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Peer Review File

Time-space constraints point to a dynamic evolution of global mantle isotopic anomalies linked to supercontinent-superocean evolution since 700 Ma

Corresponding Author: Mr Piero Azevedo Berquo de Sampaio

Attachments originally included by the reviewers as part of their assessment appear 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 Mr Azevedo Berquo de Sampaio,

Your manuscript titled "Time-space constraints point to a dynamic evolution of global mantle isotopic anomalies linked to supercontinent-superocean evolution since 700 Ma" has now been seen by 3 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.

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:
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We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

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,

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

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

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors provide an examination of the isotopic makeup of the Earth's mantle, which sheds light on its complex history over the past 700 million years. They built an important isotope database and based their geodynamic model on methods like paleogeographic reconstructions. This kind of approach is often poorly executed in geochemical/petrological studies, particularly in scientific investigations into to mantle dynamics, evolution, and composition. Additionally, they also demonstrate that the mantle's isotopic composition has evolved through significant enrichment, leading to mantle heterogeneity. The authors explain this pattern as the result of subduction zone retreat and the addition of crustal material during the formation and disintegration of supercontinents.
Overall, the article is clear and well-written, demonstrating maturity in conveying ideas. The text and figures are of excellent quality, indicating that the authors have thoroughly reviewed and revised the manuscript. In summary, this research expands our knowledge of the Earth's mantle evolution and its connection to broader geodynamic processes, contributing to ongoing debates in the scientific community. However, the presentation of the work could be improved with a some adjustments. I

recommend accepting the manuscript after a minor revision that addresses the following comments/suggestions.

Reviewer #2 (Remarks to the Author):

This is a thought-provoking manuscript that combines geochemical data from ophiolites and present-day oceans with paleogeographic reconstructions to illuminate the geochemical evolution of the African mantle domain. The hypothesis is that the geochemical evolution of the African mantle domain is linked to subduction history associated with the breakup of Rodinia by extroversion (closure of the external superocean). The test is the analysis of 947 Nd isotopic data from 88 ophiolites that are related either to mantle plumes or to oceanic plates. A new Delta_DM index is introduced to express the depletion of the mantle over time. The results suggest that mantle plumes became more enriched than oceanic plates from around 400 Ma and that this enrichment peaked at around 90 Ma. The interpretation is that there was increased subduction of continental material from around 650 Ma associated with the breakup of supercontinent Pangea, and that it took 200-300 Myr for this continental material to sink to the source of deep mantle plumes and come back to the surface.

I found the article novel, well written and well presented. It brought a few questions to mind.

It would be helpful to show how the distinction is made between plume-related and plate-related ophiolites. Is this distinction unequivocal? This is important because the findings of the study hinge on this distinction. Supplementary Figure 3 shows that subsets of plate-related ophiolites are not distinguishible. It would be helpful to present a similar figure showing the distinction between plume-related and plate-related ophiolites.

On Figure 1c, ophiolites are linked to oceans. How was this done? Could the ophiolites be shown on a figure or animation similar to Supplementary Figure 1 to illustrate this association?

Is there sufficient data to resolve the trends in ophiolite depletion? There are generally fewer data back in time, and not many data points between 300 Ma and 200 Ma.

Are the results only compatible with the geodynamic scenario of Li et al. (2019)? Is there a reason not to consider alternative tectonic scenarios, such as the ones mentioned in Merdith et al. (2021, https://doi.org/10.1016/j.earscirev.2020.103477) or Müller et al. (2022, https://doi.org/10.5194/se-13-1127-2022)? Taking a step back from the uncertain details of global tectonic scenarios, could the new analysis be used to test ideas/debates about the closure of supercontinent Rodinia by extroversion or introversion?

Finally, why is the enrichment only observed in the last 400 Myr? Was continental crust not recycled in the mantle before about 650 Ma? This is alluded to in the concluding statement without explanation: 'This would imply that geochemical anomalies in the mantle might be a transient feature in the Earth’s mantle, reflective of the planet’s geodynamic evolution instead of static features since earth Earth.' (L. 202-204)

Reviewer #3 (Remarks to the Author):

This manuscript presents a comprehensive analysis of the isotopic evolution of the Earth's mantle, focusing particularly on the African Mantle Domain (AMD) and its relationship with supercontinent-superocean evolution since 700 million years ago (Ma). The authors analyse the geochemical composition of mid-ocean ridge basalts and plume-induced basalts, both from present-day oceans and preserved in ancient ophiolites. By comparing the isotopic signatures of these rocks across time, the researchers aim to reconstruct the history of mantle enrichment processes. The plume-derived rocks in the African Mantle Domain show a trend of increasing enrichment from 400 Ma to 100 Ma, followed by a depletion trend. This enrichment is attributed to the subduction of continental crust during the assembly of Gondwana and Pangea. The authors propose a model where the Rodinia-to-Pangea supercontinent cycle played a crucial role in creating the isotopic dichotomy between the African Mantle Domain and the Pacific Domain. Overall, this paper provides a valuable contribution to our understanding of mantle evolution and the formation of isotopic heterogeneity. The proposed model is convincing and well-supported by the data. It is well-structured, thoroughly researched, and effectively communicates complex scientific concepts to the reader. The incorporation of multiple lines of evidence, including isotopic data, geological reconstructions, and regression analyses, adds credibility to the study's findings. Overall, the paper makes a significant contribution to the field of mantle geochemistry and our understanding of Earth's dynamic evolution. However, I still have some minor concerns on this manuscript which needs to be addressed in detail before it is suitable for publication. They are as follows:
1. They authors acknowledges the restricted window into the past provided by present-day ocean floor basalts (limited to the last 200 Ma) but doesn't delve into alternative methods for studying the deeper mantle, such as Sr, Pb isotopes which are crucial in understanding of the mantle isotopic heterogeneity.
2. Secondly, while the continental crust subduction hypothesis is well-supported, a more detailed exploration of the mechanisms by which this crustal material is recycled into the mantle would be valuable. The authors could explore additional mechanisms for continental crust recycling into the mantle, besides subduction.
3. While the focus on the African Mantle Domain provides valuable insights, it would be beneficial to discuss the implications of the findings for mantle dynamics on a global scale. The paper could further discuss potential uncertainties in the data and analysis methods and their implications for the interpretation of results.
4. Please provide a more detailed discussion of potential uncertainties in the data, methodology, and interpretations, and how they were addressed or accounted for in the analysis.

