nature portfolio

Peer Review File

Carbon emissions along divergent plate boundaries modulate icehouse-greenhouse climates

Corresponding Author: Dr Ben Mather

This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Communications Earth & Environment.

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

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Decision Letter:

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

Dear Dr Mather,

Your manuscript titled "Oceanic carbon sequestration and Earth's emissions modulate icehouse-greenhouse climates" has now been seen by 2 new reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make 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.

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

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), a tracked-changes version of the manuscript (as a PDF file) and the 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 **

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.

Please do not hesitate to contact us 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,

Alireza Bahadori, PhD
Associate Editor
Communications Earth & Environment
Consulting Editor
Communications Sustainability

EDITORIAL POLICIES AND FORMATTING

We ask that you ensure your manuscript complies with our editorial policies. Please ensure that the following formatting requirements are met, and any checklist relevant to your research is completed and uploaded as a Related Manuscript file type with the revised article.

Editorial Policy: Policy requirements (Download the link to your computer as a PDF.)

For Manuscripts that fall into the following fields:
• Behavioural and social science
• Ecological, evolutionary & environmental sciences
• Life sciences
An updated and completed version of our Reporting Summary must be uploaded with the revised manuscript
You can download the form here:
https://www.nature.com/documents/nr-reporting-summary.zip

Furthermore, please align your manuscript with our format requirements, which are summarized on the following checklist: Communications Earth & Environment formatting checklist

and also in our style and formatting guide Communications Earth & Environment formatting guide .

*** DATA: Communications Earth & Environment endorses the principles of the Enabling FAIR data project (http://www.copdess.org/enabling-fair-data-project/). We ask authors to make the data that support their conclusions available in permanent, publically accessible data repositories. (Please contact the editor if you are unable to make your data available).

All Communications Earth & Environment manuscripts must include a section titled "Data Availability" at the end of the Methods section or main text (if no Methods). More information on this policy, is available at http://www.nature.com/authors/policies/data/data-availability-statements-data-citations.pdf.

In particular, the Data availability statement should include:
- Unique identifiers (such as DOIs and hyperlinks for datasets in public repositories)
- Accession codes where appropriate
- If applicable, a statement regarding data available with restrictions
- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

DATA SOURCES: All new data associated with the paper should be placed in a persistent repository where they can be freely and enduringly accessed. We recommend submitting the data to discipline-specific, community-recognized repositories, where possible and a list of recommended repositories is provided at http://www.nature.com/sdata/policies/repositories.

If a community resource is unavailable, data can be submitted to generalist repositories such as figshare or Dryad Digital Repository. Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

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

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

1. The title “Oceanic carbon sequestration and Earth’s emissions modulate icehouse-greenhouse climates” refers to a

fundamental and well-acknowledged concept in Earth system science. While it effectively captures the broader relationship between volcanic outgassing and carbon sequestration, it doesn't seem to convey a novel research insight or specific finding. The title would be more impactful if it emphasized a specific aspect of the study, such as new mechanisms or temporal dynamics, to underscore the paper's unique contribution to the field.

2. One of the main findings in this study, mid-ocean ridge and rift CO2 degassing surpass arc CO2 emissions, sounds reasonable. First, the authors should refer original carbon geodynamics papers (Marty and Jambon, 1987, EPSL; Sano and Williams, 1996, GRL). Second, it is noted that their model did not take into account metamorphic CO2 degassing in non-volcanic settings such as collisional orogens. Several papers have emphasized the role of orogenic CO2 degassing in deep carbon release and its impacts on surface carbon cycle and thus climate change. Regarding this, watch a recent review by Zhang et al. (2024, NSR, https://doi.org/10.1093/nsr/nwae089), field-based studies on deep CO2 emissions from active collisional orogens such as the Himalayas and the Tibetan Plateau (Liu et al., 2024, CEE, https://doi.org/10.1038/s43247-024-01438-z; Becker et al., 2008, EPSL, https://doi.org/10.1016/j.epsl.2007.10.046), and thermodynamic modeling work by Nicoli et al. (2022, CG, https://doi.org/10.1016/j.chemgeo.2022.121025), Stewart & Penman (2024, PNAS, https://doi.org/10.1073/pnas.2401961121), and Tamang et al. (2024, JP, https://doi.org/10.1093/petrology/egae021). These studies should contribute to the numerical modeling of carbon cycle through deep time, which may alter the conclusion of this paper.

3. Note that some terms require further clarification. A few examples are given below.

Line 17. The "carbon recycling" in this sentence is ambiguous and may lead to confusion. Carbon recycling is not equivalent to carbon sequestration, which specifically refers to the long-term storage of carbon, for example in oceanic lithosphere or sediments.

Line 272-273. "Atmospheric degassing" is not precise word to describe the emissions of CO2 into Earth's atmosphere.

Reviewer #2 (Remarks to the Author):

In this manuscript, "Oceanic carbon sequestration and Earth's emissions modulate icehouse-greenhouse climates", Mather et al. reevaluate the role of carbon in/outgassing at different tectonic settings on long-term global climate variability through the Phanerozoic. While drawing upon previous work utilising palaeotectonic reconstruction to interrogate the deep carbon cycle, novel to this manuscript is the decoupling of arc emissions from crustal carbonate destabilisation and subducted carbon—principally as sedimentary carbonate—to assess arc emission contributions before the evolution of widespread biomineralisation at 120 Ma. To achieve this, the authors use petrological modelling to quantify carbon and water loss during subduction; these petrological models are subsequently coupled to their palaeotectonic reconstructions. By assessing the portion of subducted carbon that contributes to arc carbon emissions at the present day, the authors suggest that carbon outgassing at tectonic settings exceeds carbon influx during greenhouse climates, whereas the opposite is true for icehouse climates. For the timeframe before 120 Ma, the authors conclude that carbon emissions from divergent settings have a greater importance in controlling Earth's climate state than the arc-dominant present day.

I enjoyed reading and reviewing this manuscript, and I believe that the research presented is interesting, original, and suitable for Communications Earth & Environment. The outcomes and implications reported by this manuscript are of interest to a crossroads of earth scientists, including the wider deep carbon community.

However, I have several comments on the manuscript in its current form which I believe can be addressed with minor revisions. I would appreciate some consideration of forearc volatile sequestration in their model setup. At present, their model considers slab devolatilisation from the trench to 125 km, whereas it is recognised that mantle wedge forearcs may host significant proportions of carbon (e.g., Barry et al., 2019, Nature) and water (e.g., Peacock and Bostock, 2024, Can. J. Earth Sci.) that do not contribute to arc magmatism. Additionally, the closure of pores is likely to be substantial during shallow subduction, which would reduce the amount of water that contributes to the melting sub-arc mantle (e.g., Hacker, 2008, Geochem. Geophys. Geosyst.). I would appreciate the authors considering the effects of this into their model, either by considering a narrower depth range to assess sub-arc mantle wedge water concentration, or through a discussion of water loss before the slab reaches sub-arc depths. In addition, I highlight that the authors' description of the construction of their model, comprising multiple datasets, components, and parameterisations, could benefit significantly from further clarification, e.g., by including more (sub)figures and equations in their methods illustrating how slab water content is assessed.

I attach a copy of the annotated manuscript pdf to this review with specific comments enclosed; I hope that the authors find these comments constructive and helpful, and wish them well with the remainder of the review process.

Dr. Kevin Wong
University of Cambridge, 8 May 2025

References cited in review

Abers, G. A., van Keken, P. E., & Hacker, B. R. (2017). The cold and relatively dry nature of mantle forearcs in subduction zones. Nature Geoscience, 10(5), 333–337. https://doi.org/10.1038/ngeo2922
Barry, P. H., Moor, J. M. de, Giovannelli, D., Schrenk, M., Hummer, D. R., Lopez, T., Pratt, C. A., Segura, Y. A., Battaglia, A.,

Beaudry, P., Bini, G., Cascante, M., d'Errico, G., di Carlo, M., Fattorini, D., Fullerton, K., Gazel, E., González, G., Halldórsson, S. A., … Lloyd, K. G. (2019). Forearc carbon sink reduces long-term volatile recycling into the mantle. Nature, 568(7753), 487. https://doi.org/10.1038/s41586-019-1131-5
Cappelli, L., Wallace, P. A., Randazzo, A., Kamau, P. M., Njoroge, R. W., Otieno, V., Tubula, M. S., Mariita, N. O., Mangi, P., & Fontijn, K. (2023). Diffuse soil CO2 emissions at rift volcanoes: Structural controls and total budget of the Olkaria Volcanic Complex (Kenya) case study. Journal of Volcanology and Geothermal Research, 107929. https://doi.org/10.1016/j.jvolgeores.2023.107929
Clift, P. D. (2017). A revised budget for Cenozoic sedimentary carbon subduction. Reviews of Geophysics, 55(1), 97–125. https://doi.org/10.1002/2016RG000531
Foley, S. F., & Fischer, T. P. (2017). An essential role for continental rifts and lithosphere in the deep carbon cycle. Nature Geoscience, 10(12), 897–902. https://doi.org/10.1038/s41561-017-0002-7
Gibson, S. A., & McKenzie, D. (2023). On the role of Earth's lithospheric mantle in global volatile cycles. Earth and Planetary Science Letters, 602, 117946. https://doi.org/10.1016/j.epsl.2022.117946
Halpaap, F., Rondenay, S., Perrin, A., Goes, S., Ottemöller, L., Austrheim, H., Shaw, R., & Eeken, T. (2019). Earthquakes track subduction fluids from slab source to mantle wedge sink. Science Advances, 5(4), eaav7369. https://doi.org/10.1126/sciadv.aav7369
Hauri, E. H., Maclennan, J., McKenzie, D., Gronvold, K., Oskarsson, N., & Shimizu, N. (2017). CO2 content beneath northern Iceland and the variability of mantle carbon. Geology, 46(1), 55–58. https://doi.org/10.1130/G39413.1
Hunt, J. A., Zafu, A., Mather, T. A., Pyle, D. M., & Barry, P. H. (2017). Spatially Variable CO2 Degassing in the Main Ethiopian Rift: Implications for Magma Storage, Volatile Transport, and Rift-Related Emissions. Geochemistry, Geophysics, Geosystems, 18(10), 3714–3737. https://doi.org/10.1002/2017GC006975
Lee, H., Muirhead, J. D., Fischer, T. P., Ebinger, C. J., Kattenhorn, S. A., Sharp, Z. D., & Kianji, G. (2016). Massive and prolonged deep carbon emissions associated with continental rifting. Nature Geoscience, 9(2), 145–149. https://doi.org/10.1038/ngeo2622
Merdith, A. S., Daniel, I., Sverjensky, D., Andreani, M., Mather, B., Williams, S., & Vitale Brovarone, A. (2023). Global Hydrogen Production During High-Pressure Serpentinization of Subducting Slabs. Geochemistry, Geophysics, Geosystems, 24(10), e2023GC010947. https://doi.org/10.1029/2023GC010947
Muirhead, J. D., Fischer, T. P., Oliva, S. J., Laizer, A., van Wijk, J., Currie, C. A., Lee, H., Judd, E. J., Kazimoto, E., Sano, Y., Takahata, N., Tiberi, C., Foley, S. F., Dufek, J., Reiss, M. C., & Ebinger, C. J. (2020). Displaced cratonic mantle concentrates deep carbon during continental rifting. Nature, 582(7810), Article 7810. https://doi.org/10.1038/s41586-020-2328-3
Peacock, S. M., & Bostock, M. G. (2024). Serpentinization of the forearc mantle wedge in subduction zones: Revisiting Roy D. Hyndman's seminal contributions 25 years later. Canadian Journal of Earth Sciences, cjes-2024-0082. https://doi.org/10.1139/cjes-2024-0082
Vitale Brovarone, A., Sverjensky, D. A., Piccoli, F., Ressico, F., Giovannelli, D., & Daniel, I. (2020). Subduction hides high-pressure sources of energy that may feed the deep subsurface biosphere. Nature Communications, 11(1), Article 1. https://doi.org/10.1038/s41467-020-17342-x
Vitale Brovarone, A., Wong, K., Giovannelli, D., de Pins, B., Gaillard, F., Massuyeau, M., Nestola, F., Pamato, M. G., & Daniel, I. (2025). Forms and fluxes of carbon: Surface to deep. In A. Anbar & D. Weis (Eds.), Treatise on Geochemistry (Third edition) (Vol. 2, pp. 647–698). Elsevier. https://doi.org/10.1016/B978-0-323-99762-1.00142-X
Werner, C., Fischer, T. P., Aiuppa, A., Edmonds, M., Cardellini, C., Carn, S., Chiodini, G., Cottrell, E., Burton, M. R., Shinohara, H., & Allard, P. (2019). Carbon Dioxide Emissions from Subaerial Volcanic Regions: Two decades in review. In B. Orcutt, R. Dasgupta, & I. Daniel (Eds.), Deep Carbon: Past to Present. Cambridge University Press.
Williams, S. E., & Gubbins, D. (2019). Origin of Long-Wavelength Magnetic Anomalies at Subduction Zones. Journal of Geophysical Research: Solid Earth, 124(9), 9457–9473. https://doi.org/10.1029/2019JB017479
Wilson, C. R., Spiegelman, M., van Keken, P. E., & Hacker, B. R. (2014). Fluid flow in subduction zones: The role of solid rheology and compaction pressure. Earth and Planetary Science Letters, 401, 261–274. https://doi.org/10.1016/j.epsl.2014.05.052

Communications Earth & Environment is committed to improving transparency in authorship. As part of our efforts in this direction, we are now requesting that all authors identified as 'corresponding author' create and link their Open Researcher and Contributor Identifier (ORCID) with their account on the Manuscript Tracking System prior to acceptance. ORCID helps the scientific community achieve unambiguous attribution of all scholarly contributions. You can create and link your ORCID from the home page of the Manuscript Tracking System by clicking on 'Modify my Springer Nature account' and following the instructions in the link below. Please also inform all co-authors that they can add their ORCIDs to their accounts and that they must do so prior to acceptance.
https://www.springernature.com/gp/researchers/orcid/orcid-for-nature-research

For more information please visit http://www.springernature.com/orcid

If you experience problems in linking your ORCID, please contact the Platform Support Helpdesk.

Version 1:

Decision Letter:

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

Dear Dr Mather,

Your revised manuscript titled "Carbon emissions along divergent plate boundaries modulate icehouse-greenhouse climates" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment, provided you state clearly that you've not taken metamorphic CO2 into account, fully explain the reasons for this omission, outline the resulting limitations, and tone down the conclusions accordingly.

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

EDITORIAL REQUESTS:

Please review our specific editorial comments and requests regarding your manuscript in the attached "Editorial Requests Table".

*****Please take care to match our formatting and policy requirements. We will check revised manuscript and return manuscripts that do not comply. Such requests will lead to delays. *****

Please outline your response to each request in the right hand column. Please upload the completed table with your manuscript files as a Related Manuscript file.

If you have any questions or concerns about any of our requests, please do not hesitate to contact me.

SUBMISSION INFORMATION:

In order to accept your paper, we require the files listed at the end of the Editorial Requests Table; the list of required files is also available at https://www.nature.com/documents/commsj-file-checklist.pdf .

OPEN ACCESS:

Communications Earth & Environment is a fully open access journal. Articles are made freely accessible on publication. For further information about article processing charges, open access funding, and advice and support from Nature Portfolio, please visit https://www.nature.com/commsenv/open-access

At acceptance, you will be provided with instructions for completing the open access licence agreement on behalf of all authors. This grants us the necessary permissions to publish your paper. Additionally, you will be asked to declare that all required third party permissions have been obtained, and to provide billing information in order to pay the article-processing charge (APC).

Please use the following link to submit the above items:
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 **

We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Alireza Bahadori, PhD
Senior Editor
Communications Earth & Environment
Consulting Editor
Communications Sustainability

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

I recommended that the metamorphic CO2 degassing should be taken into account. However, the authors refuted to do so in the second paragraph of P by P responses. They give the reasons (1) it is a minor component (Kerrick & Caldeira, 1999), (2) it is comparable to other source (Pradhan & Sen, 2024). Even though the old paper (1) suggested minor, new paper (2) said comparable, which means not negligible. Finally, they did not calculate the metamorphic one. I cannot agree with them and suggest that the paper should be returned.
In more details, they stated that in collisional orogens, metamorphic CO2 release is largely compensated by silicate weathering, as concluded in an earlier study. However, if they only consider volcanic carbon sources versus total carbon sinks, the portion of CO2 sinks compensated by metamorphic CO2 should be subtracted; otherwise, the carbon sinks would be overestimated in the carbon balance assessment.
They argued that the Cenozoic climate cooling could deny the role of collisional orogens as a major CO2 emitter. However, deep CO2 emissions (volcanic + metamorphic) are regulated not only by carbonate precipitation but also by organic carbon burial. Therefore, it is unreasonable to dismiss metamorphic CO2 by comparing it directly with the total carbon sinks (carbonate precipitation + organic carbon burial), since it represents only one part of the deep CO2 sources. Hence, their argument that “if metamorphic CO2 were important, the Cenozoic climate would be warmer than observed” is not convincing.

