

Dense iron oxyhydroxides as possible reservoirs of water under deep mantle conditions

Corresponding Author: Dr Hongsheng Yuan

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

In this study, two hexagonal iron oxyhydroxides synthesised by heating mantle assemblages at high pressure and temperature and at a range of hydrations. The authors argue that the presence of these phases is evidence of a mechanism for water storage in the lower mantle and that these phases may be the same “H-phase” found in similar experiments from previous studies.

This is a well written and compelling study from a team of highly capable and experienced researchers. Overall I found the study to be robust, informative and highly relevant to ongoing research into how our planet’s water cycle has evolved over the past 4.5 billion years since accretion. Whilst the introduction, methods and results are of high quality and generally publishable, I have some concerns surrounding the discussion and conclusions of the study which I would like to see the authors address. Once these points are addressed, I believe this study will be well suited for publication in Nature Geosciences and will be of great benefit and utility to the geosciences community.

An overarching theme of the discussion is that these phases should be common in the lower mantle and should be efficient scavengers of water. Furthermore, the ex-situ analysis of these phases demonstrates that they are stable water bearing phases at lower temperatures and pressures. This, to me, suggests that as the deep Earth has cooled, these phases have only held onto their water content more and more tightly. A significant increase in temperature would be required for these phases to release any water. The authors do appeal to temperature variations from the core being a source of temperature perturbations which could trigger volatile release from these lower mantle phases (lines 300-303). However, thermal variations in the core are expected to be incredibly small (Stevenson 1987) because the convective vigour of the core ensures that it stays well mixed on relatively short timescales. Furthermore, all plausible thermal histories of the core feature a gradual cooling since the moon forming impact, with no prospect of remelting the lower mantle due to core energetics (e.g. Driscoll and Davies 2023). All of this (particularly lines 290-299) leads me to the opinion that if these phases can efficiently trap large quantities of water in the deep mantle, there is no obvious mechanism to release it. Taken to an extreme, what is to stop these phases and the core to sequester all of Earth’s water via mantle circulation, leading to a dry surface environment?

Perhaps I am misunderstanding these parts of the discussion and some gentle rewording and additional references (e.g. for large core thermal perturbations) would address my confusion.

Some minor comments:

Line 76: Can you be more specific about the water content of this starting material? Do you have measurements. If not, please provide some justification or reassurance for the reader.

Figure 1: There is a mix of multiple legends and annotations. Can the legends be combined? Or given some format which makes them more distinct?

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Line 245: Can you comment on why “most studies” do not find these phases?

Line 284: I think a clearer explanation of why this behaviour is paradoxical would be helpful.

Line 288: Please provide some numbers with these comments on density.

Line 310+: I was waiting for some discussion on what happens when these phases come into contact with the liquid outer core. It has been suggested that the core is a significant water reservoir and this should at least be acknowledged in the text because these phases might either provide a conveyor for water to enter the core or a means of limiting core water content as they will have a different partition coefficient with liquid Fe compared to other lower mantle phases.

Reviewer #2

(Remarks to the Author)

General Comments:

This paper uses in situ and ex situ measurements of two synthesized iron oxyhydroxides, Fe₅O₁₂H_x and Fe₇O₁₂H_x, at high pressures and temperatures, identifying them as potential mineralogical hosts for hosting hydrogen in a primordial basal magma ocean. Additionally, one of these two phases may correspond to the previously identified H-phase of FeOOH, with important mineralogical consequences for the stability of known oxyhydroxides in the deep Earth. This would provide a significant new mechanism for hosting water in the deep Earth, a topic of broad significance to the wider geological community.

The primary observation of the two previously unreported hydrous phases seems both robust and compelling. Similarly, most of the writing is clear and straightforward. However, the second half of the manuscript would benefit from a minor restructuring that creates a more sequential buildup to the conclusion that both phases are chemically and gravitationally stable at BMO conditions. In particular, the discussion of unit cell volume, hydration, and density was markedly less clear than the rest of the manuscript. Replacing Figure 4 with Extended Data Figure 5, expanding on the discussion of the hydrogen content calculation, and building this argument up more carefully would help readers make the connection to both hydration and density. Otherwise, density seems to disappear from the conversation between the introduction and its reappearance in line 298. Since there is data for both of the new phases at 80 GPa, a comparison of the density of these phases to other potential hosts of water in the deep Earth (like pyrite-type FeO₂H) would be useful in the absence of an EoS.