Overall, this paper offers a significant contribution to our understanding of mantle dynamics and the formation of isotopic heterogeneity. The proposed model is persuasive and well-supported by the presented data. I recommend this article to be accepted if the above points are addressed and significantly modified.

Jiten Pattnaik
9 April 2024

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Author Rebuttal letter: The author's response to these comments can be found at the end of this file.

Version 1:

Decision Letter:

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Dear Mr Azevedo Berquo de Sampaio,

Your manuscript titled "Time-space constraints point to a dynamic evolution of global mantle isotopic anomalies linked to supercontinent-superocean evolution since 700 Ma" 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 under the open access CC BY license (Creative Commons Attribution v4.0 International License).

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

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

Best regards,

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

REVIEWERS' COMMENTS:

Reviewer #2 (Remarks to the Author):

The authors have carefully addressed my comments, which I hope contributed to improving the manuscript.

My remaining suggestions are minor.

- Consider showing ophiolites within 50 Myr of their age in Figure 3 to tie the manuscript together.

- L. 233-236: 'geochemical anomalies in the mantle might be a transient feature in the Earth’s mantle'. Consider making this statement more precise because some geochemical heterogeneities formed within the first 50 Myr of Earth's history seem to be preserved in some modern flood basalts (Rizo et al., 2016, Science). This generally links to a comment on the first version of the manuscript that I read, which I would like to recast as a question: 'How confident can we be that there was no subduction of continental material in the African Domain at any point between 4.56 Ga and 0.9 Ga?'

L. 181: Jackson and Macdonald (2022) argued for a delay of 300 Myr. They provided adequate references for slab transit times. Koppers et al. (2021) discussed plume transit times, which might be helpful.

Minor linear suggestions:

L. 43 and L. 260: 'existence' instead of 'tenure'?

L. 68: '900'... 'Ma'?

L. 86: is using the possessive form for objects grammatically correct? 'Given the representativeness of the Nd isotopic system in the dataset' (by the way, that is still a quite obscure statement)

L. 144, L. 158 and elsewhere: 'suggested' (use the past tense to refer to work that was carried out and published at some point in the past)

L. 168: 'occur' rather than 'happen'?

L. 175: 'started' instead of 'starts'

L. 181: 'sinking' instead of 'downgoing'

L. 184: 'roughly 300 Ma' (no full stop)

L. 197: consider deleting 'its constituent'

L. 202: 'Australie' or (ou?) 'Australia'?

L. 230: 'after late Cretaceous times'

- L. 310: the equation formatting was off (including five empty boxes) in the version that I read

References:

Koppers, A.A., Becker, T.W., Jackson, M.G., Konrad, K., Müller, R.D., Romanowicz, B., Steinberger, B. and Whittaker, J.M., 2021. Mantle plumes and their role in Earth processes. Nature Reviews Earth & Environment, 2(6), pp.382-401.

Rizo, H., Walker, R.J., Carlson, R.W., Horan, M.F., Mukhopadhyay, S., Manthos, V., Francis, D. and Jackson, M.G., 2016. Preservation of Earth-forming events in the tungsten isotopic composition of modern flood basalts. Science, 352(6287), pp.809-812.

Reviewer #3 (Remarks to the Author):

Reply 1: We assume this comment is in reference to Sr and Pb isotopes in kimberlites and older continental mantle plume-derived rocks (LIPs). We avoided using continental basalts to avoid the issue of contamination by continental crust during the ascent and storage of the magmas, which makes the identification of true EM signatures ambiguous (source vs. contamination). Deep-time analyses of kimberlite isotopic signatures have already been done elsewhere (Woodhead et al. 2019; Giuliani et al. 2020), and, as noted (Lines 191 – 192), the data indicate the appearance of isotopically enriched reservoirs in the same time frame as found by our study. We added Lines 83 – 86 to the RM, which read: “We focused on oceanic basalts because continental basalts are likely contaminated by continental crust during ascent and storage through the crust that makes any interpretation based on enriched isotopic signatures difficult.” With regards to using the Sr and Pb isotopic systems for oceanic rocks, there is a low availability of data (Lines 80 – 81 of the RM). Furthermore, the Nd isotopic system is more resilient to seawater alteration and metamorphism than the Sr and Pb systems. For these reasons, we opted to focus our analysis on Nd isotopic systematics of oceanic basalts.