Reviewer #2 (Remarks to the Author):

After reading through the latest version of the manuscript by Mather et al., “Carbon emissions along divergent plate boundaries modulate icehouse-greenhouse climates”, I am satisfied that most of my requested revisions have been made by the authors, and I thank the authors for their time and effort in revising this manuscript according to both my comments and those of Reviewer 1. Below I briefly flag up some minor comments on the revised manuscript which should be straightforward for the authors to address.
I wish the authors the best with the remainder of the publication process.
Kevin Wong
Carnegie Institution for Science
23 October 2025

Lines 240–241: “The present-day rift degassing is skewed towards rifts with active plume volcanism and cratonic lithosphere involvement”. The authors here cite Brune et al. (2017, Nat. Geosci.) which does not mention plume volcanism nor cratonic lithosphere. Perhaps the recent review by Brune et al. (2023, Nat. Rev. Earth Environ.) is more appropriate here.
Methods Line 59: Missing word: “We only include chemically-bound water [in our] calculation…”
Methods Line 63: More of a broad comment, and feel free to address this briefly in your methods if you want, but how do thermal parameters (Φ) of different subduction systems vary over time, and what effect they may have on your study? For example, Cascadia at the present day has a low thermal parameter (van Keken et al., 2011, J. Geophys. Res. Solid Earth) and experiences significant slab devolatilisation at shallow pressures, evident from a highly serpentinised mantle wedge (e.g., Abers et al. 2017, Nat. Geosci.; Peacock and Bostock, 2024, Can. J. Earth Sci.). Of course, slab volatile release is not necessarily always correlated with thermal parameter; the subducting slab entering the Nankai trough also has a low thermal parameter but there is less evident serpentinisation from forearc metamorphic fluid release. It would nonetheless be interesting to first-order estimate the degree of ‘chemically-bound’ water lost from these low thermal parameter slabs in your model before 125 km depth.
Methods Line 258: Typo: “Perplex” to “Perple_X”
Methods Line 278: Reference to “mean global temperature” missing.
Data: Suggesting again it would be helpful to include water fluxes alongside the carbon fluxes.

Abers, G. A., van Keken, P. E., & Hacker, B. R. (2017). The cold and relatively dry nature of mantle forearcs in subduction zones. Nature Geoscience, 10(5), 333–337. https://doi.org/10.1038/ngeo2922
Brune, S., Kolawole, F., Olive, J.-A., Stamps, D. S., Buck, W. R., Buiter, S. J. H., Furman, T., & Shillington, D. J. (2023). Geodynamics of continental rift initiation and evolution. Nature Reviews Earth & Environment, 1–19. https://doi.org/10.1038/s43017-023-00391-3
Peacock, S. M., & Bostock, M. G. (2024). Serpentinization of the forearc mantle wedge in subduction zones: Revisiting Roy D. Hyndman’s seminal contributions 25 years later. Canadian Journal of Earth Sciences, cjes-2024-0082. https://doi.org/10.1139/cjes-2024-0082
van Keken, P. E., Hacker, B. R., Syracuse, E. M., & Abers, G. A. (2011). Subduction factory: 4. Depth-dependent flux of H2O from subducting slabs worldwide. Journal of Geophysical Research: Solid Earth, 116(B1). https://doi.org/10.1029/2010JB007922

href="http://www.nature.com/authors">www.nature.com/authors for information about policies, services and author benefits**

Response to reviewer comments

We have significantly improved the manuscript in line with the reviews. In particular, we have acknowledged in the text where some uncertainties exist in our reconstruction of carbon fluxes – the thickness of carbonate platforms. We have also rewritten the abstract and introduction sections to more clearly define the background and aims of the study. We thank the reviewers for their helpful suggestions and believe the manuscript is in much better shape thanks to their insights.

Our responses to the reviewers' comments below are highlighted in blue. Please also find a track changes document attached to this submission.

Reviewer #1

I have now read several times the new version of the manuscript of Mather et al. entitled "Oceanic carbon sequestration and Earth's emissions modulate icehouse-greenhouse climates". This manuscript is very interesting and I recognize that the manuscript has greatly improved, and it is now much easier to read. However, I still have some major concerns regarding the main text which include:

- In the abstract, when the authors mention volcanic arcs they should specify whether they mean island or continental arcs or both for clarity.

> We have clarified in the abstract that we are examining degassing along island AND continental arcs.

- A main limitation of their method is the fact that the authors do not consider the thickness of the carbonate platforms in their model, which could result in underestimating the CO2 fluxes released by continental arcs. I understand that reconstructing the thickness of carbonate platforms is challenging and a lot of the platforms that existed during the Phanerozoic are probably now eroded; but the authors should acknowledge the fact that they are only estimating a lower estimate, and not an upper estimate as they state L. 86. As such, there is a possibility that more CO2 could be outgassed by continental arcs than assumed by the authors. How would this affect their results? Perhaps the authors could use the estimates of Cao et al. https://www.sciencedirect.com/science/article/abs/pii/S0012821X1500343X to refine their modeling?

> We agree that our estimate of carbon outgassing from carbonate platforms could be too small, especially if predominantly thick carbonate platforms release significant quantities of carbon into the atmosphere. Conversely, our estimate of carbon outgassing from carbonate platforms may be too high because we assume carbonate platforms withstand tectonic destruction and do not become depleted of carbon through time. We have clarified these competing factors in a paragraph at the end of the first section, "Volcanic arc emissions from carbonate platforms".
>
> Lines 90-101: "A limitation of our study is that we do not consider the thickness of carbonate platforms in our calculations. Estimating carbonate platform thickness through time is challenging owing to highly uncertain carbonate accumulation rates, which are sensitive to fluctuations in sea level and variations in sedimentation/erosion rates(Dutkiewicz et al., 2019). If it follows that thicker platforms release more carbon into the atmosphere, and vice-versa, then our reconstructions of carbonate platform emissions would deviate from Figure 1a. On one hand, more carbon could be outgassed by continental arcs due to the subduction of thicker carbonate platforms; on the other hand, less carbon could be outgassed because we assume in our calculation that carbonate platforms that have been mapped withstand tectonic destruction throughout the Phanerozoic, and do not experience time-dependent carbon depletion. It is difficult to reconcile these two competing factors, nevertheless, our estimate of carbonate platform emissions are calibrated against present-day arc estimates(Bekaert et al., 2021; Fischer et al., 2019), and roughly agrees with other reconstructions of carbonate platform emissions through time (Supplementary Figure 2)."
>
> We have also been careful to include min-mean-max ranges in our carbon degassing estimates to reflect the level of uncertainty in the empirical relationships that govern carbon influx and outflux from various carbon reservoirs in the Earth. We thank the reviewer for their suggestion.

- Are icehouse climates linked to a decrease in CO2 fluxes emitted by MOR or to an increase in the subduction influxes? As I understand, most MOR will be subducted at some point. Wouldn't this imply to have more CO2 being subducted over time, which would drive to an icehouse climate?

> Icehouse climates are, indeed, linked to both a decrease in MOR outflux and an increase in subduction influxes. The last section of the paper explores how the balance between atmospheric outflux and oceanic plate influx drives major climate trends (Figure 4). We note that the oceanic plate influx is calculated by subtracting the subduction influx from the oceanic plate influx (i.e. balancing how much

carbon is removed from the surface vs. how much is added, lines 114-115). Thus we do implicitly account for any subducting MOR segments in our calculations.

- I understand that the authors used the P-T path of several slabs to infer how much carbon would be released during slab decarbonation. Did they consider end-members between warm and cooler slabs? This is essential as warmer slab P-T paths would likely result in larger CO2 fluxes released by arc volcanoes. The authors consider that only 22% of the subducted carbon is released beneath all arcs regardless their slab thermal state and their diversity in terms of subduction inputs and C outputs…, is this number realistic? How does this compare to modern day C fluxes?

Indeed, we consider the temperature of the slab as one of several parameters to calculate how much carbon is released during slab decarbonation. The details of this calculation are specified in the methods section under "slab devolatilization at sub-arc depths". Principally, we use the age of subducting oceanic crust to estimate the thermal structure of the subducting slab. This is calculated for every individual subduction zone segment across the globe and integrated by trench length to obtain the global slab carbon outflux for a given timestep.

We arrive at ~22% because of the total 59-78 Mt C/a which is released from the slab, only 11-18 Mt C/a is estimated to be emitted at the surface at present-day (Bekaert et al., 2021). The remainder is embedded in the overriding plate or circulated within the upper mantle (lines 131-137). We use this number to calibrate the arc emissions from slab devolatilization back through time, which assimilates the thermal state of the slabs, their volatile-carrying capacity, and devolatilization rates informed by thermodynamics.

- Are the higher Sr isotopic ratios shown in the figures related to higher erosion rates of MOR basalts to counterbalance global warmings?

Higher seawater Sr isotope ratios are associated with the weathering of silica-rich minerals. Mafic weathering acts to bring down the Sr isotope ratio (lines 199-203). The fluctuations in the seawater Sr curve is primarily related to (1) the mid-ocean ridge length, which brings down the Sr ratio due to increased mafic weathering (van der Meer et al., 2017), and (2) the length of continental arcs which are clustered in the tropics where erosion rates are higher (McKenzie et al., 2016).

- Did the authors consider the release of C during slab melting? Slab melting has been suggested to play a central role in releasing C beneath volcanic arcs.
https://www.nature.com/articles/nature16174

Yes, we follow the release of carbon and water from the slab, which result from changes in mineral phases under increasing pressure and temperature. The thermodynamic models find stable mineral phases over a specified pressure and temperature range via Gibbs free energy minimisation using the thermodynamic database of (Holland & Powell, 1998). Each mineral phase may store a different concentration of chemically-bound water and carbon as shown in Supplementary Figure 4. We compute the P-T conditions of the down-going plate at sub-arc depths (< 125 km) based on the age and dip angle of the slab. From there we look-up the concentration of water and carbon that is stable within the slab at those P-T conditions and subtract it from the initial concentration of water and carbon immediately prior to subduction. This number represents the total reduction in slab water and carbon concentration from the trench to sub-arc depths (< 125 km). The slab outflux is then obtained by multiplying by the density and integrating the along trench length and convergence rate. This routine is repeated for the three major volatile-carrying reservoir in the slab: (1) deep sea sediments, (2) altered oceanic crust, and (3) serpentinized/metasomatized mantle lithosphere. We have amended the methods section to more clearly articulate this methodology.

We compute P-T lookup tables up to 9 GPa and 1700 K, which equate to sub-arc depths where the vast majority of carbon and water are released from the slab to promote arc volcanism. The paper cited by the reviewer pertains to melting slabs acting as a barrier to deep mantle carbon subduction. We only track the volatiles released from the slab which get released along volcanic arcs. We do not track what happens to any remaining portion of volatiles or the release of more volatiles from the slab as it descends deeper into the mantle. Whether these volatiles interact with the Earth's climate is very uncertain and likely to play a very minor role compared to slab devolatilization along volcanic arcs.

Reviewer #2

Review of the revised version of the paper by Mather et al. "Oceanic carbon sequestration and Earth's emissions modulate icehouse-greenhouse climates"

By reviewing this revised version, I feel that no significant changes have been made to the manuscript. What I did not like in the first version is still here.

The summary is still not well structured and the main scientific question is still not clearly outlined. https://www.nature.com/documents/nature-summary-paragraph.pdf

> We have significantly overhauled the abstract following the Nature summary paragraph guidelines.

To me, the introduction also needs rewriting. After dealing with the main scientific question, the authors should point out what is missing so that the effort made in the study is a clear consequence of this missing information. Here the transition between the main intro (lines 23-45) and the aim (45-49) is not well accomplished. If, for example, the extension of the model from 250 to 540 Ma is new, then this should also appear in the intro and the aim of the study.

> We have also rewritten the introduction and changed its structure. We have bridged the background section with the study aim more seamlessly and emphasised the extension of the model from 250 to 540 Ma for MOR degassing.

The author should keep in mind that they might potentially be read by a large audience. The phrasing "carbonate platform degassing" (lines 52-53) is still in use with the definition appearing several lines after its first occurrence (lines 60-61). The authors should clearly explain this mechanism to the reader before going on.

> We now define “carbonate platform degassing” in the introduction section on line 35.

In figure 2, the authors introduce a new legend about active and inactive carbonate platforms. What do they mean by active and inactive? Does this change the CO2 cycle through time in their model? Please explain.

> Active carbonate platforms indicate platforms which are actively growing at the time of the reconstruction. This does not change how carbon is released from carbonate platforms in our model. We have amended the figure caption to explain that active and inactive carbonate platforms are treated the same in our model.

The authors cite Supp Fig 4 before Supp Fig 3. Please change the order.

> Fixed.

The paragraph in lines 133-141 is essentially a repetition (no significant deep-sea carbonates before 120 Ma) of the previous paragraph. Please rewrite.

> We have reframed the sentence to focus on how our prediction of volcanic arc degassing improves the current state of the art.

The authors do not take into account the CO2 emitted by plumes (line 227). When large volumes are emitted (e.g. Deccan traps) does this change their result significantly?

> We do not model carbon degassing from plume-related volcanism for multiple reasons. Foremost, it is difficult to know how long plumes have been active through geological time and how many there have been. Even at present day there is some doubt over how many plumes exist (Malamud & Turcotte, 1999). Furthermore, it is unclear how the plume buoyancy flux (how efficient the plume is at transporting material to the surface) varies through time. Some pulses in plume activity do exist – the Deccan traps are a good example highlighted by the reviewer – but there are likely other pulses of high plume activity that are less well preserved. Lastly, it is unclear how much carbon plumes transport to the atmosphere via volcanism, and whether plumes underneath continental regions release more carbon than oceanic plumes.

The repeated listing of CO2 flux values makes the reading boring. Could this be replaced by a figure or a table?

> These numbers can already be read from the figures, we simply provide them here in case the reader wants a more precise estimate of CO2 flux values. We also provide spreadsheets in the supplementary section which can be downloaded alongside the paper.

After all these negative remarks, I still feel that the paper is important and should be published in Nature Geoscience. I encourage the authors to revise their manuscript.

A few typos :
Line 35: approximatley
Line 196: atmorspheric

Fixed.

References

Bekaert, D. V., Turner, S. J., Broadley, M. W., Barnes, J. D., Halldórsson, S. A., Labidi, J., et al. (2021). Subduction-Driven Volatile Recycling: A Global Mass Balance. *Annual Review of Earth and Planetary Sciences*, *49*(1). https://doi.org/10.1146/annurev-earth-071620-055024

Brune, S., Williams, S. E., & Müller, R. D. (2017). Potential links between continental rifting, CO2 degassing and climate change through time. *Nature Geoscience*, *10*(12), 941–946. https://doi.org/10.1038/s41561-017-0003-6

Clift, P. D. (2017). A revised budget for Cenozoic sedimentary carbon subduction. *Reviews of Geophysics*, *55*(1), 97–125. https://doi.org/10.1002/2016RG000531

Connolly, J. A. D. (2009). The geodynamic equation of state: What and how. *Geochemistry Geophysics Geosystems*, *10*(10), 1–19. https://doi.org/10.1029/2009GC002540

Dutkiewicz, A., Müller, R. D., Cannon, J., Vaughan, S., & Zahirovic, S. (2019). Sequestration and subduction of deep-sea carbonate in the global ocean since the Early Cretaceous. *Geology*, *47*(1), 91–94. https://doi.org/10.1130/G45424.1

Fischer, T. P., Arellano, S., Carn, S., Aiuppa, A., Galle, B., Allard, P., et al. (2019). The emissions of CO2 and other volatiles from the world's subaerial volcanoes. *Scientific Reports*, *9*(1), 1–11. https://doi.org/10.1038/s41598-019-54682-1

Holland, T. J. B., & Powell, R. (1998). An internally consistent thermodynamic data set for phases of petrological interest. *Journal of Metamorphic Geology*, *16*(3), 309–343. https://doi.org/10.1111/j.1525-1314.1998.00140.x

Malamud, B. D., & Turcotte, D. L. (1999). How many plumes are there? *Earth and Planetary Science Letters*, *174*(1–2), 113–124.