Lastly, the introduction is compelling in its advocacy for identifying phases that could anchor water in the deep Earth. Unfortunately, this introduction overlooks the stabilizing effect of Al substitution on FeO₂H and other hydrous phases. This needs to be addressed in both the text and Fig. 1, as intermediate compositions of (Al,Fe)OOH are known to be stable at higher P-T conditions than the Fe-endmember. This weakness was underlined by Al-bearing DHMS being suddenly introduced into the broader picture in line 295.

Recommendations:

Add DOI's to all references

Error bars are only present in Figure 1 and are not defined in the corresponding figure legend.

Only abbreviate bridgmanite where needed (figures), not throughout the entire manuscript. Other minerals with longer names (e.g., ferropericlasite) are written out.

Figure 1:

L86 purple empty hexagons, not "diamonds"

L89 add 'bottom left' to description of black dashed line for clarity.

L89 "c-FeO₂H_x" is not a widely used abbreviation, therefore describe as cubic or pyrite-type.

Figure 2: Replace either blue or purple with a different color as these are too similar to clearly see. Address in text or caption how overlapping peaks (i.e., bridgmanite and Fe₅O₁₂H) were treated during peak fitting and when obtaining lattice parameters

L132 The sentence beginning "While the use of..." was not clearly written.

L152 The term "quench product" is unclear as you just used the term quench to mean thermal quenching but I believe here you mean the depressurized version of the high-pressure phase.

L174 The "varying shades of blue" look really green on my screen.

L200 This paragraph on hydrogen incorporation just isn't as clearly written as the rest of the manuscript. Swapping the figure as previously mentioned would help, but this section needs work.

L226 I am unconvinced these phases are "hyper-hydrous" in the conventional use of the term. They don't only exist in environments with large amounts of water, as stated earlier in this manuscript (L134-5). It seems like instead the authors want to emphasize that this would be the primary water host in any assemblage we could reasonably expect it to be in, but this term does not convey that. Similarly, the term "universal water-accepter" is cumbersome and unclear.

Reviewer #3

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Geoscience initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Version 1:

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Reviewer #1

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I thank the authors for their careful and thorough response to my comments. I think that the changes to the manuscript do enhance its utility and intrigue. I believe the study is now of suitable quality to be published in Nature Geosciences.

Whilst there are still things which puzzle me slightly (e.g. the whole question of mantle entrainment) I think that addressing them is far beyond the scope of this study and perhaps this is just a hallmark of a timely and provocative set of results. I feel that the manuscript should now be published such that the broader community can interrogate and assimilate the work. I look forward to seeing where this goes!

I would add that this has been a pleasant experience both in reading about some excellent work and in dealing with a courteous and thoughtful group of authors, so thank you.

Reviewer #2

(Remarks to the Author)

The authors have done a good job of incorporating the feedback provided during the first round of review. In particular, the discussion that was rewritten beginning on line 215 is vastly improved. There are, however, a few very minor issues which could be cleaned up during the typesetting & proofing stage:

- 1) In a few places, the authors use both tildes and numerical ranges to indicate uncertainty (e.g., L158 and L177). If previously reported values fall into a range, it seems sufficient to state 9-14 rather than ~9 to ~14. If the authors are still concerned about understating the uncertainty, I would recommend stating 'approximately 9-14' as it is much easier to read.
- 2) Most of the spelling is in American English except the spelling of 'coloured'.
- 3) There are faint grey lines around part b of Figure 2, which could probably be cleaned up.

This tiny tweaks aside, this is a much-improved manuscript and a valuable contribution to the geoscience community.

Reviewer #3

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Reviewer #1 (Remarks to the Author):

In this study, two hexagonal iron oxyhydroxides synthesised by heating mantle assemblages at high pressure and temperature and at a range of hydrations. The authors argue that the presence of these phases is evidence of a mechanism for water storage in the lower mantle and that these phases may be the same “H-phase” found in similar experiments from previous studies.