Comment: Thank you for your clarification on focusing specifically on Nd isotopic systematics in oceanic basalts. The rationale regarding the contamination issues linked with continental basalts and the resilience of the Nd isotopic system to seawater alteration and metamorphism is well-founded. It's reassuring to learn that analyses of kimberlite isotopic signatures in deep time, conducted elsewhere, align with the findings of your study, further bolstering the credibility of your conclusions. Overall, the author's response effectively addresses the comment and provides a clear justification for your methodological approach.

Reply 2: In the OM we listed different ways continental crust can be recycled (Lines 157 – 160; Stern and Scholl. 2010). Please see comment 5 from Reviewer #2 to see how this point was addressed.

Comment: Thank you. I'm satisfied with this response.

Reply 3: While the focus on the African Mantle Domain provides valuable insights, it would be beneficial to discuss the implications of the findings for mantle dynamics on a global scale.
Conducting the same type of statistical analysis for the ophiolites in the Pacific Domain is, at the moment, impossible due to the scarcity of data. Supplementary Fig. 10 shows the Pacific Domain plume-derived ophiolites and present-day plume-derived rocks in comparison to the African Domain data to demonstrate how enriched isotopic signatures only become prevalent in the Pacific Domain after 10 Ma. The global implications of our observations and interpretations are stated in numerous passages of the OM. Lines 172 – 179 compare African Domain with Pacific Domain. In Lines196 – 222 we relate the whole Earth’s tectonic evolution since the break-up of Rodinia to our observations and the rise of the Earth’s isotopic dichotomy. Implications of our findings for global scale mantle dynamics is once again addressed in Lines 220 – 223. We added another passage to directly link our observations (and interpretations) to the global picture. Lines 220 – 222 in the RM now read: “…which concentrated subduction in the African Domain relative to the Pacific Domain, creating the prevalence of enriched isotopic signatures in the African Domain since late Palaeozoic.”

Comment: Thank you for providing a thorough explanation of the challenges associated with statistical analysis on the Pacific Domain ophiolites due to data scarcity. The clarification on how Supplementary Fig. 10 depicts the comparison between Pacific and African Domain data, highlighting the timing of enriched isotopic signatures in the Pacific Domain, is appreciated. Overall, it appears that the manuscript effectively covers the global implications of our research findings.

Reply 4: As noted in the response to comment 3 from Reviewer #2, we added a new analysis and an accompanying supplementary figure that considers potential sampling bias by randomly sampling the same number of samples from a bootstrapped dataset identical to the presentday Nd isotopic distribution. This analysis shows that the samples older than 300 Ma are mostly significantly different from the present-day distribution. This approach helps to overcome one of the main shortfall of the ophiolite dataset. Other sources of uncertainty include alteration of the primary isotopic signatures and statistical uncertainty. Regarding alteration of isotopic signatures, Lines 286 – 289 elaborate on the high resistance of the Nd system to seawater alteration, and we specify in Line 281 that only samples with no more than amphibolite facies metamorphism were considered, which mitigates metamorphic disturbances of the isotopic system. For the statistical analysis, all of the parameters used in the Gaussian Process regression, GAM regressions and LOWESS fitting were given in the Methods section, as well as the results of statistical tests of the GAM regressions. We believe that the coherent results between the different statistical methods (GP, GAMs, LOWESS) strengthen our interpretations (Supplementary Fig. 6). The regression using only ophiolites also shows an enrichment trend and that the observed enrichment is not an artifact of combining datasets (Supplementary Fig.7).

Comment:Thank you for providing additional insights into the new analysis and supplementary figures aimed at addressing potential sampling bias in the dataset. The incorporation of this analysis, which involves sampling from a bootstrapped dataset, significantly strengthens the study and addresses a notable limitation in the ophiolite dataset. Furthermore, the clarification on other sources of uncertainty, such as alteration of isotopic signatures and statistical uncertainty, is valued. Overall, these enhancements and clarifications enhance the robustness and credibility of our study's findings. Thank you for comprehensively addressing these aspects.

Jiten Pattnaik
9th June 2024

Author Rebuttal letter:

Author Response

Below we address Reviewer #2âs remaining minor comments. Our replies are shown in blue. The revised manuscript and supplementary materials with the tracked changes are also attached. (OM = Original Manuscript; RM = Revised Manuscript).

Reviewer #2:
The authors have carefully addressed my comments, which I hope contributed to improving the manuscript.

My remaining suggestions are minor.

Comment 1:

Consider showing ophiolites within 50 Myr of their age in Figure 3 to tie the manuscript together.

Figure 3 was modified to incorporate this suggestion. The first 3 panels now include the ophiolites within 50 Myr of the age depicted on the panel. The caption for Fig. 3 was also updated accordingly.

Comment 2:

- L. 233-236: 'geochemical anomalies in the mantle might be a transient feature in the Earthâs mantle'. Consider making this statement more precise because some geochemical heterogeneities formed within the first 50 Myr of Earth's history seem to be preserved in some modern flood basalts (Rizo et al., 2016, Science). This generally links to a comment on the first version of the manuscript that I read, which I would like to recast as a question: 'How confident can we be that there was no subduction of continental material in the African Domain at any point between 4.56 Ga and 0.9 Ga?