Mason, E., Edmonds, M., & Turchyn, A. V. (2017). Remobilization of crustal carbon may dominate volcanic arc emissions. *Science*, *294*(July), 290–294. https://doi.org/10.1126/science.aan5049

McKenzie, N. R., Horton, B. K., Loomis, S. E., Stockli, D. F., Planavsky, N. J., & Lee, C.-T. A. (2016). Continental arc volcanism as the principal driver of icehouse-greenhouse variability. *Science*, *352*(6284), 444–447. https://doi.org/10.1126/science.aad5787

van der Meer, D. G., van den Berg van Saparoea, A. P. H., van Hinsbergen, D. J. J., van de Weg, R. M. B., Godderis, Y., Le Hir, G., & Donnadieu, Y. (2017). Reconstructing first-order changes in sea level during the Phanerozoic and Neoproterozoic using strontium isotopes. *Gondwana Research*, *44*, 22–34. https://doi.org/10.1016/j.gr.2016.11.002

Müller, R. D., Mather, B., Dutkiewicz, A., Keller, T., Merdith, A., Gonzalez, C. M., et al. (2022). Evolution of Earth's tectonic carbon conveyor belt. *Nature*, *605*(7911), 629–639. https://doi.org/10.1038/s41586-022-04420-x

Zhong, X., & Galvez, M. E. (2022). The Subducting Slab as a Chromatographic Column: Regimes of Sub-Solidus Mass Transport as a Function of Lithospheric Hydration State, With Special Reference to the Fate of Carbonate. *Journal of Geophysical Research: Solid Earth*, *127*(6), e2021JB023851. https://doi.org/10.1029/2021JB023851

Reviewer #1

1. The title “Oceanic carbon sequestration and Earth’s emissions modulate icehouse-greenhouse climates” refers to a fundamental and well-acknowledged concept in Earth system science. While it effectively captures the broader relationship between volcanic outgassing and carbon sequestration, it doesn’t seem to convey a novel research insight or specific finding. The title would be more impactful if it emphasized a specific aspect of the study, such as new mechanisms or temporal dynamics, to underscore the paper’s unique contribution to the field.

> The title was changed in a previous revision to satisfy a reviewer who thought the title was too specific. We have changed it back in this version to “Carbon emissions along divergent plate boundaries modulate icehouse-greenhouse climates”.

2. One of the main findings in this study, mid-ocean ridge and rift CO2 degassing surpass arc CO2 emissions, sounds reasonable. First, the authors should refer original carbon geodynamics papers (Marty and Jambon, 1987, EPSL; Sano and Williams, 1996, GRL). Second, it is noted that their model did not take into account metamorphic CO2 degassing in non-volcanic settings such as collisional orogens. Several papers have emphasized the role of orogenic CO2 degassing in deep carbon release and its impacts on surface carbon cycle and thus climate change. Regarding this, watch a recent review by Zhang et al. (2024, NSR, https://doi.org/10.1093/nsr/nwae089), field-based studies on deep CO2 emissions from active collisional orogens such as the Himalayas and the Tibetan Plateau (Liu et al., 2024, CEE, https://doi.org/10.1038/s43247-024-01438-z; Becker et al., 2008, EPSL, https://doi.org/10.1016/j.epsl.2007.10.046), and thermodynamic modeling work by Nicoli et al. (2022, CG, https://doi.org/10.1016/j.chemgeo.2022.121025), Stewart & Penman (2024, PNAS, https://doi.org/10.1073/pnas.2401961121), and Tamang et al. (2024, JP, https://doi.org/10.1093/petrology/egae021). These studies should contribute to the numerical modeling of carbon cycle through deep time, which may alter the conclusion of this paper.

> We have added references to the Marty and Jambon, 1987, EPSL; Sano and Williams, 1996, GRL papers.
>
> Regarding the metamorphic CO_2 degassing along collisional orogens: we refute that emissions from these settings significantly contribute to the atmospheric carbon budget above the other processes we have documented in the manuscript. Kerrick and Caldeira, 1999, EPSL[1] conclude the following, “Assuming a global climate-silicate-weathering feedback, we estimate that metamorphic CO_2 degassing from the Himalayan orogen would have produced a warming of <0.5ºC, and enhanced weathering in this orogen would have produced a cooling of <0.2ºC; thus, direct climate effects of this degassing and weathering were likely to have been minor.” In addition, Pradhan and Sen, 2024, EPSL[2] conclude: “metamorphic CO_2 in the collisional orogen is comparable to other carbon sources and sinks.”
>
> If, for argument’s sake, carbon emissions from continental collisions were significant emitters of CO_2, then one would expect that the Earth would have entered a greenhouse climate throughout the Cenozoic, due to the abundance of continental collisional zones. However, the climate record tells us the Earth entered an icehouse climate during the Cenozoic. If one were to scale the present-day CO_2 emissions by the length of continental collisional zones back through geological time, then these emissions would have drastically reduced by the early Cenozoic.

3. Note that some terms require further clarification. A few examples are given below.

Line 17. The “carbon recycling” in this sentence is ambiguous and may lead to confusion. Carbon recycling is not equivalent to carbon sequestration, which specifically refers to the long-term storage of carbon, for example in oceanic lithosphere or sediments.

> We have amended the text to “carbon sequestration”. Thanks.

Line 272-273. “Atmospheric degassing” is not precise word to describe the emissions of CO2 into Earth’s atmosphere.

We have changed this to “atmospheric carbon influx”. Thanks.

Reviewer #2

In this manuscript, "Oceanic carbon sequestration and Earth’s emissions modulate icehouse-greenhouse climates", Mather et al. reevaluate the role of carbon in/outgassing at different tectonic settings on long-term global climate variability through the Phanerozoic. While drawing upon previous work utilising palaeotectonic reconstruction to interrogate the deep carbon cycle, novel to this manuscript is the decoupling of arc emissions from crustal carbonate destabilisation and subducted carbon—principally as sedimentary carbonate—to assess arc emission contributions before the evolution of widespread biomineralisation at 120 Ma. To achieve this, the authors use petrological modelling to quantify carbon and water loss during subduction; these petrological models are subsequently coupled to their palaeotectonic reconstructions. By assessing the portion of subducted carbon that contributes to arc carbon emissions at the present day, the authors suggest that carbon outgassing at tectonic settings exceeds carbon influx during greenhouse climates, whereas the opposite is true for icehouse climates. For the timeframe before 120 Ma, the authors conclude that carbon emissions from divergent settings have a greater importance in controlling Earth’s climate state than the arc-dominant present day.

I enjoyed reading and reviewing this manuscript, and I believe that the research presented is interesting, original, and suitable for Communications Earth & Environment. The outcomes and implications reported by this manuscript are of interest to a crossroads of earth scientists, including the wider deep carbon community.

However, I have several comments on the manuscript in its current form which I believe can be addressed with minor revisions. I would appreciate some consideration of forearc volatile sequestration in their model setup. At present, their model considers slab devolatilisation from the trench to 125 km, whereas it is recognised that mantle wedge forearcs may host significant proportions of carbon (e.g., Barry et al., 2019, Nature) and water (e.g., Peacock and Bostock, 2024, Can. J. Earth Sci.) that do not contribute to arc magmatism. Additionally, the closure of pores is likely to be substantial during shallow subduction, which would reduce the amount of water that contributes to the melting sub-arc mantle (e.g., Hacker, 2008, Geochem. Geophys. Geosyst.). I would appreciate the authors considering the effects of this into their model, either by considering a narrower depth range to assess sub-arc mantle wedge water concentration, or through a discussion of water loss before the slab reaches sub-arc depths.

We thank the reviewer for their detailed comments and have significantly revised the manuscript and, in particular, the methods section in line with their comments. With regard to slab devolatilization: we agree that all of the water contained within pore space of the subducting slab is lost well before the sub-arc mantle. We only document the concentration of water in pore space for completeness. Only the chemically-bound component is considered in our devolatilization calculations. We include the following line in the methods section (lines 54-55), “We only include chemically-bound water calculation, since pore water would be expelled from the slab long before the slab has a chance to reach arc depths.”

In addition, I highlight that the authors’ description of the construction of their model, comprising multiple datasets, components, and parameterisations, could benefit significantly from further clarification, e.g., by including more (sub)figures and equations in their methods illustrating how slab water content is assessed.

We have included a new supplementary figure (Figure S1) which shows how we fitted a curve to water outflux against half spreading rates at mid-ocean ridges. We have also included an additional equation describing the pore water content in deep-sea ocean sediments has been added to the methods section. The equations governing water concentration in oceanic crust was described in Jarrard (2003) in their Table 1, of which there are many, so we direct readers to that paper instead of repeating them again here.

I attach a copy of the annotated manuscript pdf to this review with specific comments enclosed; I hope that the authors find these comments constructive and helpful, and wish them well with the remainder of the review process.

> We thank the reviewer for their detailed comments. Please find a track-changes document attached to this revision where we have amended the manuscript in line with the suggested revisions.

Response to editor comments

Thank you for reviewing our manuscript. In line with your feedback, we have revised the manuscript to state that we have not taken metamorphic carbon degassing into account, the reasons for this omission and associated limitations, and toned down the conclusions. We believe the manuscript is in very good shape now thanks to the insightful feedback. Note that we have now included the methods section in the main body of the manuscript as per the editorial checklist, thus the line numbers from the reviewers have changed.

Response to reviewer comments

Reviewer #1 (Remarks to the Author):

I recommended that the metamorphic CO2 degassing should be taken into account. However, the authors refuted to do so in the second paragraph of P by P responses. They give the reasons (1) it is a minor component (Kerrick & Caldeira, 1999), (2) it is comparable to other source (Pradhan & Sen, 2024). Even though the old paper (1) suggested minor, new paper (2) said comparable, which means not negligible. Finally, they did not calculate the metamorphic one. I cannot agree with them and suggest that the paper should be returned.
In more details, they stated that in collisional orogens, metamorphic CO2 release is largely compensated by silicate weathering, as concluded in an earlier study. However, if they only consider volcanic carbon sources versus total carbon sinks, the portion of CO2 sinks compensated by metamorphic CO2 should be subtracted; otherwise, the carbon sinks would be overestimated in the carbon balance assessment.
They argued that the Cenozoic climate cooling could deny the role of collisional orogens as a major CO2 emitter. However, deep CO2 emissions (volcanic + metamorphic) are regulated not only by carbonate precipitation but also by organic carbon burial. Therefore, it is unreasonable to dismiss metamorphic CO2 by comparing it directly with the total carbon sinks (carbonate precipitation + organic carbon burial), since it represents only one part of the deep CO2 sources. Hence, their argument that “if metamorphic CO2 were important, the Cenozoic climate would be warmer than observed” is not convincing.

> We thank the reviewer for detailing their stance on metamorphic CO2 release along collisional orogens. We are still not convinced that this component would contribute a significant amount of carbon that would alter the conclusions we draw in the manuscript. The Cenozoic period has a relatively large proportion of continental collisional zones, which would suggest that CO2 emissions from continental collisions would be at their highest. However, the papers we cited (Kerrick & Caldeira 1999 and Pradhan & Sen 2024) suggest their emissions were either not significant or not more any more significant than other carbon fluxes. Thus their contributions before the Cenozoic must be even more reduced than in recent times because there were fewer continental collisional zones.

In any case, we have sought to resolve the matter in our manuscript by clearly stating that we do not consider metamorphic CO2 degassing in our carbon contributions and what the implications are, "A limitation of our study is that we do not consider metamorphic CO_2 degassing along collisional orogens. The abundance of collisional zones during the Cenozoic would suggest that metamorphic CO_2 degassing should be proportionally high (Macdonald *et al.* 2019). While this process has been shown to emit significant quantities of carbon in the Himalayas (Becker *et al.* 2008) and the Tibetan Plateau (Liu *et al.* 2024), other studies have suggested that metamorphic CO_2 degassing is minor (Kerrick & Caldeira 1999) or no more significant than other carbon sources (Pradhan & Sen 2024) at the global scale. Regardless, the drop in collisional orogens prior to the Cenozoic would significantly reduce the role of metamorphic CO_2 degassing on icehouse-greenhouse climate variations." (Lines 267-275)

Reviewer #2 (Remarks to the Author):

After reading through the latest version of the manuscript by Mather et al., "Carbon emissions along divergent plate boundaries modulate icehouse-greenhouse climates", I am satisfied that most of my requested revisions have been made by the authors, and I thank the authors for their time and effort in revising this manuscript according to both my comments and those of Reviewer 1. Below I briefly flag up some minor comments on the revised manuscript which should be straightforward for the authors to address.
I wish the authors the best with the remainder of the publication process.
Kevin Wong
Carnegie Institution for Science
23 October 2025

Lines 240–241: "The present-day rift degassing is skewed towards rifts with active plume volcanism and cratonic lithosphere involvement". The authors here cite Brune et al. (2017, Nat. Geosci.) which does not mention plume volcanism nor cratonic lithosphere. Perhaps the recent review by Brune et al. (2023, Nat. Rev. Earth Environ.) is more appropriate here.

Added.

Methods Line 59: Missing word: "We only include chemically-bound water [in our] calculation…"

Fixed.

Methods Line 63: More of a broad comment, and feel free to address this briefly in your methods if you want, but how do thermal parameters (Φ) of different subduction systems vary over time, and what effect they may have on your study? For example, Cascadia at the present day has a low thermal parameter (van Keken et al., 2011, J. Geophys. Res. Solid Earth) and experiences significant slab devolatilisation at shallow pressures, evident from a highly serpentinised mantle wedge (e.g., Abers et al. 2017, Nat. Geosci.; Peacock and Bostock, 2024, Can. J. Earth Sci.). Of course, slab volatile release is not necessarily always correlated with thermal parameter; the subducting slab entering the Nankai trough also has a low thermal parameter but there is less evident serpentinisation from forearc metamorphic fluid release. It would nonetheless be interesting to first-order estimate the degree of 'chemically-bound' water lost from these low thermal parameter slabs in your model before 125 km depth.

> We agree the thermal parameter (Φ) offers an interesting first-order proxy for slab devolatilistation. In particular, it scales well with the uptake of plate bending-related serpentinization at subduction zones. However, in Figure 1b we show that the overall proportion of carbon influx to the oceanic plate from this process is relatively small. The thermal parameter is thus better seen as an enabler of slab devolatilization: if the subducting slab is laden with volatiles, then a high Φ would lead to its efficient devolatilization. We have added to lines 73-74 of the methods section explaining this.

Methods Line 258: Typo: "Perplex" to "Perple_X"

> Fixed.

Methods Line 278: Reference to "mean global temperature" missing.

> Added reference to Scotese et al 2021 ESR.

References

Becker, J.A., Bickle, M.J., Galy, A. & Holland, T.J.B., 2008. Himalayan metamorphic CO2 fluxes: Quantitative constraints from hydrothermal springs. *Earth and Planetary Science Letters*, **265**, 616–629. doi:10.1016/j.epsl.2007.10.046

Kerrick, D.M. & Caldeira, K., 1999. Was the Himalayan orogen a climatically significant coupled source and sink for atmospheric CO2 during the Cenozoic? *Earth and Planetary Science Letters*, **173**, 195–203. doi:10.1016/S0012-821X(99)00229-0

Liu, W., Zhang, M., Liu, Y., Cui, L., Sano, Y., Zhou, X., Li, Y., *et al.*, 2024. Massive crustal carbon mobilization and emission driven by India underthrusting Asia. *Commun Earth Environ*, **5**, 271. doi:10.1038/s43247-024-01438-z

Macdonald, F.A., Swanson-Hysell, N.L., Park, Y., Lisiecki, L. & Jagoutz, O., 2019. Arc-continent collisions in the tropics set Earth's climate state. *Science*, **364**, 181–184. doi:10.1126/science.aav5300

Pradhan, S. & Sen, I.S., 2024. Metamorphic CO2 fluxes offset the net geological carbon sink in the Himalayan-Tibetan orogen. *Earth and Planetary Science Letters*, **647**, 119018. doi:10.1016/j.epsl.2024.119018

Oceanic carbon sequestration and Earth's emissions modulate icehouse-greenhouse climates

Ben R. Mather[1], R. Dietmar Müller[1], Adriana Dutkiewicz[1], Sabin Zahirovic[1]

[1] – EarthByte Group, School of Geosciences, The University of Sydney, Sydney, NSW 2006, Australia
* Corresponding author: ben.mather@sydney.edu.au

SUMMARY

The exchange of carbon between oceanic plates, the deep Earth, and the atmosphere plays a significant role in modulating global climate[1,2]. Icehouse-greenhouse climate fluctuations have been attributed to changes in palaeogeography and solid Earth degassing[3], particularly along continental arcs[2,4,5], to arc weathering[5] and to the sequestration of carbon into oceanic carbonate-rich sediments[6]. However, the proportions of these contributions and their effect on modulating global climate are poorly constrained. Here we show that the changing balance between volcanic outgassing and carbon sequestration into oceanic lithosphere is the key driver for major climate shifts. Combined volcanic arc, mid-ocean ridge and continental rift emissions exceed carbon recycling during greenhouse climates. In contrast, oceanic plate sequestration exceeds emissions during icehouse climates. Our results challenge the long-held view that carbon degassing along volcanic arcs is the main contributor to atmospheric carbon[2,4,5], suggesting instead that mid-ocean ridge and rift degassing surpass arc emissions before 100 Ma.