This is a well written and compelling study from a team of highly capable and experienced researchers. Overall I found the study to be robust, informative and highly relevant to ongoing research into how our planet’s water cycle has evolved over the past 4.5 billion years since accretion. Whilst the introduction, methods and results are of high quality and generally publishable, I have some concerns surrounding the discussion and conclusions of the study which I would like to see the authors address. Once these points are addressed, I believe this study will be well suited for publication in Nature Geosciences and will be of great benefit and utility to the geosciences community.

Response: We sincerely thank the reviewer for the positive assessment of our work and for the encouraging comments regarding the robustness and relevance of our study. We are particularly gratified that the reviewer recognized the potential significance of identifying the long-standing "H-phase" mystery, which we have now further emphasized in the revised manuscript. We have carefully addressed the concerns regarding the discussion and conclusions, specifically concerning the geodynamic mechanisms of water release and the thermal constraints of the core. We believe these revisions have significantly strengthened the paper's impact on the understanding of the Earth’s deep-water cycle.

An overarching theme of the discussion is that these phases should be common in the lower mantle and should be efficient scavengers of water. Furthermore, the ex-situ analysis of these phases demonstrates that they are stable water bearing phases at lower temperatures and pressures. This, to me, suggests that as the deep Earth has cooled, these phases have only held onto their water content more and more tightly. A significant increase in temperature would be required for these phases to release any water. The authors do appeal to temperature variations from the core being a source of temperature perturbations which could trigger volatile release from these lower mantle phases (lines 300-303). However, thermal variations in the core are expected to be incredibly small (Stevenson 1987) because the convective vigour of the core ensures that it stays well mixed on relatively short timescales. Furthermore, all plausible thermal histories of the core feature a gradual cooling since the moon forming impact, with no prospect of remelting the lower mantle due to core energetics (e.g. Driscoll and Davies 2023). All of this (particularly lines 290-299) leads me to the opinion that if these phases can efficiently trap large quantities of water in the deep mantle, there is no obvious mechanism to release it. Taken to an extreme, what is to stop these phases and

the core to sequester all of Earth's water via mantle circulation, leading to a dry surface environment?

Perhaps I am misunderstanding these parts of the discussion and some gentle rewording and additional references (e.g. for large core thermal perturbations) would address my confusion.

Response: We sincerely thank the reviewer for this profound and insightful observation. We agree that our previous reliance on core-side thermal perturbations as the primary trigger for water release was geodynamically insufficient, especially given the thermal constraints and cooling history of the core (e.g., Stevenson, 1987; Driscoll and Davies, 2023).

To address this concern and clearly illustrate the mechanism for water release that prevents an irreversible total sequestration scenario, we have thoroughly reoriented and revised the section titled "**Global water cycling and hydration of the Earth's interior**". Instead of a thermal trigger at the core–mantle boundary (CMB), we now present a dynamic, convection-driven release framework based on our new experimental constraints. The key revisions are detailed below:

1. **Convective entrainment via mantle plumes:** While these dense iron oxyhydroxides serve as efficient reservoirs for long-term volatile storage within ULVZs, they are not permanently isolated from global mantle circulation. Because the margins of LLSVPs are considered the primary sites of plume generation, portions of these basally segregated hydrous assemblages can be mechanically entrained into buoyant mantle upwellings and transported toward shallower depths.
2. **Decompression-driven destabilization:** Crucially, our experimental observations demonstrate that these hexagonal phases do not form at pressures below approximately 70 GPa, defining a clear lower pressure limit for their structural stability. Consequently, as the entrained material ascends within a plume, the decreasing pressure shifts local phase equilibria and destabilizes the hydrogen-rich crystal frameworks. This structural breakdown drives the redistribution of water into other coexisting mantle minerals.
3. **Dehydration melting and surface return:** If the local water storage capacity of the surrounding solid assemblage is exceeded during this redistribution, dehydration melting will occur under shallower mantle conditions. This process provides a geodynamically viable pathway for water to escape the deep mantle, return to the convecting mantle, and ultimately reach the Earth's surface over geological timescales.
4. **Dynamic volatile equilibrium:** This revised framework directly clarifies that these hexagonal phases operate as a dynamic, long-term deep reservoir rather than a permanent, dead-end sink for Earth's water budget. Such a continuous convective cycle successfully helps reconcile the long-term preservation of a deep primordial water reservoir with the distinct primordial isotopic signatures

observed in ocean island basalts.