Thank you for bringing the need for further clarification. We have reworded L. 242 â 246in the last sentence of the RM to make our meaning clearer:

âThis would imply that geochemical anomalies in the mantle might be evolving features reflective of the planetâs geodynamic evolution through time, instead of static features present since early Earth.â
Comment 3:

L. 181: Jackson and Macdonald (2022) argued for a delay of 300 Myr. They provided adequate references for slab transit times. Koppers et al. (2021) discussed plume transit times, which might be helpful.

Jackson and MacDonald (2022), as well as references they cited, argue for a 300 Myr delay, were cited in the manuscript when providing the estimate. We have made some minor changes to make the delay estimate more explicit (L. 185 â 188 of the RM):

âThe estimates for processing time of recycled material in the mantle vary, but estimates based on experiments33, and on the calculated slab sinking rates of 1.1 to 1.9 cm/yr34 and plume upwelling times of up to 80-100 Myrs35, are in the order of 200â300 Myrs7.â

Minor suggestions:

We amended the manuscript following the minor suggestions made by the reviewer, except for switching âAustralieâ to âAustraliaâ in L. 208 of the RM, because we are following the original nomenclature of Cawood et al. (2021). We also slightly reworded L. 88 â 92 in RM to make its meaning clearer:

âThe Nd isotopic system not only has the largest number of analyses available in the dataset, it is also resistant to alteration processes such as seawater alteration18. Therefore, â¦â

References:

Cawood, P. A., Martin, E. L., Murphy, J. B., Pisarevsky, S. A. (2021). "Gondwana's interlinked peripheral orogens." Earth and Planetary Science Letters 568: 117057.

Author Response

We have addressed all reviewer's comments, with our replies shown below in blue. The revised manuscript and supplementary materials with the tracked changes are also attached. Please OM: Original Manuscript. RM: Revised Manuscript

Reviewer #1:

Comment: The authors provide an examination of the isotopic makeup of the Earth's mantle, which sheds light on its complex history over the past 700 million years. They built an important isotope database and based their geodynamic model on methods like paleogeographic reconstructions. This kind of approach is often poorly executed in geochemical/petrological studies, particularly in scientific investigations into to mantle dynamics, evolution, and composition. Additionally, they also demonstrate that the mantle's isotopic composition has evolved through significant enrichment, leading to mantle heterogeneity. The authors explain this pattern as the result of subduction zone retreat and the addition of crustal material during the formation and disintegration of supercontinents.

Overall, the article is clear and well-written, demonstrating maturity in conveying ideas. The text and figures are of excellent quality, indicating that the authors have thoroughly reviewed and revised the manuscript. In summary, this research expands our knowledge of the Earth's mantle evolution and its connection to broader geodynamic processes, contributing to ongoing debates in the scientific community. However, the presentation of the work could be improved with a some adjustments. I recommend accepting the manuscript after a minor revision that addresses the following comments/suggestions.

We greatly appreciate the reviewer's appreciation and enthusiastic support of our work, and the effort put into the reviewing of the manuscript.

Reviewer #2:

This is a thought-provoking manuscript that combines geochemical data from ophiolites and present-day oceans with paleogeographic reconstructions to illuminate the geochemical evolution of the African mantle domain. The hypothesis is that the geochemical evolution of the African mantle domain is linked to subduction history associated with the breakup of Rodinia by extroversion (closure of the external superocean). The test is the analysis of 947 Nd isotopic data from 88 ophiolites that are related either to mantle plumes or to oceanic plates. A new Delta_DM index is introduced to express the depletion of the mantle over time. The results suggest that mantle plumes became more enriched than oceanic plates from around 400 Ma and that this enrichment peaked at around 90 Ma. The interpretation is that there was increased subduction of continental material from around 650 Ma associated with the breakup of supercontinent Pangea, and that it took 200-300 Myr for this continental material to sink to the source of deep mantle plumes and come back to the surface. I found the article novel, well written and well presented. It brought a few questions to mind.

Thank you for the thoughtful and constructive feedback. It is exciting to see the ideas so well-received.

Comment 1: It would be helpful to show how the distinction is made between plume-related and plate-related ophiolites. Is this distinction unequivocal? This is important because the findings of the study hinge on this distinction. Supplementary Figure 3 shows that subsets of plate-related ophiolites are not distinguishible. It would be helpful to present a similar figure showing the distinction between plume-related and plate-related ophiolites.

We added Supplementary Fig. 5, highlighting the two groups' different trace element patterns, with the plume-related group markedly more enriched in incompatible trace elements.

Lines 95-97 in the RM read: "*The difference between the plume-related and upper mantle-derived samples is highlighted by their distinct incompatible trace element patterns (Supplementary Fig. 5).*"

The caption for Supplementary Fig. 5 in the revised Supplementary Material reads: "*Supplementary Figure 5.* **Incompatible trace element distributions** between the upper mantle- and plume-related datasets normalised to primitive mantle[1]. Median, 25th and 75th percentiles are for the distribution of each element within each dataset."

Comment 2: On Figure 1c, ophiolites are linked to oceans. How was this done? Could the ophiolites be shown on a figure or animation similar to Supplementary Figure 1 to illustrate this association?