Earth has experienced multiple climate extremes throughout the Phanerozoic, including icehouse states in the late Ordovician, late Paleozoic, and Cenozoic[5]. Long-lived climate fluctuations are attributed to increased atmospheric CO_2 concentrations contributing to greenhouse climates, and lower CO_2 content contributing to icehouse climates[2]. A key source of atmospheric carbon is from the degassing of mid-ocean ridges (MORs) and continental rifts. The rate of MOR degassing is proportional to the rate of crustal production, which has varied considerably through time but has only previously been estimated for times younger than 250 Ma[7]. Carbon emissions along continental rifts contribute 14.5 ± 1.5 Mt C/a at present[8], which scales with the estimated rift length through time. Island and continental arc magmatism is widely considered to add more carbon into the atmosphere than any other geologic process[4,5]. Volcanic arc emissions are derived from decarbonated subducting slabs[9], and metamorphic decarbonation when arc magmas ascends though crustal carbonate platforms[10]. The metamorphic decarbonation of carbonate bodies from arc magmatism, termed "carbonate platform degassing", has been proposed to contribute significant amounts of carbon to the atmosphere[10–12]. At present day, approximately 20–30% of active volcanic outgassing (18.4 ± 4.3 Mt C/a[9]) and 80–100% of diffuse outgassing (25.4 ± 12.3 Mt C/a[13]) along volcanic arcs is attributed to carbonate platform degassing[11]. However, their carbon emissions through time are biased by present-day estimates because the area of extinct carbonate platforms preserved and juxtaposed with subduction zones via subduction initiation and margin conversion has progressively increased towards the present day. From the total arc outgassing of 44.1 ± 16.6 Mt C/a, approximately 13.4–44.5 Mt C/a is partitioned from carbonate platforms. The remainder is contributed by decarbonated subducting slabs. At present day, the carbon emissions from arc volcanoes derived from decarbonated slabs is primarily controlled by the recycling and devolatilisation of thick calcareous deep-sea sediments at sub-arc depths[14]. However, the influx of these carbonate carbon-rich sediments into subduction zones has been significant only over the last 120 Myr[6] due to the evolution and proliferation of planktic calcifiers that comprise deep-sea carbonates[15]. Prior to 120 Ma, sediment-hosted carbonate carbon subduction was insignificant, which would reduce slab-derived carbon emissions along volcanic arcs back through time. For most studies, present-day outgassing measurements at arc volcanoes are linearly scaled back through geological time using continental arc length estimates[5,12], and do not account for the change in composition of the subducting slab nor in the abundance of extinct carbonate platforms along subduction zones. In this study, we track the evolution of subducting carbon reservoirs and their devolatilisation, using a plate tectonic model for the last 540 Myr[16] coupled to a Perple_X thermodynamic model[17], to more accurately reconstruct arc emissions derived from decarbonated slabs. In addition, we track the presence of carbonate platforms and their intersections with volcanic arcs to reconstruct the total volcanic arc emissions through the Phanerozoic. Lastly, we extend rates of MOR and continental rift degassing from 250 Ma to 540 Ma to compare their importance relative to volcanic arc degassing in icehouse/greenhouse climate variations through Earth's history.

Volcanic arc emissions from carbonate platforms

To estimate the total carbon outgassing at volcanic arcs, we first quantify carbonate platform degassing rates by reconstructing carbonate platform-subduction zone intersections over the Phanerozoic using a plate tectonic model[16]. Previous studies have tracked the intersections of carbonate platforms with subduction zones to estimate their relative contribution to arc emissions since 410 Ma[18,19]. We follow a similar approach but extend the reconstruction of carbonate platforms, based on those mapped at the present day, back to 500 Ma[20] and scale carbonate platform emissions relative to the rate of mantle melting, rather than scaling by arc length[19]. The magnitude of arc volcanism is largely controlled by the hydration of the sub-arc mantle[21], which in-turn promotes partial melting, magmatism and associated metamorphic decarbonation when magma ascends though crustal carbonate bodies[10]. We reconstruct the oceanic plate water reservoirs (Supplementary Figure 1) and estimate the quantity of water leaving the subducting slab at sub-arc depths shallower than 125 km, hydrating the mantle wedge, and calculate the melting rate using a model of hydrous mantle melting[22] (refer to Methods). The result is used to scale the present-day rate of carbonate platform degassing back through the Phanerozoic.

The carbon outflux from carbonate platforms constitutes all volcanic arc emissions before 120 Ma, gradually increasing from ~12 Mt C/yr at 500 Ma to 28 Mt C/yr at present day (Figure 1a, Supplementary Figure 2). This is because the surface area of carbonate platforms gradually expands through time until the present day (Figure 2, Movie S1). An alternative model of crustal arc growth, based on arc magma addition rates[23] scaled by total arc lengths[4], predicts generally lower carbon emissions than our curve (Supplementary Figure 2). The discrepancy is particularly prevalent after the mid-Cretaceous when carbon emissions drop sharply but not nearly as much as an alternate model of arc emissions[12], which predicts much higher rates of degassing before the present day because arc lengths are scaled by zircon populations rather than plate reconstructions (Supplementary Figure 2). While zircons may be a useful constraint on the arc length where other geological data are sparse[5], their record suffers from a preservation bias. We therefore argue that plate reconstructions could be more useful for mapping continental arc lengths through time (Figure 3a) because they assimilate a diverse range of geological data to constrain subduction zone and arc locations[24].

A limitation of our study is that we do not consider the thickness of carbonate platforms in our calculations. Estimating carbonate platform thickness through time is challenging owing to highly uncertain carbonate accumulation rates, which are sensitive to fluctuations in sea level and variations in sedimentation/erosion rates[6]. If carbonate platform decarbonation flux into the atmosphere scales with platform thickness, then our reconstructions of carbonate platform emissions would deviate from Figure 1a. On the one hand, more carbon could be outgassed by continental arcs due to the subduction of thicker carbonate platforms; on the other hand, less carbon could be outgassed because we assume in our calculation that carbonate platforms that have been mapped withstand tectonic destruction throughout the Phanerozoic, and do not experience time-dependent carbon depletion. It is difficult to reconcile these two competing factors; nevertheless, our estimate of carbonate platform emissions are calibrated against present-day arc estimates[9,13], and roughly agrees with other reconstructions of carbonate platform emissions through time (Supplementary Figure 2).

Plate influx and volcanic arc emissions from slab devolatilisation

The rate of carbon sequestration into oceanic plates is estimated using a simplified model of three major volatile-carrying reservoirs: deep-sea carbonate sediments[6], hydrothermally altered ocean crust[7], and partly serpentinised and metasomatised lithospheric mantle[25] (Figure 1b). Serpentinisation related to plate bending at subduction zones stores additional carbon in the oceanic plate (4–15 Mt C/a at present day)[1] and is scaled by the slab dip angle[26]. Partially serpentinised mantle lithosphere due to slow spreading rates along MORs contributes an additional 5–11 Mt C/a into present-day oceanic plates[27]. For deep-sea carbonate sediments, we assume that up to 20% of the their total flux reflects a net increase in marine carbonate carbon sequestration since the Late Cretaceous, driven by a doubling in the flux of terrigenous sediments into the oceans during the Cenozoic[28] resulting in additional atmospheric carbon drawdown driven by increased continental silicate weathering (see Methods). The rate of oceanic plate influx is balanced by the subduction influx, (i.e., the amount of carbon being subducted is subtracted from the carbon being sequestered into the oceanic plates). During the subduction of carbon reservoirs stored within oceanic plates, volatiles are released from the down-going slab stimulating partial melting and arc magmatism[22]. The carbon loss at sub-arc depths (≤ 125 km) is calculated using a simplified thermodynamic model of each reservoir (Supplementary Figure 3). The majority of sediment-hosted carbon is released at depths ≤ 125 km, while the other carbon-carrying reservoirs are ultimately recycled into deeper portions of the upper mantle[29].

Assuming that 20–30% of present-day volcanic outgassing is from carbonate platforms[11], the remaining proportion must be derived from the decarbonation of subducting slabs[14]. To estimate the outgassing related to slab outflux, we estimate the influx of carbon into major carbon-carrying reservoirs within oceanic lithosphere and track their migration as they are propelled by plate tectonics towards subduction zones (Methods, Supplementary Figure 4). Summing all the carbon reservoirs in the oceanic lithosphere, the total carbon entering subduction zones at the present day is 101–138 Mt C/a (Figure 1c). From our thermodynamic calculations of subducting slabs, ~59–78 Mt C/a of this total is released from the slab at sub-arc depth, which is in broad agreement with the 46–55% of the total subducting carbon from previous carbon flux estimates[1]. In order to fit a present-day volcanic arc degassing rate of 11–18 Mt C/a (not attributed to carbonate platforms), we estimate that ~22% of carbon released from the slab enters the atmosphere while the remainder is embedded in the overriding plate or is circulated within the upper mantle. Taking 22% of reconstructed slab carbon outflux through the Phanerozoic, we find that volcanic arc emissions are near zero prior to 120 Ma and increase to 11–18 Mt C/a towards the present day due to the subduction of deep-sea carbonate sediments[7] (Figure 1a, orange curve). A long-term increase in pelagic carbonate sediments since the mid-Cretaceous was punctuated by a doubling of carbonate carbon sequestration post-50 Ma to ~310 Mt C/a[6]. Most of this flux is thought to reflect a shift of carbonate deposition from shallow to deep water, while up to 20% of the total flux may reflect a net increase in deep-sea carbonate carbon flux since the Late Cretaceous, which reaches up to 50 Mt C/a (Figure 1b). Consequently, present-day arc degassing rates are skewed by the relatively recent subduction of pelagic carbonate sediments (Figure 1a). Therefore, the assumption that the present-day partitioning of subducting carbon reservoirs can be used to estimate rates of arc degassing through deep time is flawed.

Multiple studies have inferred the rate of volcanic arc degassing through deep time using the present-day rate of arc degassing paired with reconstructions of arc length from zircon populations[5] or plate reconstructions[2,18] to explain climate variability. Our work provides an alternate calculation which accounts for the absence of significant volumes of subducting deep-sea carbonates prior to 120 Ma[6], which are the source of most slab-derived carbon emissions from arc volcanoes (Figure 1c). Furthermore, we incorporate plate convergence rates which are intrinsic to the amount of carbon that is recycled and outgassed along volcanic arcs (Figure 3b). By extension, our volcanic arc degassing curve (Figure 1a) considers the competing effects of a decline in the plate destruction rate (the product of convergence rate and trench length, Figure 3c) and the dramatic increase in subducting carbonate sediments since ~120 Ma.

A limitation of our study is that we do not consider the deposition of continental sediments or organic carbon in our sedimentary carbon budget. However, it is estimated that deep-sea carbonate contributes over 80% of the total sedimentary carbon flux at the present day[30]. More than 45 Mt C/a would need to be contributed by continental sediments or organic carbon into subduction zones to increase arc emissions before 120 Ma and offset emissions from MORs and continental rifts. A subduction influx of 45 Mt C/a would exceed the subduction influx from all other carbon-carrying reservoirs in subducting slabs (deep-sea sediments, altered oceanic crust, serpentinised and metasomatized mantle lithosphere) between the early Cambrian and Early Jurassic. It is therefore unlikely that the subduction of continental sediments or organic carbon could provide a sufficient boost in carbon emissions along arcs to outweigh the contribution from MOR and continental rift outgassing.

Carbon emissions along spreading ridges and continental rifts

The rate of MOR outgassing is proportional to the rate of seafloor spreading. To estimate the rate of carbon emissions at mid-ocean ridges, we connect an established model of volatile extraction[31] to tectonic reconstructions of MOR geometries and spreading rates through the Phanerozoic[16] (Figure 1). Our range of 10–16 Mt C/a for the present day (Figure 1a) is consistent with previous estimates using the same volatile extraction model[7]. The breakup of Pangea is characterised by fast spreading rates during the Late Cretaceous (Figure 3b) and a corresponding peak in MOR outflux of 30–40 Mt C/a (Figure 1a). Other peaks at 360 and 450 Ma correspond to periods of increased crustal production associated with the assembly of Pangea (Figure S1). MOR degassing decreases from 130 Ma to the present day due to the extinction and stepwise subduction of MOR segments[7]. However, it is worth considering that the seafloor spreading record, and thus MOR lengths and spreading rates, are synthetic in the pre-Pangea timeframe. A combination of geological and paleomagnetic data are used as constraints where and how fast plates diverge. However, ocean basins entirely surrounded by subduction zones carry larger uncertainties than oceans bounded by passive margins, leading to significant associated uncertainties in estimating these parameters in deep time.

The present-day rate of rift degassing is 14.5 ± 1.5 Mt C/a[8], which we scale back through time by the rift length[32,33]. Mapped rifts in the Cambrian and early Paleogene were formed during continental breakup and/or terrain detachment and migration, while younger rifts include a substantial portion formed in convergent settings such as back-arc basins[33]. The uncertainty of estimating rift degassing is relatively high due to the preservation bias of rifts in estimating the present-day degassing rate, and the age of rift initiation and cessation[8]. The present-day rift degassing is skewed towards rifts with active plume volcanism and cratonic lithosphere involvement[8]. Nevertheless, rift degassing combined with MOR degassing exceeds carbon emissions from volcanic arcs before 100 Ma. Prior to the middle Cretaceous, carbon emissions from MORs and rifts contribute ~ 62% of total carbon emissions into the atmosphere (Figure 1a).

Icehouse – greenhouse climate variations

Compilations of palaeotemperature [34] and atmospheric CO_2 records[35] reveal the diversity of past Earth climates (Figure 4). Continental arc weathering has been suggested as a major driver of global climate[36], where a high abundance of continental arc sutures within tropical latitudes leads to higher erosion rates that lower atmospheric CO_2 concentration. An increased flux of silicate-rich sediment from the continents into the oceans is correlated to higher seawater $^{87}Sr/^{86}Sr$ ratios during some greenhouse periods[37]; however, this is complicated by more intensive mafic weathering (associated with hydrothermal flux at MORs and off-axis alteration of the seafloor), which lowers the $^{87}Sr/^{86}Sr$ ratio[38]. To more comprehensively understand past climates, the variations in $^{87}Sr/^{86}Sr$ ratios, MOR and continental arc lengths (Figure 3a), spreading and convergence rates (Figure 3b), and rates of oceanic crustal production and destruction (Figure 3c) must be considered in concert with the exchange of carbon between the solid Earth and the atmosphere. By combining our estimates of carbon outgassing at volcanic arcs and MORs (Figure 1a) with carbon influx into major carbon-carrying reservoirs in oceanic lithosphere (Figure 1b), we calculate the ratio of carbon partitioning between the Earth's atmosphere and ocean basins (Figure 4c, refer to Methods). Changes in this ratio predict climate trends: values > 1 indicate an increase in carbon being outgassed into the atmosphere, while values < 1 indicate that more carbon is being sequestered into oceanic plates. Our curve tracks most of the icehouse-greenhouse climate fluctuations through the Phanerozoic era, including the early Paleozoic greenhouse, early Devonian greenhouse, late Paleozoic icehouse, Mesozoic-Early Cretaceous greenhouse, late Cretaceous "coolhouse" climate, and the Cenozoic icehouse (Figure 4). From a statistical analysis of mean global temperature (Figure 4a) and the ratio of atmospheric outflux to oceanic plate influx (Figure 4c), we find a negligible time lag between signals and a moderately strong correlation coefficient of 0.6 (Supplementary Figure 5).

The early Paleozoic greenhouse is characterised by a high ratio of carbon outflux from MORs compared to oceanic plate sequestration (Figure 4c, 2f), leading to moderate surface temperatures during the Cambrian (20–25 °C, Figure 4a). Global temperatures decline from the early to middle Ordovician (20–15 °C) as the rate of crustal destruction at subduction zones exceeds crustal production at MORs (Figure 3c). This distinct early to middle Ordovician cooling trend from ~490–430 Ma[34] culminated in the short-lived Hirnantian glaciation at the end of the Ordovician at 445–444 Ma[39], featuring substantial ice sheets extending to 35° latitude[34] (Figure 4c). The abrupt drop in global temperature has recently been attributed to an asteroid impact[40], which cannot be reproduced by our atmospheric outflux / oceanic plate influx curve. Low seawater $^{87}Sr/^{86}Sr$ ratios during the late Ordovician to early Silurian have been linked to the weathering of predominantly mafic and ultramafic rocks associated with the Taconic arc-continent collision[41] (Figure 4a), but may also be explained by a spike in oceanic crustal production rates from 460–435 Ma (Figure 3c).

The high atmospheric CO_2 concentration during the early Devonian greenhouse (Figure 2e, 4b) corresponds to a peak in our curve due to low convergence rates (3–6 cm/yr, Figure 3b), thus higher rates of oceanic plate influx (Figure 1b) in response to the diminished recycling of carbon-rich reservoirs into the mantle (Figure 1c). Seawater $^{87}Sr/^{86}Sr$ ratios increased despite low convergence rates and a diminished potential for silicate weathering (Figure 4a). Without a significant proportion of continental arc sutures within tropical latitudes to draw down atmospheric CO_2 via continental silicate weathering[36], the increase in seawater $^{87}Sr/^{86}Sr$ ratios likely reflects a decline in seafloor spreading rates (Figure 3b), and thus reduced hydrothermal flux and seafloor alteration at MORs.