We believe this updated, physically motivated model aligns perfectly with modern geodynamic constraints while highlighting the crucial role of our discovered phases in modulating planetary volatile cycles.

Some minor comments:

Line 76: Can you be more specific about the water content of this starting material? Do you have measurements. If not, please provide some justification or reassurance for the reader.

Response: We thank the reviewer for raising this point. Although we did not perform direct measurements on the specific microscopic sample loadings, well-established literature indicates that mineral powders not pre-dried in a vacuum oven consistently retain adsorbed atmospheric moisture at the thousand ppm level [e.g., American Mineralogist (2015), 100 2336–2339; Chemistry of Materials (2007), 19, 825–833]. We have revised the text (Line 79) to explicitly state this estimated value.

Line 87: The “H-phase” hasn’t been mentioned yet. I think it is very interesting that you might have identified a mysterious phase from past studies and this should feature in the introduction.

Response: We agree that introducing the “H-phase” earlier enhances the narrative. We have added a dedicated explanation in the Introduction to contextualize the debate regarding the stability of iron-bearing bridgmanite and the unidentified “H-phase”.

Figure 1: There is a mix of multiple legends and annotations. Can the legends be combined? Or given some format which makes them more distinct?

Line 88: “in blue” are you referring to the arrow? Be more specific with these descriptions, although, this caption is rather long.

Response: We have significantly revised Figure 1 and its caption to improve clarity and reduce clutter. Following the reviewer’s suggestion, data points are now represented by distinct black symbols (circles, triangles, and squares), which are explicitly defined in the revised caption. Additionally, we have clarified that the “blue” refers specifically to the blue arrow denoting subduction slab profiles. The caption has been restructured to be more concise while providing precise spatial guidance (e.g., “bottom left”) for the reader.

Line 127: in what way is this dramatic? I think this statement needs backing-up with the implication that makes it dramatic.

Response: We thank the reviewer for the suggestion. We have removed the subjective word "dramatic" and rephrased the sentence to focus strictly on the experimental facts (Lines 85-91). The reaction is highly significant because the bridgmanite assemblage undergoes an unexpected transformation between 2,500 and 2,800 K to form two previously unobserved hexagonal iron oxyhydroxides. Remarkably, this reaction occurs even with trace adsorbed moisture (e.g., <0.1 wt% for Run#WH-S1), demonstrating that very low hydrogen concentrations are sufficient to destabilize lower-mantle silicates and stabilize these new hexagonal phases.

Line 245: Can you comment on why “most studies” do not find these phases?

Response: The primary reason is that most studies investigating bridgmanite stability employ rigorous pre-drying protocols (e.g., vacuum heating) to achieve strictly anhydrous conditions. Our results demonstrate that these iron oxyhydroxides are hydrogen-stabilized phases; consequently, they do not form in truly dry systems. This suggests that past sightings of the “H-phase” in supposedly dry experiments were likely caused by accidental hydrogen contamination. We have added a sentence (Lines 175–180) to clarify that these phases can only form if at least a small amount of hydrogen is present.

Line 284: I think a clearer explanation of why this behaviour is paradoxical would be helpful.

Response: Thank you for this insightful comment. Upon revision, we realized that the term "paradoxical" was indeed potentially confusing as the underlying mechanisms are physically consistent. We have therefore removed the "paradoxical" framing. Instead, we now provide a more direct and transparent explanation of the two key factors:

1. How these phases maintain high density ($\sim 6\text{--}7\text{ g/cm}^3$) despite their high hydrogen content due to their iron-rich frameworks.
2. How the scavenging of water from the melt into these solid phases accelerates (rather than delays) the solidification of the basal magma ocean (BMO) by elevating the residual melt's solidus.

We believe this direct causal approach is clearer than the previous "paradox" phrasing.

Line 288: Please provide some numbers with these comments on density.

Response: To explicitly demonstrate gravitational stability, we added a new panel (Fig. 4b) comparing the densities of $\text{Fe}_5\text{O}_{12}\text{H}_x$ and $\text{Fe}_7\text{O}_{12}\text{H}_x$ with the PREM profile and cubic FeO_2H_x . We have also updated the text (Lines 181-190) to include specific calculated density values based on our experimental volumes. For instance, at 82 GPa, the density of $\text{Fe}_7\text{O}_{12}\text{H}_x$ is 6.98 g/cm^3 , and $\text{Fe}_5\text{O}_{12}\text{H}_x$ is 6.08 g/cm^3 . Both phases are substantially denser than the ambient bulk silicate mantle (PREM) at corresponding depths. This

significant density contrast strongly supports their tendency to segregate and settle to the base of the mantle.