Knowing the locations of the ophiolites and their surrounding continental terranes and geological contexts, we can use local and global full plate paleogeographic reconstructions to link the ophiolites to broader oceanic basins. There is uncertainty in the along-strike positioning of ophiolites in each suture. Therefore, we believe it is conservative to show the broader area from which they might be derived using plate boundaries and motions than to reconstruct their precise movements individually. We added Supplementary Fig. 2, which schematically represents how an ophiolite can be linked to a broader oceanic area. We also made changes to the text.

Lines 76-79 in the RM read: *"These ophiolites can be linked back to now-closed paleo-oceanic basins using the geotectonic context of their surrounding terranes and global plate-tectonic reconstruction models[2,3]. This is demonstrated schematically in Supplementary Fig. 2."*

The caption for Supplementary Fig. 2 in the Supplementary Material reads: "*Supplementary Figure 2.* **Schematic model** depicting how ophiolites can be linked to oceanic basins. Using full plate reconstruction models, we can associate an ophiolite complex to a now-consumed area of an oceanic basin. Strike-slip movements of accreted terranes along suture zones make more precise reconstructions difficult to ascertain."

Comment 3: Is there sufficient data to resolve the trends in ophiolite depletion? There are generally fewer data back in time, and not many data points between 300 Ma and 200 Ma.

Although data density decreases over time, we added a new statistical test to account for sampling bias.

Lines 134-137 in the RM now read: "*Furthermore, the means of each 10 Ma bin for the plume-derived samples older than 300 Ma are significantly different at the 95% level from the means of the same sample size drawn from the present-day ocean plume Nd isotopic dataset (see Methods, Supplementary Fig. 8), ruling out the trend being due to sampling density bias.*"

We also added an in-depth explanation of the approach to the Methods section, where lines 319-329 read: "*To account for sampling bias, we compared the means of each of the 10 Ma bins of the plume dataset to the means of one thousand re-sampling of the same sample size drawn from the Nd isotopic dataset of present-day plume-derived rocks in order to test the statistical significance of the limited sample sizes per age bin. For example, if 15 analyses were used to determine the mean at a given age bin, the means of 1000 sets of 15 analyses drawn from the present-day dataset were used to determine the confidence intervals at the 1σ (68%) and 2σ (95%) levels. Although for some samples older than 300*

Ma, the mean is not distinguishable from the present-day distribution at the 95% confidence level, the mean Δ_{DM} value for rocks older than 300 Ma is significantly different at the 95% confidence level from the present-day distribution, while for rocks younger than 300 Ma the means cannot be statistically distinguished (Supplementary Fig 9)".

The caption for Supplementary Fig. 8 in the Supplementary Material reads: "*Supplementary Figure 9.* **Comparison of the mean for each age bin with means from the equivalent number of samples drawn from the present-day ocean Nd isotopic dataset.** Plume-related samples older than 300 Ma are generally more depleted than younger samples, with their mean Δ_{DM} significantly different from that the present-day oceanic plume-related rocks. Δ_{DM} values pre- and post-300 Ma do not show such a clear trend. Grey shaded bands represent the bootstrapped 1σ (darker) and 2σ (lighter) confidence intervals for the mean at each age bin."

We believe that this approach adds even more confidence to the reported findings, because it mitigates the issue of sampling bias.

Comment 4: Are the results only compatible with the geodynamic scenario of Li et al. (2019)? Is there a reason not to consider alternative tectonic scenarios, such as the ones mentioned in Merdith et al. (2021, https://doi.org/10.1016/j.earscirev.2020.103477) or Müller et al. (2022, https://doi.org/10.5194/se-13-1127-2022)? Taking a step back from the uncertain details of global tectonic scenarios, could the new analysis be used to test ideas/debates about the closure of supercontinent Rodinia by extroversion or introversion?

This is a very good point, thank you! Besides Li et al. (2019), the results are compatible with other full-plate tectonic reconstructions published so far (Merdith et al., 2021; Li et al. 2023). Although the shapes of oceanic basins might vary between reconstruction models, their relative positions in terms of continental break-up and amalgamation remain similar. For example: in all models the Paleo-Pacific Ocean opens by break-up of Rodinia between the present-day eastern Australian and western North American margins (with or without South China and other continental blocks in-between), which eventually evolved into the Panthalassa superocean and now the Pacific Ocean; on the other hand, Pangaea assembled through the closure of the previous superocean, i.e., an extroversion assembly of Pangaea. Therefore, the post-900 Ma external and internal oceans are consistent between these tectonic reconstructions. In the revised manuscript, we added a note about this point (lines 223-231).