MOR outflux decreased from 26 to 15 Mt C/a at the onset of the late Paleozoic icehouse (Figure 1a), due to low oceanic crustal production rates which persisted from 320–260 Ma (Figure 3c). The combination of low MOR outflux and increased continental arc weathering[36], reflected in elevated seawater $^{87}Sr/^{86}Sr$ ratios (Figure 4a), significantly lowered the atmospheric CO_2 content to establish one of the most extensive glaciations in geologic history[42]. After a temperature spike following the late Paleozoic icehouse, likely triggered by plume-related volcanism associated with the breakup of Pangea[43] (Figure 2d), the Mesozoic-early Cretaceous greenhouse climate was sustained by a moderately positive atmospheric outflux to oceanic plate influx ratio (Figure 4c). A brief "coolhouse" climate during the middle Cretaceous[44], largely unexplained by other models, appears to be associated with a reduced rift degassing of 10 Mt C/a and MOR degassing of 7 Mt C/a (Figure 1a, 2c). A return to warmer temperatures in the early Paleogene is driven by a reduced oceanic plate influx (Figure 1b) as less carbon was being removed from the oceanic plates via subduction (Figure 1c, 2b).

The Cenozoic icehouse is marked by a substantial increase in oceanic plate influx from 51 Mt C/a at the start of the Cenozoic (66 Ma) to 77 Mt C/a at the present day due to the proliferation of planktic calcifiers that comprise deep-sea carbonates[15] (Figure 1b). The sequestration of significant quantities of carbon into ocean basins outweighs the carbon being removed at subduction zones (Figure 1c) and since ~22% of carbon released from the slab at sub-arc depths is estimated to reach the atmosphere, the plate influx significantly outweighs the atmospheric outflux leading to a cooler climate. Increased continental silicate weathering, indicated by high seawater $^{87}Sr/^{86}Sr$ ratios (Figure 4a), is due to the clustering of continental arcs within tropical latitudes during the Cenozoic[45] (Figure 2a), and may have further enhanced the removal of atmospheric CO_2.

Conclusions

Our findings challenge the notion that continental arc emissions are the primary drivers of atmospheric carbon levels[5]. Our breakdown of arc emissions from carbonate platforms and subducting plate devolatilisation reveals that this mechanism only holds for the last 100 Myr. While carbonate platform degassing emits a significant quantity of carbon at the present day and exceeds subduction outflux at volcanic arcs prior to 120 Ma, their combined contributions amount to only ~38% of total emissions including MOR and rift degassing. Thus, carbon emissions from MORs and continental rifts are the dominant source of atmospheric carbon for most of the Phanerozoic. We find that long-term variations in global climate are primarily driven by the balance between atmospheric degassing (MORs, rifts and volcanic arcs), and the sequestration of carbon into oceanic plate reservoirs. The oceanic plate influx is in turn modulated by the recycling of carbon-rich reservoirs into the Earth's mantle. This has significant implications for palaeoclimate reconstructions through deep time, where the balance between atmospheric outflux and oceanic plate influx is a major factor in maintaining a habitable environment for life, while large fluctuations between atmospheric outflux and oceanic plate influx may drive icehouse-greenhouse climate variations.

REFERENCES

1. Kelemen, P. B. & Manning, C. E. Reevaluating carbon fluxes in subduction zones, what goes down, mostly comes up. *Proceedings of the National Academy of Sciences of the United States of America* **112**, E3997–E4006 (2015).
2. Mills, B. J. W., Scotese, C. R., Walding, N. G., Shields, G. A. & Lenton, T. M. Elevated CO2 degassing rates prevented the return of Snowball Earth during the Phanerozoic. *Nat Commun* **8**, 1110 (2017).
3. Merdith, A. S. *et al.* Phanerozoic icehouse climates as the result of multiple solid-Earth cooling mechanisms. *Science Advances* **11**, eadm9798 (2025).
4. Cao, W., Lee, C.-T. A. & Lackey, J. S. Episodic nature of continental arc activity since 750 Ma: A global compilation. *Earth and Planetary Science Letters* **461**, 85–95 (2017).

5. McKenzie, N. R. *et al.* Continental arc volcanism as the principal driver of icehouse-greenhouse variability. *Science* **352**, 444–447 (2016).
6. Dutkiewicz, A., Müller, R. D., Cannon, J., Vaughan, S. & Zahirovic, S. Sequestration and subduction of deep-sea carbonate in the global ocean since the Early Cretaceous. *Geology* **47**, 91–94 (2019).
7. Müller, R. D. *et al.* Evolution of Earth's tectonic carbon conveyor belt. *Nature* **605**, 629–639 (2022).
8. Brune, S., Williams, S. E. & Müller, R. D. Potential links between continental rifting, CO2 degassing and climate change through time. *Nature Geosci* **10**, 941–946 (2017).
9. Bekaert, D. V. *et al.* Subduction-Driven Volatile Recycling: A Global Mass Balance. *Annual Review of Earth and Planetary Sciences* **49**, (2021).
10. Mason, E., Edmonds, M. & Turchyn, A. V. Remobilization of crustal carbon may dominate volcanic arc emissions. *Science* **294**, 290–294 (2017).
11. Aiuppa, A., Fischer, T. P., Plank, T., Robidoux, P. & Di Napoli, R. Along-arc, inter-arc and arc-to-arc variations in volcanic gas CO2/ST ratios reveal dual source of carbon in arc volcanism. *Earth-Science Reviews* **168**, 24–47 (2017).
12. Chu, X., Lee, C.-T. A., Dasgupta, R. & Cao, W. The contribution to exogenic CO2 by contact metamorphism at continental arcs: A coupled model of fluid flux and metamorphic decarbonation. *Am J Sci* **319**, 631–657 (2019).
13. Fischer, T. P. *et al.* The emissions of CO2 and other volatiles from the world's subaerial volcanoes. *Scientific Reports* **9**, 1–11 (2019).
14. Plank, T. & Manning, C. E. Subducting carbon. *Nature* **574**, 343–352 (2019).
15. Erba, E. The first 150 million years history of calcareous nannoplankton: Biosphere–geosphere interactions. *Palaeogeography, Palaeoclimatology, Palaeoecology* **232**, 237–250 (2006).

16. Müller, R. D. *et al.* A tectonic-rules-based mantle reference frame since 1 billion years ago – implications for supercontinent cycles and plate–mantle system evolution. *Solid Earth* **13**, 1127–1159 (2022).
17. Connolly, J. A. D. The geodynamic equation of state: What and how. *Geochemistry Geophysics Geosystems* **10**, 1–19 (2009).
18. Pall, J. *et al.* The influence of carbonate platform interactions with subduction zone volcanism on palaeo-atmospheric CO2 since the Devonian. *Climate of the Past* **14**, 857–870 (2018).
19. Zahirovic, S. *et al.* Subduction and carbonate platform interactions. *Geoscience Data Journal* **9**, 371–383 (2022).
20. Kiessling, W., Flügel, E. & Golonka, J. Patterns of Phanerozoic carbonate platform sedimentation. *Lethaia* **36**, 195–225 (2003).
21. England, P. C. & Katz, R. F. Melting above the anhydrous solidus controls the location of volcanic arcs. *Nature* **467**, 700–703 (2010).
22. Katz, R. F., Spiegelman, M. & Langmuir, C. H. A new parameterization of hydrous mantle melting. *Geochemistry, Geophysics, Geosystems* **4**, 1–19 (2003).
23. Ratschbacher, B. C., Paterson, S. R. & Fischer, T. P. Spatial and Depth-Dependent Variations in Magma Volume Addition and Addition Rates to Continental Arcs: Application to Global CO2 Fluxes since 750 Ma. *Geochemistry, Geophysics, Geosystems* **20**, 2997–3018 (2019).
24. Merdith, A. S. *et al.* Extending full-plate tectonic models into deep time: Linking the Neoproterozoic and the Phanerozoic. *Earth-Science Reviews* **214**, 103477 (2021).
25. Alt, J. C. *et al.* The role of serpentinites in cycling of carbon and sulfur: Seafloor serpentinization and subduction metamorphism. *Lithos* **178**, 40–54 (2013).
26. Mather, B. R., Müller, R. D., Alfonso, C. P., Seton, M. & Wright, N. M. Kimberlite eruptions driven by slab flux and subduction angle. *Scientific Reports* **13**, 1–12 (2023).

27. Merdith, A. S., Atkins, S. E. & Tetley, M. G. Tectonic Controls on Carbon and Serpentinite Storage in Subducted Upper Oceanic Lithosphere for the Past 320 Ma. *Frontiers in Earth Science* **7**, 1–23 (2019).

28. Salles, T. *et al.* Hundred million years of landscape dynamics from catchment to global scale. *Science* **379**, 918–923 (2023).

29. Van Keken, P. E., Hacker, B. R., Syracuse, E. M. & Abers, G. A. Subduction factory: 4. Depth-dependent flux of H2O from subducting slabs worldwide. *Journal of Geophysical Research: Solid Earth* **116**, (2011).

30. Clift, P. D. A revised budget for Cenozoic sedimentary carbon subduction. *Reviews of Geophysics* **55**, 97–125 (2017).

31. Keller, T., Katz, R. F. & Hirschmann, M. M. Volatiles beneath mid-ocean ridges: Deep melting, channelised transport, focusing, and metasomatism. *Earth and Planetary Science Letters* **464**, 55–68 (2017).

32. Merdith, A. S., Williams, S. E., Brune, S., Collins, A. S. & Müller, R. D. Rift and plate boundary evolution across two supercontinent cycles. *Global and Planetary Change* **173**, 1–14 (2019).

33. Şengör, A. M. C. & Natal'in, B. A. Rifts of the world. in *Mantle plumes: their identification through time* (Geological Society of America, 2001). doi:10.1130/0-8137-2352-3.389.

34. Scotese, C. R., Song, H., Mills, B. J. W. & van der Meer, D. G. Phanerozoic paleotemperatures: The earth's changing climate during the last 540 million years. *Earth-Science Reviews* **215**, 103503 (2021).

35. Foster, G. L., Royer, D. L. & Lunt, D. J. Future climate forcing potentially without precedent in the last 420 million years. *Nature Communications* **8**, 1–8 (2017).

36. Macdonald, F. A., Swanson-Hysell, N. L., Park, Y., Lisiecki, L. & Jagoutz, O. Arc-continent collisions in the tropics set Earth's climate state. *Science* **364**, 181–184 (2019).

37. Gernon, T. M. *et al.* Global chemical weathering dominated by continental arcs since the mid-Palaeozoic. *Nat. Geosci.* **14**, 690–696 (2021).

38. van der Meer, D. G. *et al.* Reconstructing first-order changes in sea level during the Phanerozoic and Neoproterozoic using strontium isotopes. *Gondwana Research* **44**, 22–34 (2017).

39. Finnegan, S. *et al.* The Magnitude and Duration of Late Ordovician–Early Silurian Glaciation. *Science* **331**, 903–906 (2011).

40. Glikson, A. Y. An asteroid impact origin of the Hirnantian (end-Ordovician) glaciation and mass extinction. *Gondwana Research* **118**, 153–159 (2023).

41. Swanson-Hysell, N. L. & Macdonald, F. A. Tropical weathering of the Taconic orogeny as a driver for Ordovician cooling. *Geology* G38985.1 (2017) doi:10.1130/G38985.1.

42. Walzer, U. & Hendel, R. Natural climate change and glaciations. *Earth-Science Reviews* **241**, 104435 (2023).

43. Dang, Z., Zhang, N., Li, Z.-X. & Yan, P. Pangaea's breakup: the roles of mantle plumes, orogens and subduction retreat. *Geological Society, London, Special Publications* **542**, SP542-2022–345 (2023).

44. Goldberg, S. L., Present, T. M., Finnegan, S. & Bergmann, K. D. A high-resolution record of early Paleozoic climate. *Proc. Natl. Acad. Sci. U.S.A.* **118**, e2013083118 (2021).

45. Li, S., Goldstein, S. L. & Raymo, M. E. Neogene continental denudation and the beryllium conundrum. *Proceedings of the National Academy of Sciences* **118**, e2026456118 (2021).

46. Faccenda, M. Water in the slab: A trilogy. *Tectonophysics* **614**, 1–30 (2014).

47. Veizer, J. *et al.* 87Sr/86Sr, δ13C and δ18O evolution of Phanerozoic seawater. *Chemical Geology* **161**, 59–88 (1999).

48. Halverson, G. P., Wade, B. P., Hurtgen, M. T. & Barovich, K. M. Neoproterozoic chemostratigraphy. *Precambrian Research* **182**, 337–350 (2010).

49. Witkowski, C. R., Weijers, J. W. H., Blais, B., Schouten, S. & Sinninghe Damsté, J. S. Molecular fossils from phytoplankton reveal secular PCO2 trend over the Phanerozoic. *Science Advances* **4**, eaat4556 (2018).

Acknowledgements
This study was supported by BHP through the Spatio TEmporaL expLorAtion for Resources 'STELLAR' project and the AuScope Simulation, Analysis & Modelling node funded by the Australian Government through the National Collaborative Research Infrastructure Strategy, NCRIS. We gratefully acknowledge funding from the Australian Research Council through grants DE210100084 (SZ) and FT190100829 (AD).

Author contributions
BM interrogated the plate reconstruction models and coupled them with the thermodynamic modelling framework, wrote the paper and prepared the figures. RDM and AD conceived and directed the project. SZ digitised the carbonate platforms back to 500 Ma. All authors participated in the discussion and interpretation of the results and in the preparation of the manuscript.

Competing interests The authors declare no competing interests.

Data availability: The datasets generated and analysed during the current study are available in the Zenodo repository, https://doi.org/10.5281/zenodo.15315706

Code availability: Plate reconstruction software, GPlates, is freely available from www.gplates.org/download.html. Thermodynamic modelling software, Perple_X, and associated thermodynamic databases are freely available from www.perplex.ethz.ch.

Supplementary information is available for this paper.
Correspondence and requests for materials should be addressed to B.M.

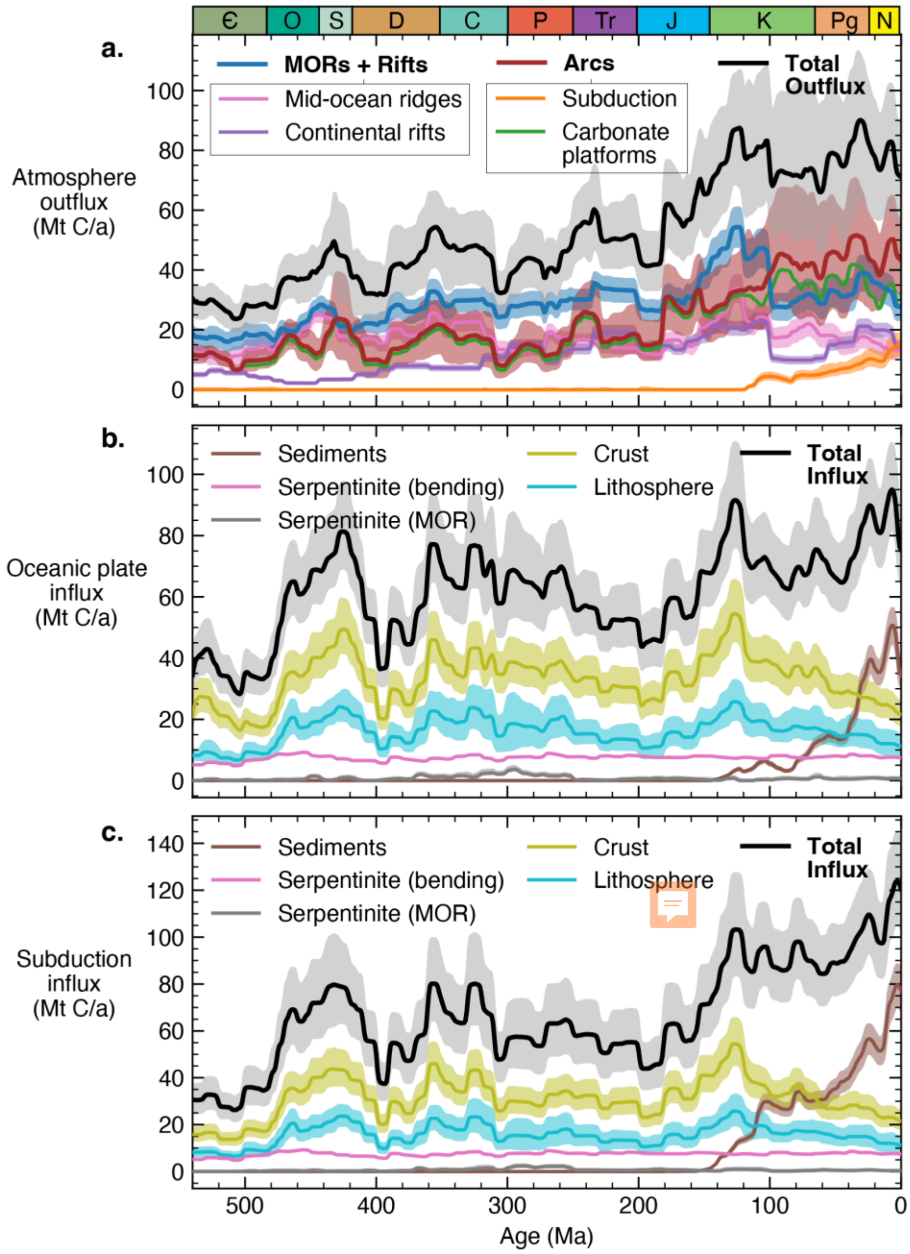


Figure 1: Fluxes of carbon into the atmosphere and mantle. a. Contributions of mid-ocean ridge, continental rift, volcanic arc, and carbonate platform outflux to atmospheric carbon. Total volcanic arc degassing (maroon line) is partitioned into the release of carbon from subducting slabs (orange line) and degassing from carbonate platforms (green line). Mid-ocean ridge (pink line) and continental rift degassing (purple line) combine to the total outflux from divergent plate boundaries (blue line). **b.** Carbon is sequestered into four main volatile-carrying reservoirs in the oceanic plate: deep-sea carbonate sediments[6] (brown), hydrothermally altered ocean crust[7] (yellow line), partially serpentinised mantle lithosphere (related to plate bending[46]: pink line, and slow spreading rates at MORs[27]: grey line), and metasomatised mantle lithosphere[25,31] (cyan line). We assume that 0–20% of deep-sea sedimentary carbon is sequestered from the atmosphere, with the remainder being sourced from shallow marine environments. **c.** The rate of carbon recycling into the mantle is controlled by the concentration of carbon within each reservoir, variations in the plate convergence rate and trench lengths. The increase in the subduction of sediment-hosted inorganic carbon is due to the substantial increase in deep-sea carbonates after 120 Ma[6]. Top panel indicates geological time periods (□ – Cambrian, O – Ordovician, S – Silurian, D – Devonian, C – Carboniferous, P – Permian, Tr – Triassic, J – Jurassic, K – Cretaceous, Pg – Palaeogene, N – Neogene).