Line 310+: I was waiting for some discussion on what happens when these phases come into contact with the liquid outer core. It has been suggested that the core is a significant water reservoir and this should at least be acknowledged in the text because these phases might either provide a conveyor for water to enter the core or a means of limiting core water content as they will have a different partition coefficient with liquid Fe compared to other lower mantle phases.

Response: We thank the reviewer for this insightful suggestion. We agree that the interaction between these dense oxyhydroxides and the liquid outer core is a critical aspect of Earth's volatile cycle. In the revised manuscript (Lines 235-243), we have acknowledged that this partitioning is highly sensitive to factors such as oxygen fugacity, for which experimental constraints are currently insufficient to determine the direction or magnitude of H-transfer. Furthermore, we have added a discussion on the kinetic limitations at the solid-liquid interface, noting that any reaction or exchange would likely be diffusion-controlled and potentially very slow. This balanced view clarifies that while these phases could theoretically act as a "hydrogen conveyor," the actual net flux remains an open question for future experimental studies.

Reviewer #2 (Remarks to the Author):

General Comments:

This paper uses in situ and ex situ measurements of two synthesized iron oxyhydroxides, $\text{Fe}_5\text{O}_{12}\text{H}_x$ and $\text{Fe}_7\text{O}_{12}\text{H}_x$, at high pressures and temperatures, identifying them as potential mineralogical hosts for hosting hydrogen in a primordial basal magma ocean. Additionally, one of these two phases may correspond to the previously identified H-phase of FeOOH , with important mineralogical consequences for the stability of known oxyhydroxides in the deep Earth. This would provide a significant new mechanism for hosting water in the deep Earth, a topic of broad significance to the wider geological community.

Response: We are grateful for the reviewer's positive assessment of the significance of our work. We are pleased that the reviewer recognized the importance of identifying $\text{Fe}_5\text{O}_{12}\text{H}_x$ and $\text{Fe}_7\text{O}_{12}\text{H}_x$ as primary mineralogical hosts for hydrogen in the deep Earth and their potential identity as the long-sought "H-phase". We believe these findings indeed offer a compelling new mechanism for long-term water storage in the lowermost mantle.

The primary observation of the two previously unreported hydrous phases seems both robust and compelling. Similarly, most of the writing is clear and straightforward. However, the second half of the manuscript would benefit from a minor restructuring that creates a more sequential buildup to the conclusion that both phases are chemically and gravitationally stable at BMO conditions. In particular, the discussion of unit cell volume, hydration, and density was markedly less clear than the rest of the manuscript. Replacing Figure 4 with Extended Data Figure 5, expanding on the discussion of the hydrogen content calculation, and building this argument up more carefully would help readers make the connection to both hydration and density. Otherwise, density seems to disappear from the conversation between the introduction and its reappearance in line 298. Since there is data for both of the new phases at 80 GPa, a comparison of the density of these phases to other potential hosts of water in the deep Earth (like pyrite-type FeO_2H) would be useful in the absence of an EoS.

Response: We thank the reviewer for the encouraging feedback regarding the robustness of our experimental observations and the clarity of the manuscript. We have thoroughly revised the text (Section "Water sequestration and gravitational settling at the CMB") to explicitly define the relationships between unit-cell volume, hydrogen incorporation, and the resulting mineral density. Following your suggestion, we have replaced the original Figure 4 with Extended Data Figure 5. This new Figure 4 now provides a direct visual constraint on the hydrogen content (x) by showing hydration isopleths alongside our experimental data.

To ensure density remains a central theme in the stability discussion, we have added a new panel designated as Figure 4b. This panel compares the calculated densities of both hexagonal phases against the with the PREM profile and cubic FeO_2H_x . We have also

updated the text (Lines 181-190) to include specific calculated density values based on our experimental volumes. At a pressure of 82 GPa, the calculated density of $\text{Fe}_7\text{O}_{12}\text{H}_x$ reaches approximately 6.98 g/cm^3 , while that of $\text{Fe}_5\text{O}_{12}\text{H}_x$ is 6.06 g/cm^3 . This direct comparison clearly demonstrates that both hexagonal phases remain substantially denser than the surrounding bulk silicate mantle, supporting their capacity to segregate gravitationally and serve as primary hydrogen hosts within ULVZs.