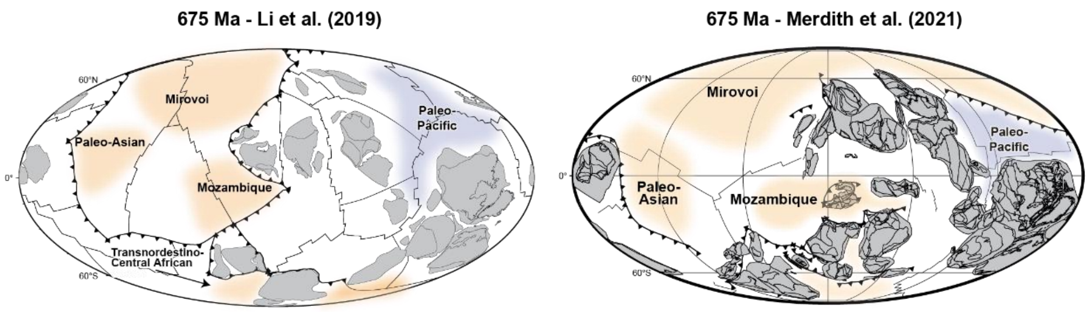

Figure 1: Comparison between Li et al. (2019) and Merdith et al. (2021) models showing that the relative positions of oceanic basins are equal in both models.

Lines 223 – 231 of the RM now read: "*We note that our analysis based on the reconstruction model of reference[13] remains valid for other recent global reconstruction models[16,17]. Although the shapes of oceanic basins might vary between reconstruction models, their relative positions in terms of continental break-up and amalgamation remain similar. For example, in all these models the Paleo-Pacific Ocean opened by the break-up of Rodinia between the present-day eastern Australian and western North American margins (with or without South China and other continental blocks in-between), which eventually evolved into the Panthalassa superocean and now the Pacific Ocean. On the other hand, in all these models Pangaea assembled through the closure of the previous superocean, i.e., an extroversion assembly of Pangaea.*"

Regarding the debates about the amalgamation of Rodinia, while we believe that the approach would be valid to attempt to resolve this issue, the sample density that far back in time is yet too low, as noted before, and thus is likely not enough to provide definitive answer regarding the extroversion *vs.* introversion debate. However, such an examination might be possible in future following the same approach that we take in this study, once an increased Precambrian data coverage deemed such an analyses feasible.

Comment 5: Finally, why is the enrichment only observed in the last 400 Myr? Was continental crust not recycled in the mantle before about 650 Ma? This is alluded to in the concluding statement without explanation: 'This would imply that geochemical anomalies in the mantle might be a transient feature in the Earth's mantle, reflective of the planet's geodynamic evolution instead of static features since earth Earth.' (L. 202-204).

Continental crust material was likely also recycled back to the mantle before 650 Ma, but probably mostly into the Rodinia (present-day Pacific) mantle domain (Supplementary Fig. 1). Such an enrichment signal is thus not expected to be seen in the African domain, as shown by our observation (Fig. 2).

In our model (Fig. 3 and Supplementary Fig. 1), the African mantle domain was an external superocean before ~700 Ma, and was therefore relatively depleted of continental materials. During the extroversion assembly of Gondwana and eventually Pangea after 700 Ma (Fig. 3 and Supplementary Fig. 1), continental materials started to be carried down to the deep mantle along subduction zones, which took 200–300 Myr to be cycled back after 400 Ma to the surface in plume-related basalts.

We made a few changes in the RM to make this point clearer. Lines 161 – 169 in the RM read:

"*…This latter model explains the generally ancient isotopic signature commonly attributed to recycled materials[7], as evidenced mostly by Pb isotopes[1] and mass-independent fractionated signatures in S isotopes[28], although younger recycled crust materials of 650–200 Ma have also been proposed for some enriched plume sources based on Sr and Os isotopes[29,30]. However, the contribution of wholesale continental crust in total crustal recycling was likely relatively minor compared to subduction erosion and sedimentary subduction[27] because of the high buoyancy of continental crust. Nevertheless, all of these processes happen in convergent margins concentrated in the African domain over the last supercontinent cycle[13,16].*"

<u>Reviewer #3:</u>

This manuscript presents a comprehensive analysis of the isotopic evolution of the Earth's mantle, focusing particularly on the African Mantle Domain (AMD) and its relationship with supercontinent-superocean evolution since 700 million years ago (Ma). The authors analyse the geochemical composition of mid-ocean ridge basalts and plume-induced basalts, both from present-day oceans and preserved in ancient ophiolites. By comparing the isotopic signatures of these rocks across time, the researchers aim to reconstruct the history of mantle enrichment processes. The plume-derived rocks in the African Mantle Domain show a trend of increasing enrichment from 400 Ma to 100 Ma, followed by a depletion trend. This enrichment is attributed to the subduction of continental crust during the assembly of Gondwana and Pangea. The authors propose a model where the Rodinia-to-Pangea supercontinent cycle played a crucial role in creating the isotopic dichotomy between the African Mantle Domain and the Pacific Domain. Overall, this paper provides a valuable contribution to our understanding of mantle evolution and the formation of isotopic heterogeneity. The proposed model is convincing and well-supported by the data. It is well-structured, thoroughly researched, and effectively communicates complex scientific concepts to the reader. The incorporation of multiple lines of evidence, including isotopic data, geological reconstructions, and regression analyses, adds credibility to the study's findings. Overall, the paper makes a significant contribution to the field of mantle geochemistry and our understanding of Earth's dynamic evolution. However, I still have some minor concerns on this manuscript which needs to be addressed in detail before it is suitable for publication.

Thank you for your thoughtful comments. We are excited to see the positive reception of the ideas on the manuscript.

Comment 1: They authors acknowledges the restricted window into the past provided by present-day ocean floor basalts (limited to the last 200 Ma) but doesn't delve into alternative methods for studying the deeper mantle, such as Sr, Pb isotopes which are crucial in understanding of the mantle isotopic heterogeneity.