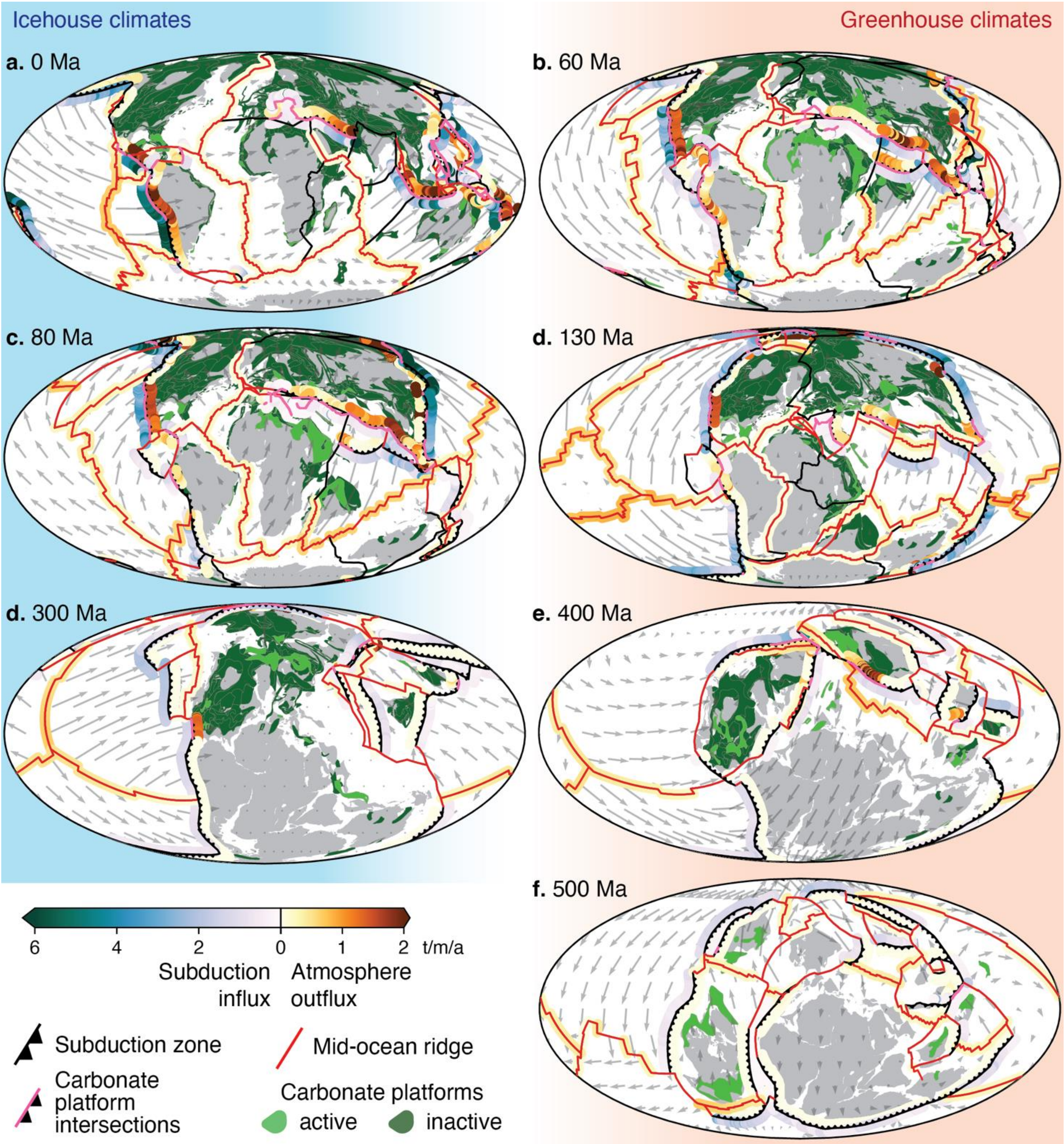


Figure 2: Location of carbonate platforms and carbon fluxes during key icehouse-greenhouse periods[5]**.** Subduction influx refers to the carbon being recycled at trenches (greens–blues) in tonnes of carbon per metre along trenches per year. Atmospheric outflux refers to carbon emitted at MORs, continental rifts, and volcanic arcs (orange colours) in tonnes of carbon per metre along each topological segment per year. Carbonate platforms are indicated by green polygons, where light green indicates active platforms, and dark green light green indicates older inactive platforms. Active and inactive carbonate platforms are treated the same in our model. Note that the total carbonate platform area increases cumulatively towards the present day and represents platforms that are currently preserved and have been mapped. The initial regions would have been larger prior to the platforms being partially eroded and buried. Volcanic arcs emit more carbon where subduction zones intersect carbonate platforms, highlighted by violet trench segments. Arc melts interact with carbonate platforms on continental arcs at shallow depth, which dissolves CO_2 in the magma passing through them and contributes to additional carbon by magma degassing.

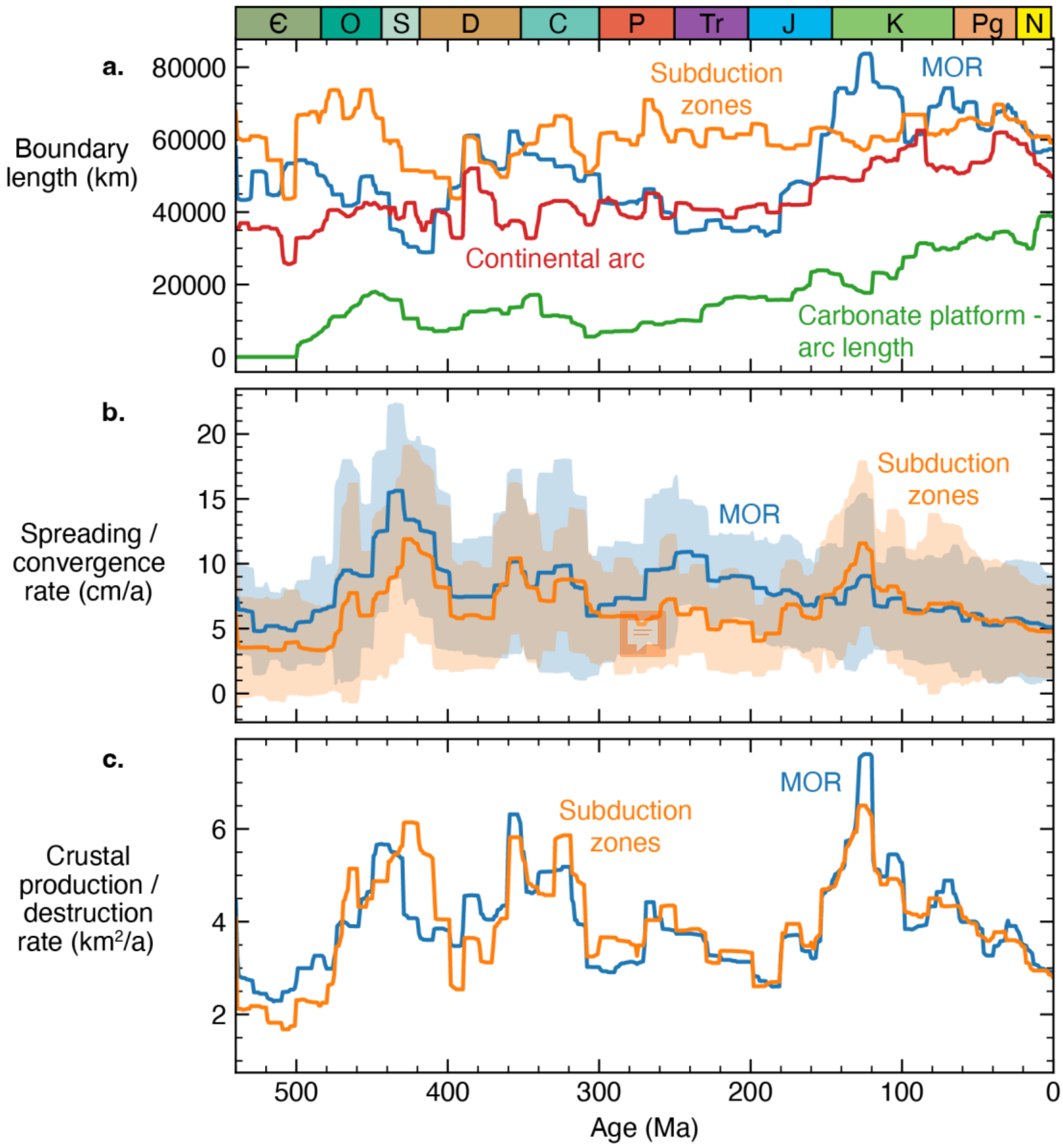


Figure 3: Plate tectonic statistics through the Phanerozoic. a. Lengths of mid-ocean ridges (blue), subduction zones (orange), continental arcs (red), and carbonate platforms which intersect continental arcs (green). **b.** Spreading and convergence rates along MORs and subduction zones, respectively. **c.** Rates of crustal production at MORs (blue) compared to crustal destruction rates at subduction zones (blue).

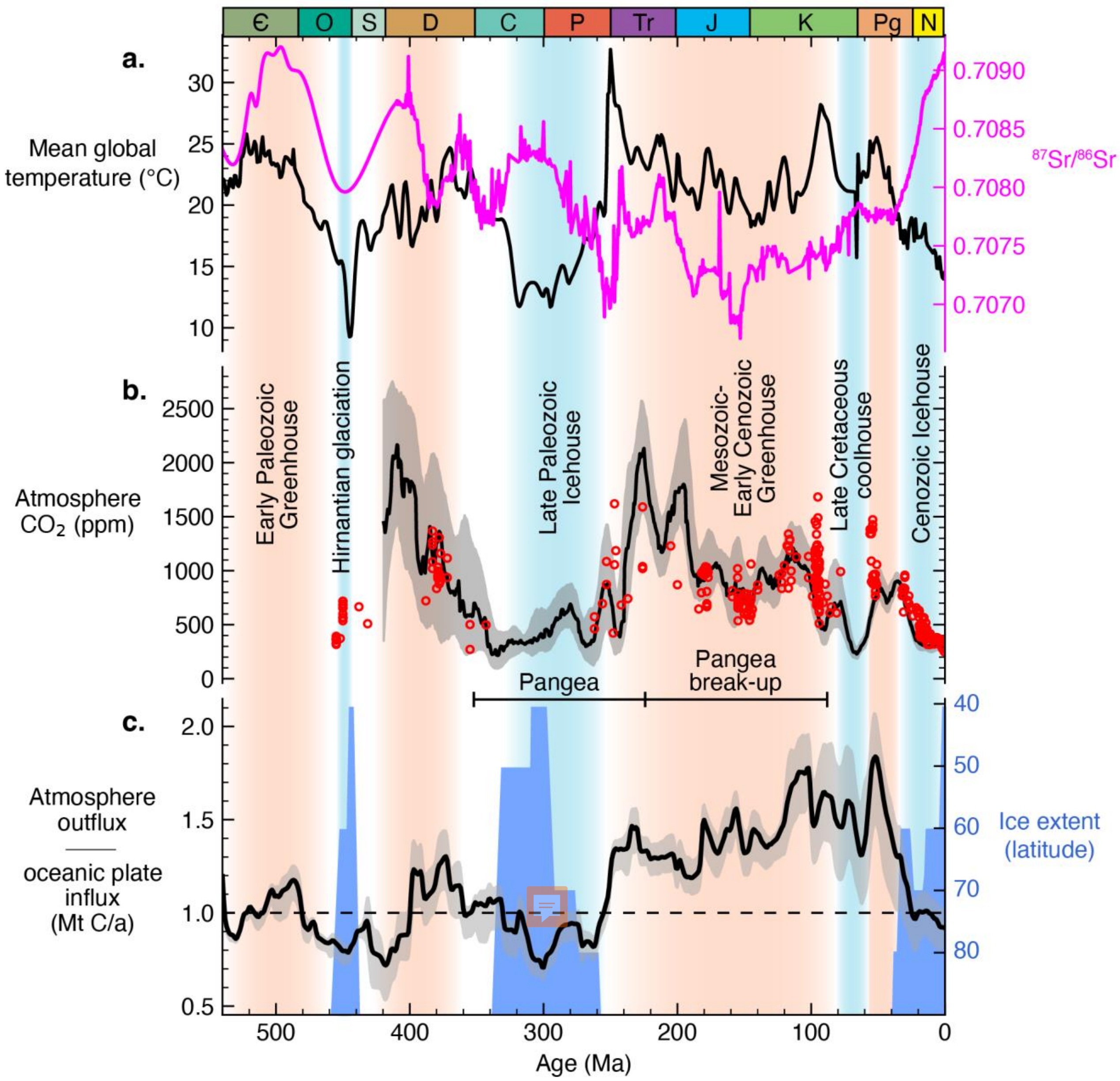


Figure 4: Icehouse – greenhouse climate states in the Phanerozoic. Changes in the concentration of atmospheric carbon are driven by the cycling of carbon between the Earth's mantle and surface. **a.** Variations in surface temperature (black line)[34] and the seawater $^{87}Sr/^{86}Sr$ curve (magenta line)[47,48]. **b.** Atmospheric CO_2 concentration from a compilation of sources (black line)[35] overlain with CO_2 estimates from phytoplankton (red circles)[49]. **c.** The ratio of atmospheric outflux to oceanic plate influx (from this study) superimposed with the magnitude of glaciation events measured by the latitudinal extent of ice sheets from poles to the equator[36]. Where the carbon outflux / influx ratio is above 1 predicts a greenhouse climate; where the ratio is below 1 predicts an icehouse climate.

METHODS

Reservoirs of water in oceanic lithosphere

Oceanic mantle – Chemically-bound water is stored in the top and bottom of oceanic mantle lithosphere. Hydration of the base of the oceanic mantle lithosphere is caused by metasomatism as new seafloor is created at mid-ocean ridges[1],

$$F = A + B_u(u - u_{\mathrm{ref}}) + B_d(d - d_{\mathrm{ref}}) + B_T(T - T_{\mathrm{ref}}) + B_c(c - c_{\mathrm{ref}}) + B_v(v - v_{\mathrm{ref}}) + C_v(v^2 - v_{\mathrm{ref}}^2)$$

where F is the water influx, u is the spreading rate, d is the grainsize of mantle lithosphere, T is the ambient mantle temperature, c is the heat capacity, and v is the volatile content. Reference values for each of these parameters (denoted with $_{\mathrm{ref}}$) and other constants are provided in Table S1. The top of the oceanic mantle is affected by serpentinisation caused by mantle unroofing during the formation of new seafloor. We employ a model which relates the half-spreading rate (cm/a), u, to mantle hydration using the following function,

$$F = \frac{A}{u + B}$$

where F is the water influx, $A = 3.18 \times 10^5$ and $B = 1$. The coefficients A and B were obtained by fitting a curve to data on the mantle hydration and hydrothermal circulation at MORs[2].

Oceanic crust – The concentration of water in altered oceanic crust is partitioned into pore and chemically bound water, which we calculate using a logarithmic model of oceanic crust age[3]. It assumes 0.3 km of upper volcanics, 0.3 km of lower volcanics, 1.4 km of dikes, and 5 km of gabbro. The bulk concentration of pore water is primarily controlled by rock type and the decay of porosity with oceanic crust age. The bulk concentration of chemically-bound water is principally controlled by rock type and density which varies logarithmically with oceanic crust age.