Lastly, the introduction is compelling in its advocacy for identifying phases that could anchor water in the deep Earth. Unfortunately, this introduction overlooks the stabilizing effect of Al substitution on FeO_2H and other hydrous phases. This needs to be addressed in both the text and Fig. 1, as intermediate compositions of $(\text{Al,Fe})\text{OOH}$ are known to be stable at higher P-T conditions than the Fe-endmember. This weakness was underlined by Al-bearing DHMS being suddenly introduced into the broader picture in line 295.

Response: We sincerely thank the reviewer for this insightful comment. We agree that Al substitution significantly enhances thermal stability. We have revised the Introduction to acknowledge the stabilizing role of Al and updated Figure 1 to include the previously reported P-T stability data for cubic $(\text{Fe,Al})\text{O}_2\text{H}_x$ (empty squares). This addition provides a more balanced context for the stability of hydrous phases at the CMB. We believe these revisions satisfy the reviewer's concerns and significantly strengthen the geodynamical context of our work.

Recommendations:

Add DOI's to all references

Response: DOIs have been added to all references.

Error bars are only present in Figure 1 and are not defined in the corresponding figure legend.

Response: We thank the reviewer for the suggestion. The captions for Figures 1 and 4 have been updated to explicitly define all error bars:

Fig. 1: Error bars represent $\pm 2\sigma$ of the mean.

Fig. 4a: Error bars for pressures and volumes are smaller than the symbol size.

Fig. 4b: Vertical error bars reflect the combined uncertainty of both the Fe/O ratio and hydrogen occupancy.

Only abbreviate bridgmanite where needed (figures), not throughout the entire manuscript. Other minerals with longer names (e.g., ferropericlase) are written out.

Response: Mineral names are now written in full throughout the main text; abbreviations (e.g., "Bdm") are restricted to figures and tables.

Figure 1:

L86 purple empty hexagons, not "diamonds"

L89 add 'bottom left' to description of black dashed line for clarity.

L89 "c-FeO₂H_x" is not a widely used abbreviation, therefore describe as cubic or pyrite-type.

Response: We thank the reviewer for these corrections. We have revised Figure 1 and its caption accordingly. To reduce clutter, we removed text annotations and used distinct symbols (e.g., empty triangles for the "H-phase"). We added "bottom left" to clarify the location of the phase D to H transition. We have replaced the abbreviation "c-FeO₂H_x" with "cubic FeO₂H_x" throughout the caption and the main text.

Figure 2: Replace either blue or purple with a different color as these are too similar to clearly see. Address in text or caption how overlapping peaks (i.e., bridgmanite and Fe₅O₁₂H) were treated during peak fitting and when obtaining lattice parameters

Response: We thank the reviewer for the suggestion. In the revised Fig. 2b, we have used contrasting colors to provide a clear visual distinction between phases. As clarified in the updated caption, initial hexagonal parameters were estimated from distinct, non-overlapping reflections—(100), (110), and (101)—while overlapping peaks (e.g., between Bdm and Fe₅O₁₂H_x) were resolved using multi-peak profile fitting during full-profile refinement.

L132 The sentence beginning "While the use of..." was not clearly written.

Response: We have revised to clearly distinguish between the intentional use of hydrous silica and the occurrence of the transformation in nominally anhydrous runs (Lines 89-91).

L152 The term "quench product" is unclear as you just used the term quench to mean thermal quenching but I believe here you mean the depressurized version of the high-pressure phase.

Response: We have replaced "quench product" with "recovered product" to specify that the phase was characterized at ambient conditions following decompression.

L174 The "varying shades of blue" look really green on my screen.

Response: We have adjusted the color palette to enhance contrast and ensure that the blue regions are accurately represented and clearly distinguishable across different displays.

L200 This paragraph on hydrogen incorporation just isn't as clearly written as the rest of the manuscript. Swapping the figure as previously mentioned would help, but this section needs work.