We assume this comment is in reference to Sr and Pb isotopes in kimberlites and older continental mantle plume-derived rocks (LIPs). We avoided using continental basalts to avoid the issue of contamination by continental crust during the ascent and storage of the magmas, which makes the identification of true EM signatures ambiguous (source vs. contamination). Deep-time analyses of kimberlite isotopic signatures have already been done elsewhere (Woodhead et al. 2019; Giuliani et al. 2020), and, as noted (Lines 191 – 192), the data indicate the appearance of isotopically enriched reservoirs in the same time frame as found by our study.

We added Lines 83 – 86 to the RM, which read: "*We focused on oceanic basalts because continental basalts are likely contaminated by continental crust during ascent and storage through the crust that makes any interpretation based on enriched isotopic signatures difficult.*"

With regards to using the Sr and Pb isotopic systems for oceanic rocks, there is a low availability of data (Lines 80 – 81 of the RM). Furthermore, the Nd isotopic system is more resilient to seawater alteration and metamorphism than the Sr and Pb systems.

For these reasons, we opted to focus our analysis on Nd isotopic systematics of oceanic basalts.

Comment 2: Secondly, while the continental crust subduction hypothesis is well-supported, a more detailed exploration of the mechanisms by which this crustal material is recycled into the mantle would be valuable. The authors could explore additional mechanisms for continental crust recycling into the mantle, besides subduction.

In the OM we listed different ways continental crust can be recycled (Lines 157 – 160; Stern and Scholl. 2010). Please see comment 5 from Reviewer #2 to see how this point was addressed.

Comment 3: While the focus on the African Mantle Domain provides valuable insights, it would be beneficial to discuss the implications of the findings for mantle dynamics on a global scale.

Conducting the same type of statistical analysis for the ophiolites in the Pacific Domain is, at the moment, impossible due to the scarcity of data. Supplementary Fig. 10 shows the Pacific Domain plume-derived ophiolites and present-day plume-derived rocks in comparison to the

African Domain data to demonstrate how enriched isotopic signatures only become prevalent in the Pacific Domain after 10 Ma.

The global implications of our observations and interpretations are stated in numerous passages of the OM. Lines 172 – 179 compare African Domain with Pacific Domain. In Lines 196 – 222 we relate the whole Earth's tectonic evolution since the break-up of Rodinia to our observations and the rise of the Earth's isotopic dichotomy. Implications of our findings for global scale mantle dynamics is once again addressed in Lines 220 – 223.

We added another passage to directly link our observations (and interpretations) to the global picture.

Lines 220 – 222 in the RM now read: "…which concentrated subduction in the African Domain relative to the Pacific Domain, creating the prevalence of enriched isotopic signatures in the African Domain since late Palaeozoic."

Comment 4: The paper could further discuss potential uncertainties in the data and analysis methods and their implications for the interpretation of results. Please provide a more detailed discussion of potential uncertainties in the data, methodology, and interpretations, and how they were addressed or accounted for in the analysis.

As noted in the response to ***comment 3*** from **Reviewer #2**, we added a new analysis and an accompanying supplementary figure that considers potential sampling bias by randomly sampling the same number of samples from a bootstrapped dataset identical to the present-day Nd isotopic distribution. This analysis shows that the samples older than 300 Ma are mostly significantly different from the present-day distribution. This approach helps to overcome one of the main shortfall of the ophiolite dataset.

Other sources of uncertainty include alteration of the primary isotopic signatures and statistical uncertainty. Regarding alteration of isotopic signatures, Lines 286 – 289 elaborate on the high resistance of the Nd system to seawater alteration, and we specify in Line 281 that only samples with no more than amphibolite facies metamorphism were considered, which mitigates metamorphic disturbances of the isotopic system.

For the statistical analysis, all of the parameters used in the Gaussian Process regression, GAM regressions and LOWESS fitting were given in the **Methods** section, as well as the results of statistical tests of the GAM regressions. We believe that the coherent results between the different statistical methods (GP, GAMs, LOWESS) strengthen our interpretations (Supplementary Fig. 6). The regression using only ophiolites also shows an enrichment trend and that the observed enrichment is not an artifact of combining datasets (Supplementary Fig. 7).

Minor changes:

- Added "$\Delta_{8/6}$ is the deviation from the Northern Hemisphere Reference Line in $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ space" to the caption of **Figure 1** to explain the $\Delta_{8/6}$ variable.
- Changed the label on the histogram of **Figure 2** to "frequency" instead of counts.
- Added a thicker outline on the histograms of **Figure 2**.