Deep sea sediments – The concentration of water in deep-sea sediments is partitioned into pore and chemically bound water based on the thickness of sediments calculated from the distance to the nearest passive margin and age of the oceanic crust[4]. From the total sediment thickness, we estimate the pore water content based on a relationship that describes the exponential decay of porosity with depth[5]. The result is the thickness of the water column obtained from the integrated pore space within deep-sea sediments, which is multiplied by the density of seawater (1,025 kg/m^3) to obtain an area density of water. The remaining thickness is chemically-bound water within compacted sediments which we assume is 7 wt. % with an average density of 1,700 kg/m^3. The flux of water entering subduction zones at the present day is 424 Mt/a and 131 Mt/a for pore and chemically-bound water, respectively, which is consistent with the ranges described in other studies[6,7].

Slab devolatilisation at sub-arc depths

Carbon and water loss at sub-arc depth (≤ 125 km) is calculated using a simplified thermodynamic model of three major volatile-carrying reservoirs in subducting slabs: deep-sea carbonate sediments[8], hydrothermally altered ocean crust[9], and partly serpentinised and metasomatised lithospheric mantle[10]. We employ a thermodynamic lookup table approach to calculate the release of water and carbon from each reservoir in the subducting slab as a function of pressure and temperature (Supplementary Figure 4). The bulk composition and solution models used to compute these lookup tables from Perple_X version 6.9[11] are identical to those published in a previous study[9]. The model utilises an internally consistent thermodynamic database[12] on a grid of 313 × 313 nodes corresponding to a resolution of 287 bars and 3.5 K. Devolatilisation in computed by subtracting the initial bulk H_2O and CO_2 wt. % from that calculated at a depth of 125 km, which is taken as a representative average of the sub-arc depth in the mantle corresponding to the maximum generation of arc magmas[13]. This number represents the total reduction in slab water and carbon concentration from the trench to sub-arc depths (≤ 125 km). The P-T conditions of the down-going plate at sub-arc depths (≤ 125 km) is calculated from the age and dip angle of the slab. We approximate slab temperatures at the plate surface, T_s, crust-sediment interface, T_c, and the oceanic Moho, T_m, using three logarithmic fitting parameters,

$$T_s = 1331 - 58.6 \ln \Phi$$
$$T_c = 1303 - 60.23 \ln \Phi$$
$$T_m = 1622 - 132.5 \ln \Phi$$

where the temperature of each interface at 125 km depth is related to the thermal parameter, ϕ, which is the product of the oceanic seafloor age, A_{plate}, orthogonal convergence velocity, v_c, and the slab dip angle[14], θ_{dip},

$$\Phi = A_{\mathrm{plate}} v_c \theta_{\mathrm{dip}}$$

Slab dip is calculated using a machine learning approach based on the kinematic and rheological properties of the subducting plate[15]. The oceanic seafloor age and convergence velocity are obtained from the plate model[16].

The slab outflux, q, is then obtained by (1) subtracting the initial bulk H_2O and CO_2 wt. % from that calculated at 125 km, (2) multiplying by the slab density, ρ_{slab}, and convergence rate, v_c, then (3) integrating along the length of all subduction zones, L,

$$q_{\mathrm{H_2O}} = \int_{i=0}^{L} \left[X_i^{\mathrm{H_2O}}(P,T) - X_i^{\mathrm{H_2O}}\right] \rho_{slab_i} v_{c_i}$$

$$q_{\mathrm{CO_2}} = \int_{i=0}^{L} \left[X_i^{\mathrm{CO_2}}(P,T) - X_i^{\mathrm{CO_2}}\right] \rho_{slab_i} v_{c_i}$$

This routine is repeated for each volatile-carrying reservoir in the slab (deep-sea sediments, altered oceanic crust, and serpentinized/metasomatized mantle lithosphere), which are then summed together to estimate the total slab outflux.

Volatile uptake from plate bending-related serpentinisation at subduction zones

Serpentinisation related to plate bending at subduction zones stores additional carbon and water in the down-going plate[6,17], which we scale by the slab dip angle[15]. Serpentinisation within bending-induced faults occur within the brittle part of the slab, which we take as slab temperatures below the 600°C isotherm computed from a plate model of lithospheric cooling[18]. Assuming approximately 10% serpentinite within the brittle layer, we obtain ranges of volatile uptake in the outer rise that is consistent with previous estimates (~ 7.5 Mt C/a[17] and ~ 1,000 Mt H_2O/a[6]). A correction is also applied to the sediments based on whether a subduction zone is accreting or eroding sediment, which is proportional to the orthogonal convergence rate[19]. No sediment is accreted for convergence velocities > 6.8 cm/a, and up to 40% of sedimentary carbon is accreted to the overriding plate for convergence velocities approaching 0 cm/a. For our calculations we apply a linear gradient between these two endmembers.

Calculation of melting rates to constrain carbonate platform degassing

The quantity of water that is devolatilised from subducting plates at sub-arc depths (≤ 125 km) determines the concentration of water in the upper mantle, which controls the melting rate and magnitude of arc volcanism. We estimate the amount of water lost from the slab at sub-arc depths from thermodynamic lookup tables of three major water-carrying reservoirs (explained above). The water outflux from the slab is converted to a bulk concentration assuming an upper mantle density of 3,300 kg/m^3 at a distance inbound from the trench corresponding to a depth of 125 km and the dip angle of the subducting slab[15]. The wet melt fraction is calculated using an established model of hydrous mantle melting[20], averaged across a pressure range of 2.5–4.5 GPa at 0.5 GPa increments and assuming a mantle temperature of 1350°C. The melting rate is calculated by multiplying the melt fraction with the density of the upper mantle, the arc depth, the convergence velocity, and integrating along all subduction zones. For the present day we obtain a present-day melting rate of 128 Mt/a for all subduction zones and 84 Mt/a for subduction zones which intersect carbonate platforms. The latter is used to scale the rate of carbonate platform degassing through the Phanerozoic. This is dimensionalised to the present-day rate of carbonate platform degassing (13.4–44.5 Mt C/a). A similar relationship is observed if we scale the present-day carbonate platform degassing rate by the intersecting arc length (Supplementary Figure 2). A departure between the outflux constrained by melting rates and arc lengths is particularly pronounced between 80–90 Ma due to fast convergence rates and increased melt production along western North America and eastern Eurasia, which favours increased carbonate platform degassing. Both curves assume that carbonate platforms withstand tectonic destruction and do not experience time-dependent depletion.

Calculation of contribution of pelagic carbonate sediment accumulation to plate influx

If the enormous increase in the pelagic carbonate carbon reservoir since the Late Cretaceous[8] merely reflected a shift from shallow to deep water carbonate deposition[21], then the growth of this reservoir would have not resulted in any net increase in carbon sequestration. However, it is more likely that a portion of this increase did represent a net increase[8], which has been substantiated by a global erosion model[22] demonstrating an enormous increase in continental erosion, and thus associated carbon drawdown from the atmosphere in the Cenozoic, which would have resulted in a net increase in marine carbonate carbon sequestration. We apply an empirical estimate for this carbon sink, using the following rationale: a eustatic sea-level fall after the Late Cretaceous sea-level highstand[23] would have enlarged the continental area available for continental silicate weathering. In addition, an increase in net accumulation rates of pelagic carbonate carbon starting in the Paleogene may have been driven by the emergence of major new orogens[24] and associated river systems[8]. Both effects were captured in the global erosion model for the last 100 million years[22], which modelled a doubling of terrigenous sediment flux to the oceans in the Cenozoic. Here, we assume that the portion of the pelagic carbonate carbon flux since 90 Ma that reflects a net increase (as opposed to a shift of carbonate sedimentation from shallow to deep water) gradually rose from 0% at 90 Ma to 20% of the total pelagic carbonate carbon flux after 50 Ma. The initial increase from 90 to 50 Ma would have been mainly driven by a global regression while the subsequent increase was driven by the combined effect of an increased area of orogens[24] and a continuing global regression and eustatic sea-level fall[23].

Calculation of ratio between atmospheric carbon outflux and oceanic plate influx

To calculate this ratio, the total carbon outgassing at mid-ocean ridges and volcanic arcs (derived from slab devolatilization and carbonate platform degassing) is divided by the rate of plate influx, which is contained within the upper portion of the oceanic plates. We include all carbon stored within deep-sea sediments[8], hydrothermally altered oceanic crust[9], and serpentinite due to slow spreading along MORs[25] and plate bending at subduction zones oceanic[17]. We exclude carbon stored within the base of the oceanic mantle lithosphere in this calculation because this carbon rarely interacts with the Earth's surface and plays no significant role in modulating greenhouse – icehouse climates.

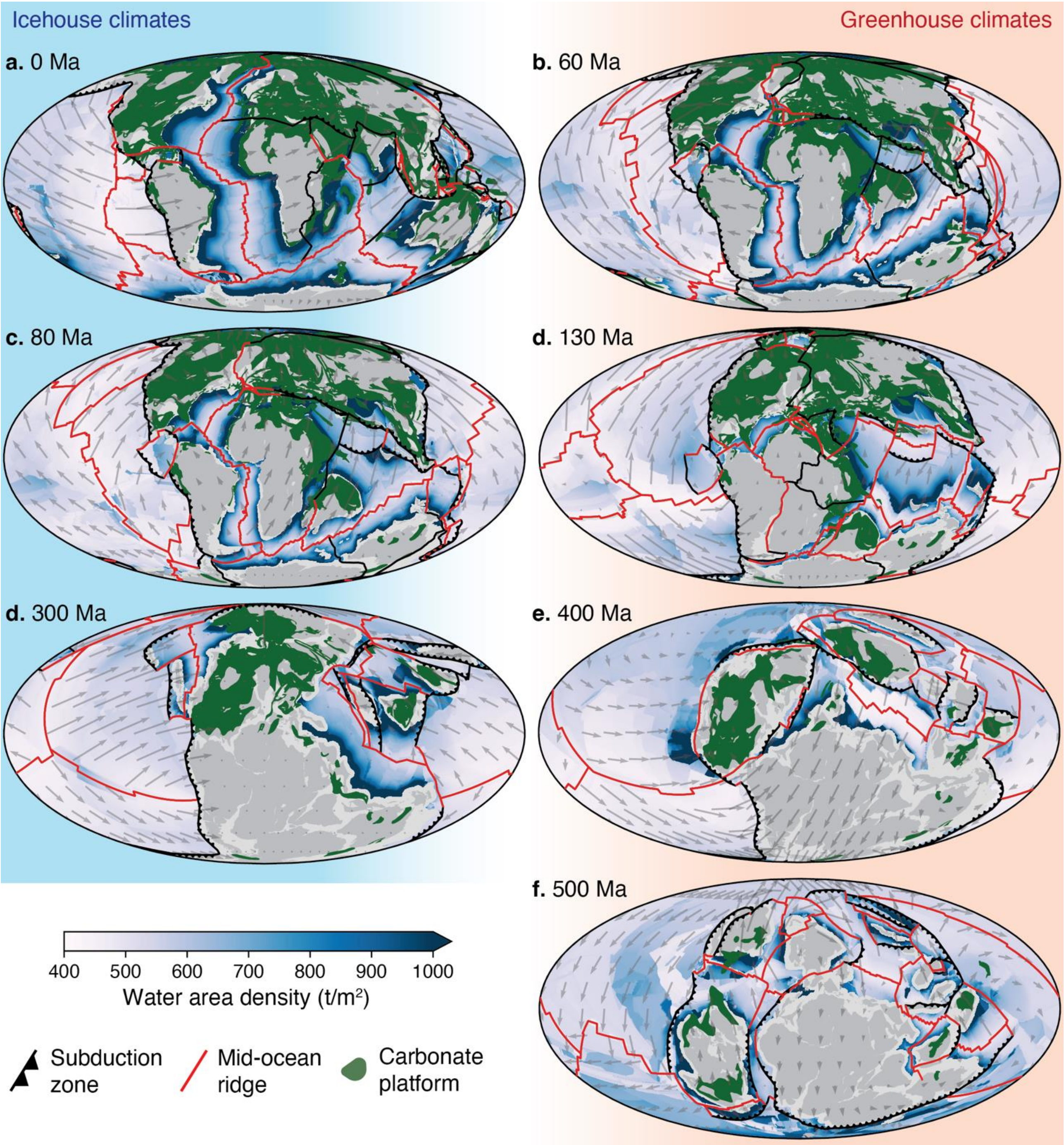
References

1. Keller, T., Katz, R. F. & Hirschmann, M. M. Volatiles beneath mid-ocean ridges: Deep melting, channelised transport, focusing, and metasomatism. *Earth and Planetary Science Letters* **464**, 55–68 (2017).
2. Iyer, K., Rüpke, L. H. & Morgan, J. P. Feedbacks between mantle hydration and hydrothermal convection at ocean spreading centers. *Earth and Planetary Science Letters* **296**, 34–44 (2010).
3. Jarrard, R. D. Subduction fluxes of water, carbon dioxide, chlorine, and potassium. *Geochemistry, Geophysics, Geosystems* **4**, (2003).
4. Dutkiewicz, A. *et al.* Predicting Sediment Thickness on Vanished Ocean Crust Since 200 Ma. *Geochemistry, Geophysics, Geosystems* **18**, 4586–4603 (2017).
5. Bahr, D. B., Hutton, E. W. H., Syvitski, J. P. M. & Pratson, L. F. Exponential approximations to compacted sediment porosity profiles. *Computers & Geosciences* **27**, 691–700 (2001).
6. Faccenda, M. Water in the slab: A trilogy. *Tectonophysics* **614**, 1–30 (2014).
7. Hacker, B. R. H2O subduction beyond arcs. *Geochemistry, Geophysics, Geosystems* **9**, (2008).
8. Dutkiewicz, A., Müller, R. D., Cannon, J., Vaughan, S. & Zahirovic, S. Sequestration and subduction of deep-sea carbonate in the global ocean since the Early Cretaceous. *Geology* **47**, 91–94 (2019).
9. Müller, R. D. *et al.* Evolution of Earth's tectonic carbon conveyor belt. *Nature* **605**, 629–639 (2022).
10. Alt, J. C. *et al.* The role of serpentinites in cycling of carbon and sulfur: Seafloor serpentinization and subduction metamorphism. *Lithos* **178**, 40–54 (2013).
11. Connolly, J. A. D. The geodynamic equation of state: What and how. *Geochemistry Geophysics Geosystems* **10**, 1–19 (2009).
12. Holland, T. J. B. & Powell, R. An internally consistent thermodynamic data set for phases of petrological interest. *Journal of Metamorphic Geology* **16**, 309–343 (1998).
13. Thompson, A. B. Water in the Earth's upper mantle. *Nature* **358**, 295–302 (1992).
14. Van Keken, P. E., Hacker, B. R., Syracuse, E. M. & Abers, G. A. Subduction factory: 4. Depth-dependent flux of H2O from subducting slabs worldwide. *Journal of Geophysical Research: Solid Earth* **116**, (2011).
15. Mather, B. R., Müller, R. D., Alfonso, C. P., Seton, M. & Wright, N. M. Kimberlite eruptions driven by slab flux and subduction angle. *Scientific Reports* **13**, 1–12 (2023).
16. Müller, R. D. *et al.* A tectonic-rules-based mantle reference frame since 1 billion years ago – implications for supercontinent cycles and plate–mantle system evolution. *Solid Earth* **13**, 1127–1159 (2022).
17. Kelemen, P. B. & Manning, C. E. Reevaluating carbon fluxes in subduction zones, what goes down, mostly comes up. *Proceedings of the National Academy of Sciences of the United States of America* **112**, E3997–E4006 (2015).
18. Grose, C. J. Properties of oceanic lithosphere: Revised plate cooling model predictions. *Earth and Planetary Science Letters* **333–334**, 250–264 (2012).
19. Clift, P. & Vannucchi, P. Controls on tectonic accretion versus erosion in subduction zones: Implications for the origin and recycling of the continental crust. *Reviews of Geophysics* **42**, (2004).
20. Katz, R. F., Spiegelman, M. & Langmuir, C. H. A new parameterization of hydrous mantle melting. *Geochemistry, Geophysics, Geosystems* **4**, 1–19 (2003).
21. Opdyke, B. N. & Wilkinson, B. H. Surface area control of shallow cratonic to deep marine carbonate accumulation. *Paleoceanography* **3**, 685–703 (1988).
22. Salles, T. *et al.* Hundred million years of landscape dynamics from catchment to global scale. *Science* **379**, 918–923 (2023).
23. Kocsis, Á. T. & Scotese, C. R. Mapping paleocoastlines and continental flooding during the Phanerozoic. *Earth-Science Reviews* **213**, 103463 (2021).

