers comments, and the relevant portions of the revised manuscript. My concerns have been addressed satisfactorily and I suggest the authors and editor devote their attention to the satisfactory response to reviewer #2's comments.
=> Thank you

Reviewer #2 (Remarks to the Author):

I reviewed a previous version of this manuscript, and I think the authors have done a good job with revisions based on feedback. The overall objectives are clearer and I found it easier to follow the descriptions of the model and experimental framework.

I have just a few remaining comments. The authors set up the possible PETM onset duration as ranging between 5 to 20 kyr, but this is not the most accurate summary of the published literature. Some of the cited references suggest a more rapid onset (around 3 kyr). Also, see the review: Turner, S.K., 2018. Constraints on the onset duration of the Paleocene–Eocene Thermal Maximum. Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 376(2130), p.20170082. I just suggest a slight rephrasing of the text where appropriate - I think the choice of experiment onset durations adequately demonstrates the authors' objectives.
=> We have rephrased as between 3 kyr and 20 kyr

My second comment is that the authors could more directly address the contrast between model results for changes in d13C gradients from the preindustrial - year 2100 period versus across the PETM. It would be very useful to know how long the suppressed gradients persist in response to the RCP forcings. In the long-PETM style simulations displayed in Fig. 5, the suppressed gradients in panels (a) and (b) reestablish prior to the end of the experiment at 10 kyr. But, the few thousand year persistence of these suppressed gradients is feasible to imagine detecting in proxy records. In contrast, a suppression of a few centuries, if that is in fact the duration of the RCP effects, would be much less likely to be detected in a typical pelagic deep ocean sediment core. The authors conclude that the similarity between the PETM data and the modeled RCP response suggests a rapid onset for the PETM, but it is also interesting if very rapid carbon emissions, comparable to the RCPs, allows vertical gradients to reestablish on timescales shorter than what pelagic deep sea cores can capture.
=> Thank you for the insightful comments. We have performed multi-millennial simulations for hypothetical futures based on our extended RCP scenarios. The results are now presented in Fig. 6a and Supplementary Fig. 9, and discussed in the main text as

"Beyond the 21^{st} century, future evolutions of oceanic δ^{13}C-DIC are highly uncertain due to uncertainty in future atmospheric CO_2 changes and potential feedbacks from the ocean. Yet, our multi-millennial simulations for hypothetical futures, assuming constant

atmospheric CO_2 after a year of 2500 and also considering geochemical effects only, reveal that the magnitude and duration of the gradient reversals are sensitive to the atmospheric $\delta^{13}C$-CO_2 excursions and the local ventilation states of the ocean. The surface ocean $\delta^{13}C$-DIC might drop by up to ~2 ‰ under RCP2.6, ~4 ‰ under RCP4.5, and up to ~6 ‰ under RCP6.0, which outpace deep ocean $\delta^{13}C$-DIC decreases of similar magnitudes (Fig. 6a). The duration over which the gradient reversal persists ranges from none under RCP2.6 to a few millennia in poorly ventilated deep North Pacific under RCP6.0 and RCP8.5 (Fig. 6a; Supplementary Fig. 9).”

Also, the authors should be aware that there is particular reason to believe that the Site 1209 record may be lacking thousands of years of the PETM onset (see Haynes, L.L. and Hönisch, B., 2020. The seawater carbon inventory at the Paleocene–Eocene Thermal Maximum. Proceedings of the National Academy of Sciences, 117(39), pp.24088-24095).

Finally, another model-data difference stems from the fact that the PETM drove a benthic foraminiferal extinction - a major factor in why there should be “a delayed…decline in benthic d13C” as the authors note on Line 308.
=> In response to these and above comments, we have revised the statement regarding the future and PETM comparison as

“A precise comparison of the $\delta^{13}C$-DIC excursions between the future and the PETM onset is hampered due to the difficulty in reconstructing high temporal resolution PETM $\delta^{13}C$ records (e.g., refs. [25,41]) and large uncertainties in age models (e.g., refs. [42,43]). In particular, benthic foraminiferal extinction and $CaCO_3$ dissolution that might have led to data gaps during the PETM onset challenge a precise determination of the magnitude and duration of the vertical gradient reversal (e.g., ref. [18]). Century-scale reversal or elimination events that are likely to occur under some RCP scenarios would not be detected in a typical pelagic deep ocean sediment core.”

Line 70 - I suggest change to ‘a recent compilation’
=> done

Figure 6c - I’m not sure I understand the purpose of this panel. It’s really challenging to see the difference between the surface and deep ocean values because of the overlapping of lines so the takeaway about reduced gradients is not obvious.
=> We removed the previous Figure 6c and added a revised Figure 6a.

Reviewer #3 (Remarks to the Author):

Dear Eun Young Kwon and colleagues,

Thank you for your revised manuscript and the clear investment made to improve it in response to the reviewers. I think the current version of the manuscript is of high quality and of interest to different research fields. A few final small comments are listed below.

Best wishes,

Anne Morée
=> Thank you

P2, l49: excursions in the surface ocean/atmosphere/mean d13C-DIC? Please specify.
=> We have added "on the Earth's surface"

P6, l178: 'remain relatively unchanged'? A one permil deep water change is definitely large for many d13C records (eg Ziegler et al their nice Fig. 2 or of course your own Fig 6). Please also refer to a figure here or in the next sentence. For which scenario is your -1 permil statement meant?
=> We have revised as "Unlike the well-ventilated thermocline and relatively young North Atlantic Deep Water, the δ^{13}C-DIC values in deep waters remain relatively unchanged with a subsurface averaged ^{13}C Suess effect less than –1 ‰ as of 2100 (Figs. 1b and 1c), becoming the waters of the most enriched δ^{13}C-DIC."

The -1 permil statement applies to all of the RCP scenarios, which is apparent in the referred figures added in this revision.

Fig 6 replace planktonic with planktic
=> done

Fig 6c nicely done although it would help the comparison to a and b if the figure size and design was more similar.
=> Our revised Fig. 6a has similar size and design to other figures in Fig. 6.

Ziegler, M., Diz, P., Hall, I. R., and Zahn, R.: Millennial-scale changes in atmospheric CO2 levels linked to the Southern Ocean carbon isotope gradient and dust flux, Nature Geoscience, 6, 457, 10.1038/ngeo1782, 2013.

24th Jan 22

Dear Dr Kwon,

Thank you for revising and resubmitting your manuscript titled "Unprecedented Reversal of Oceanic d13C Depth Gradient due to 21st Century Greenhouse Gas Emissions". After considering your responses, I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

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Best regards,

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