Reviewer #3:

This manuscript presents a comprehensive analysis of the isotopic evolution of the Earth's mantle, focusing particularly on the African Mantle Domain (AMD) and its relationship with supercontinent-superocean evolution since 700 million years ago (Ma). The authors analyse the geochemical composition of mid-ocean ridge basalts and plume-induced basalts, both from present-day oceans and preserved in ancient ophiolites. By comparing the isotopic signatures of these rocks across time, the researchers aim to reconstruct the history of mantle enrichment processes. The plume-derived rocks in the African Mantle Domain show a trend of increasing enrichment from 400 Ma to 100 Ma, followed by a depletion trend. This enrichment is attributed to the subduction of continental crust during the assembly of Gondwana and Pangea. The authors propose a model where the Rodinia-to-Pangea supercontinent cycle played a crucial role in creating the isotopic dichotomy between the African Mantle Domain and the Pacific Domain. Overall, this paper provides a valuable contribution to our understanding of mantle evolution and the formation of isotopic heterogeneity. The proposed model is convincing and well-supported by the data. It is well structured, thoroughly researched, and effectively communicates complex scientific concepts to the reader. The incorporation of multiple lines of evidence, including isotopic data, geological reconstructions, and regression analyses, adds credibility to the study's findings. Overall, the paper makes a significant contribution to the field of mantle geochemistry and our understanding of Earth's dynamic evolution. However, I still have some minor concerns on this manuscript which needs to be addressed in detail before it is suitable for publication. Thank you for your thoughtful comments.

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Thank you for your clarification on focusing specifically on Nd isotopic systematics in oceanic basalts. The rationale regarding the contamination issues linked with continental basalts and the resilience of the Nd isotopic system to seawater alteration and metamorphism is well-founded. It's reassuring to learn that analyses of kimberlite isotopic signatures in deep time, conducted elsewhere, align with the findings of your study, further bolstering the credibility of your conclusions. Overall, the author's response effectively addresses the comment and provides a clear justification for your methodological approach.

Comment 2: Secondly, while the continental crust subduction hypothesis is well-supported, a more detailed exploration of the mechanisms by which this crustal material is recycled into the mantle would be valuable. The authors could explore additional mechanisms for continental crust recycling into the mantle, besides subduction.

In the OM we listed different ways continental crust can be recycled (Lines 157 – 160; Stern and Scholl. 2010). Please see comment 5 from Reviewer #2 to see how this point was addressed.

Thank you. I'm satisfied with this response.

Comment 3: While the focus on the African Mantle Domain provides valuable insights, it would be beneficial to discuss the implications of the findings for mantle dynamics on a global scale.

Conducting the same type of statistical analysis for the ophiolites in the Pacific Domain is, at the moment, impossible due to the scarcity of data. Supplementary Fig. 10 shows the Pacific Domain plume-derived ophiolites and present-day plume-derived rocks in comparison to the African Domain data to demonstrate how enriched isotopic signatures only become prevalent in the Pacific Domain after 10 Ma. The global implications of our observations and interpretations are stated in numerous passages of the OM. Lines 172 – 179 compare African Domain with Pacific Domain. In Lines196 – 222 we relate the whole Earth's tectonic evolution since the break-up of Rodinia to our observations and the rise of the Earth's isotopic dichotomy. Implications of our findings for global scale mantle dynamics is once again addressed in Lines 220 – 223. We added another passage to directly link our observations (and interpretations) to the global picture. Lines 220 – 222 in the RM now read: "…which concentrated subduction in the African Domain relative to the Pacific Domain, creating the prevalence of enriched isotopic signatures in the African Domain since late Palaeozoic."

Thank you for providing a thorough explanation of the challenges associated with statistical analysis on the Pacific Domain ophiolites due to data scarcity. The clarification on how Supplementary Fig. 10 depicts the comparison between Pacific and African Domain data, highlighting the timing of enriched isotopic signatures in the Pacific Domain, is appreciated. Overall, it appears that the manuscript effectively covers the global implications of our research findings.

Comment 4: The paper could further discuss potential uncertainties in the data and analysis methods and their implications for the interpretation of results. Please provide a more detailed discussion of potential uncertainties in the data, methodology, and interpretations, and how they were addressed or accounted for in the analysis.

As noted in the response to comment 3 from Reviewer #2, we added a new analysis and an accompanying supplementary figure that considers potential sampling bias by randomly sampling the same number of samples from a bootstrapped dataset identical to the presentday Nd isotopic distribution. This analysis shows that the samples older than 300 Ma are mostly significantly different from the present-day distribution. This approach helps to overcome one of the main shortfall of the ophiolite dataset. Other sources of uncertainty include alteration of the primary isotopic signatures and statistical uncertainty. Regarding alteration of isotopic signatures, Lines 286 – 289 elaborate on the high resistance of the Nd system to seawater alteration, and we specify in Line 281 that only samples with no more than amphibolite facies metamorphism were considered, which mitigates metamorphic disturbances of the isotopic system. For the statistical analysis, all of the parameters used in the Gaussian Process regression, GAM regressions and LOWESS fitting were given in the Methods section, as well as the results of statistical tests of the GAM regressions. We believe that the coherent results between the different statistical methods (GP, GAMs, LOWESS) strengthen our interpretations (Supplementary Fig. 6). The regression using only ophiolites also shows an enrichment trend and that the observed enrichment is not an artifact of combining datasets (Supplementary Fig.7).

Thank you for providing additional insights into the new analysis and supplementary figures aimed at addressing potential sampling bias in the dataset. The incorporation of this analysis, which involves sampling from a bootstrapped dataset, significantly strengthens the study and addresses a notable limitation in the ophiolite dataset. Furthermore, the clarification on other sources of uncertainty, such as alteration of isotopic signatures and statistical uncertainty, is valued. Overall, these enhancements and clarifications enhance the robustness and credibility of our study's findings. Thank you for comprehensively addressing these aspects.