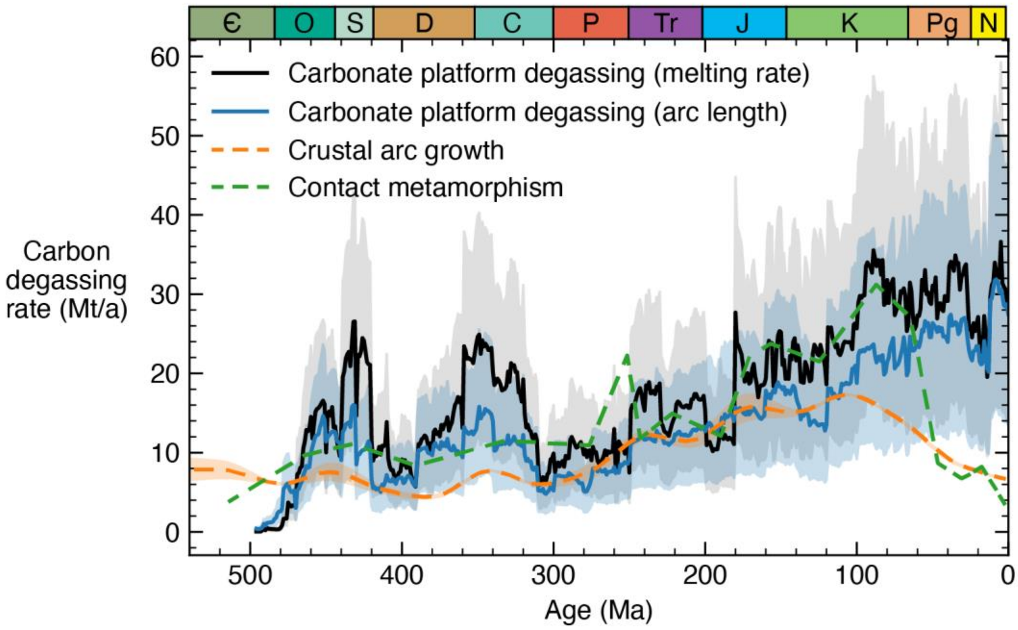
24. Müller, R. D. *et al.* A Global Plate Model Including Lithospheric Deformation Along Major Rifts and Orogens Since the Triassic. *Tectonics* **38**, 1884–1907 (2019).
25. Merdith, A. S., Atkins, S. E. & Tetley, M. G. Tectonic Controls on Carbon and Serpentinite Storage in Subducted Upper Oceanic Lithosphere for the Past 320 Ma. *Frontiers in Earth Science* **7**, 1–23 (2019).
26. Bekaert, D. V. *et al.* Subduction-Driven Volatile Recycling: A Global Mass Balance. *Annual Review of Earth and Planetary Sciences* **49**, (2021).
27. Wong, K. *et al.* Deep Carbon Cycling Over the Past 200 Million Years: A Review of Fluxes in Different Tectonic Settings. *Frontiers in Earth Science* **7**, 1–22 (2019).
28. Zahirovic, S. *et al.* Subduction and carbonate platform interactions. *Geoscience Data Journal* **9**, 371–383 (2022).
29. Ratschbacher, B. C., Paterson, S. R. & Fischer, T. P. Spatial and Depth-Dependent Variations in Magma Volume Addition and Addition Rates to Continental Arcs: Application to Global CO2 Fluxes since 750 Ma. *Geochemistry, Geophysics, Geosystems* **20**, 2997–3018 (2019).
30. Cao, W., Lee, C.-T. A. & Lackey, J. S. Episodic nature of continental arc activity since 750 Ma: A global compilation. *Earth and Planetary Science Letters* **461**, 85–95 (2017).
31. Chu, X., Lee, C.-T. A., Dasgupta, R. & Cao, W. The contribution to exogenic CO2 by contact metamorphism at continental arcs: A coupled model of fluid flux and metamorphic decarbonation. *Am J Sci* **319**, 631–657 (2019).
32. Connolly, J. A. D. Computation of phase equilibria by linear programming: A tool for geodynamic modeling and its application to subduction zone decarbonation. *Earth and Planetary Science Letters* **236**, 524–541 (2005).

Parameter	**Symbol**	**Units**	**Ref value**	**Value**	**A**	**B**	**C**
Half-spreading rate	*u*	cm/a	3	varies	0.881	0.262	
Compaction length	*d*	log10 km	1.5044	1.5044		0.359	
Potential temperature	*T*	°C	1350	1350		-2.91×10^{-4}	
Fertility	*c*	wt. %	19	19		7.50×10^{-3}	
Volatile content	*v*	wt ppm	100	150		5.40×10^{-3}	0.714

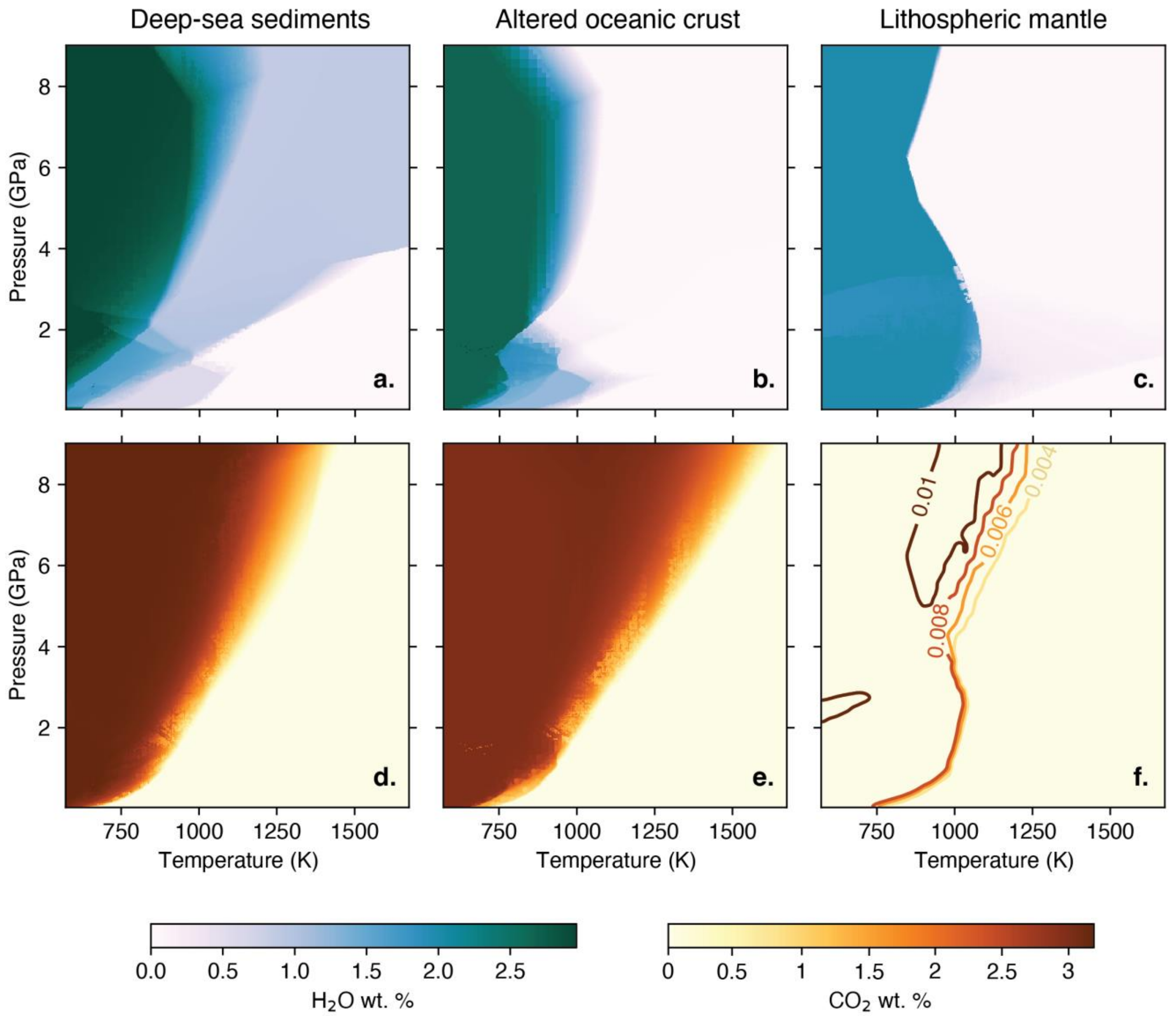
Table S1. Coefficients and reference values for calculating the flux of water into the bottom of new oceanic mantle lithosphere at mid-ocean ridges[1].



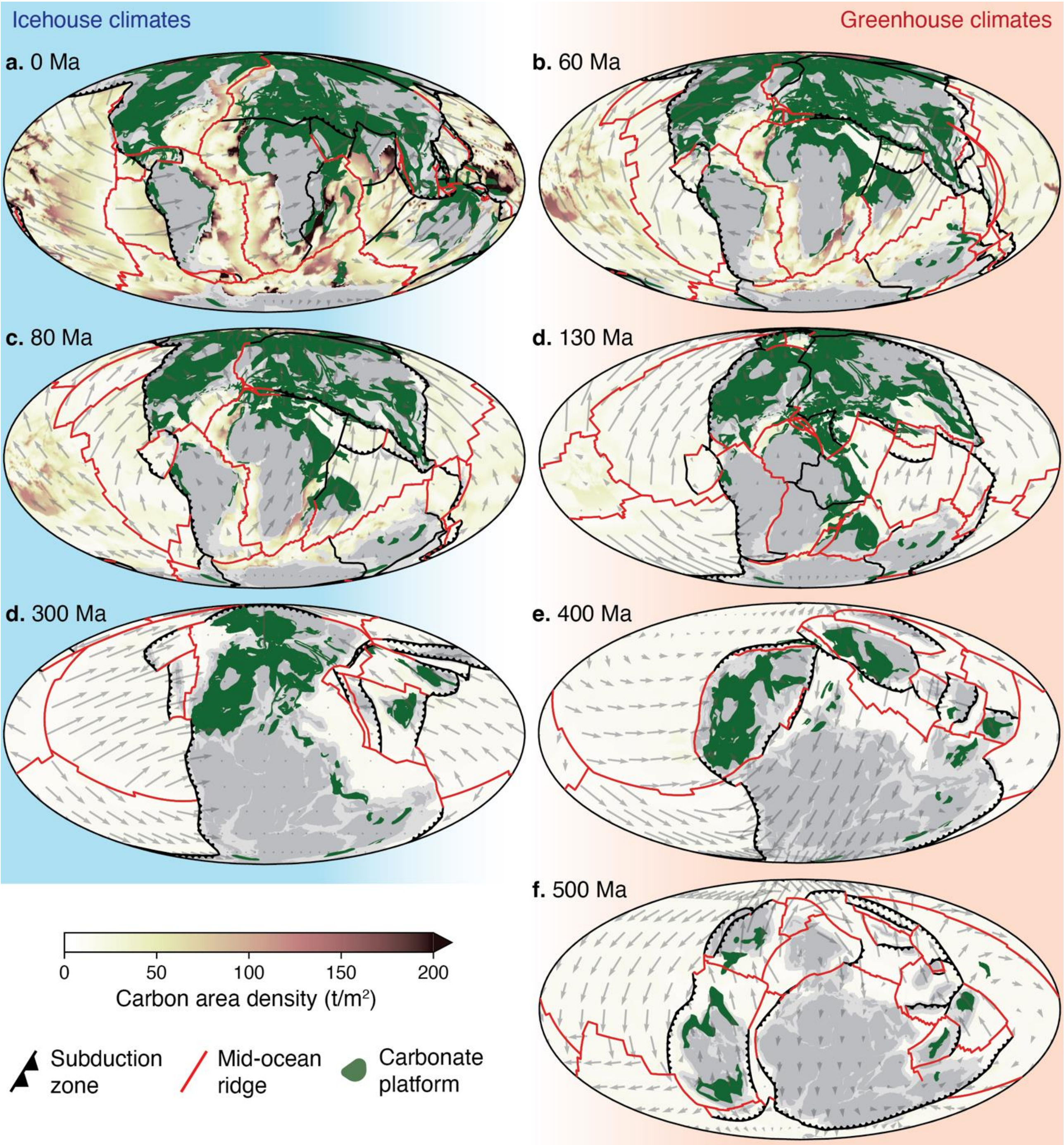
Supplementary Figure 1. Total concentration of water in oceanic plates reconstructed through major icehouse–greenhouse climates. The density of water contained within the oceanic plates (in tonnes per metre squared) corresponds to the timeseries in Figure 2.



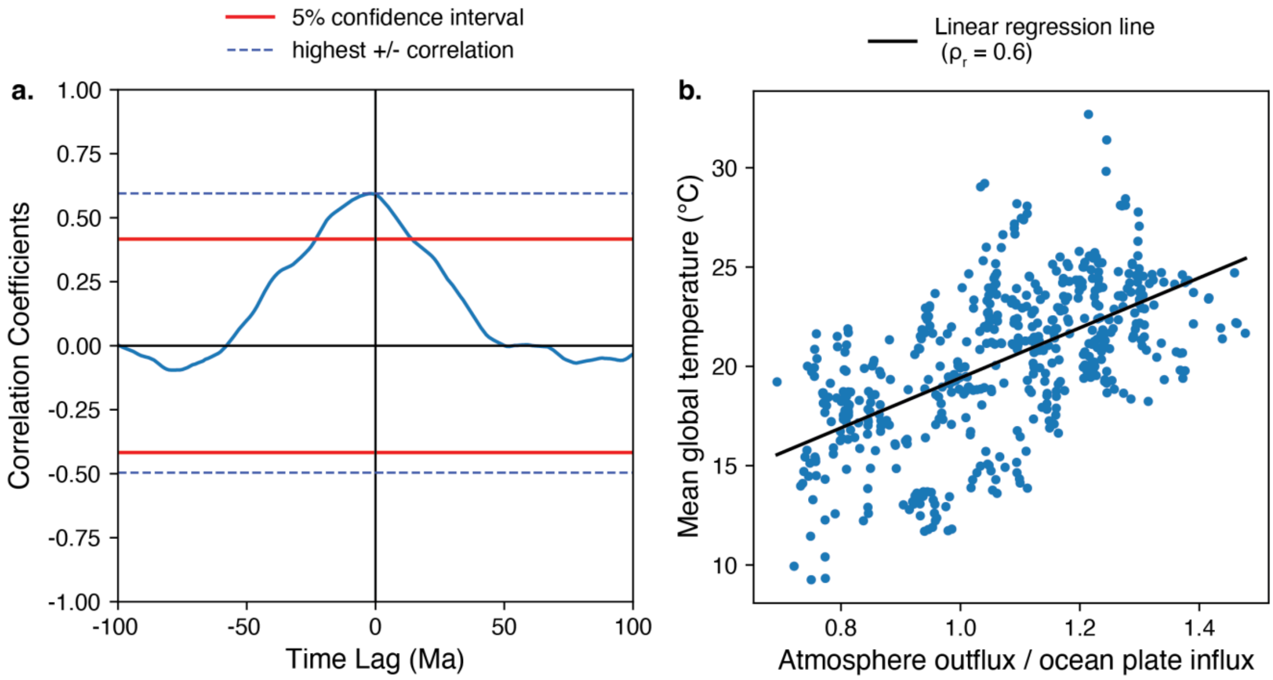
Supplementary Figure 2. Rates of carbonate platform degassing along volcanic arcs. The degassing rate is calculated by scaling the present-day carbonate platform outflux (13.4–44.5 Mt C/a)[26] against the melting rate[20] (black line) and arc length (blue line). The linear carbon outflux trend holds if we scale the carbon loss at carbonate platforms by intersecting arc length to be consistent with previous studies[27,28] (Supplementary Figure 2). The "crustal arc growth" curve is based on arc magma addition rates[29] scaled by the total arc length[30] (dashed orange line). The "contact metamorphism" curve represents an alternative model of carbonate platform outflux due to contact metamorphism[31], which is scaled by zircon populations.



Supplementary Figure 3. H_2O and CO_2 concentration of volatile-carrying reservoirs over a range of pressure and temperature. Phase diagrams were calculated using Perplex[32] and are identical to those published in a previous study[9]. The concentration of H_2O (**a**-**c**) and CO_2 (**d**-**f**) were extracted across a range of upper mantle temperature and pressure conditions to determine the loss of water and carbon at sub-arc depths. Panel **f.** is contoured by the wt. % of CO_2 because it is orders of magnitude smaller than deep-sea sediments (**d**) or altered oceanic crust (**e**).



Supplementary Figure 4. Total concentration of carbon in oceanic plates reconstructed through major icehouse–greenhouse climates. The density of carbon contained within the oceanic plates (in tonnes per metre squared) corresponds to the timeseries in Figure 2.



Supplementary Figure 5. Statistical relationship between atmosphere outflux / subduction influx ratio and mean global temperature. a. the cross correlation examines the phase offset between the two signals. The strongest correlation (ρ_r = 0.6) occurs for zero time offset, which indicates the ratio of carbon outflux to subduction influx is synchronised with global temperature (to within a maximum temporal of 1 Myr). **b.** Plot of mean global temperature against the ratio of atmosphere outflux to subduction influx. A linear regression line through the data has a Pearson correlation coefficient, ρ_r, of 0.6.