Response: Following the reviewer's suggestion, we have significantly rewritten this section to better articulate the relationship between iron deficiency and hydrogen capacity (Lines 146-163). We have completely restructured Fig.4a now incorporates the thermodynamic volume additivity model (formerly Extended Data Fig. 5), allowing for a direct visual comparison between the experimental P–V data and the calculated hydration isopleths. This makes the derivation of hydrogen concentrations ($x \geq 9$ for $\text{Fe}_5\text{O}_{12}\text{H}_x$ and $x \geq 3$ for $\text{Fe}_7\text{O}_{12}\text{H}_x$.) much more transparent. Furthermore, we added Panel (b) to explicitly address density and BMO stability, which we detail in the revised text (Lines 181-190).

L226 I am unconvinced these phases are “hyper-hydrous” in the conventional use of the term. They don't only exist in environments with large amounts of water, as stated earlier in this manuscript (L134-5). It seems like instead the authors want to emphasize that this would be the primary water host in any assemblage we could reasonably expect it to be in, but this term does not convey that. Similarly, the term “universal water-accepter” is cumbersome and unclear.

Response: As suggested, we have replaced these terms with "primary hydrogen hosts" to accurately reflect their role without implying a water-saturated environment.

Reviewer #1 (Remarks to the Author):

I thank the authors for their careful and thorough response to my comments. I think that the changes to the manuscript do enhance its utility and intrigue. I believe the study is now of suitable quality to be published in Nature Geosciences.

Whilst there are still things which puzzle me slightly (e.g. the whole question of mantle entrainment) I think that addressing them is far beyond the scope of this study and perhaps this is just a hallmark of a timely and provocative set of results. I feel that the manuscript should now be published such that the broader community can interrogate and assimilate the work. I look forward to seeing where this goes!

I would add that this has been a pleasant experience both in reading about some excellent work and in dealing with a courteous and thoughtful group of authors, so thank you.

Response: We sincerely thank the reviewer for the strong endorsement and kind words. We are grateful for your constructive feedback, which has significantly enhanced the clarity and impact of our manuscript.

We agree that lingering questions, such as mantle entrainment, present exciting challenges that extend beyond the current scope. We hope our findings will serve as a timely catalyst for the broader community to explore these puzzles in future studies.

It has been an equally pleasant and rewarding experience for us to engage with such a thoughtful and rigorous reviewer. Thank you again for your time and invaluable contributions.

Reviewer #2 (Remarks to the Author):

The authors have done a good job of incorporating the feedback provided during the first round of review. In particular, the discussion that was rewritten beginning on line 215 is vastly improved. There are, however, a few very minor issues which could be cleaned up during the typesetting & proofing stage:

Response: We sincerely thank the reviewer for the highly positive evaluation and for recognizing the improvements made to the discussion section. We also greatly appreciate your careful reading and the final polishing suggestions. We have fully addressed these minor issues in the final manuscript.

1) In a few places, the authors use both tildes and numerical ranges to indicate uncertainty (e.g., L158 and L177). If previously reported values fall into a range, it seems sufficient to state 9-14 rather than ~9 to ~14. If the authors are still concerned about understating the uncertainty, I would recommend stating 'approximately 9-14' as it is much easier to read.

Response: We completely agree that refining these expressions improves readability. We have updated the text throughout the manuscript (including Lines 158 and 177) to use the format "9–14" or "approximately 9–14" as recommended.

2) Most of the spelling is in American English except the spelling of ‘coloured’.

Response: Thank you for catching this inconsistency. We have corrected "coloured" to "colored" and performed a final check to ensure standard American English spelling is used consistently throughout the text.

3) There are faint grey lines around part b of Figure 2, which could probably be cleaned up.

Response: We apologize for this graphical oversight. We have cleaned up the artifacts in the vector file to remove the faint grey lines, and the updated Figure 2 has been provided in our final submission.

This tiny tweaks aside, this is a much-improved manuscript and a valuable contribution to the geoscience community.

Response: Once again, we thank you for your constructive reviews across both rounds, which have undeniably made this a stronger and more valuable contribution to the geoscience community.

Reviewer #3 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Geoscience initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Response: We sincerely thank the co-reviewer for their time and effort in evaluating our manuscript. We deeply appreciate your valuable contribution to this review process and the Nature Geoscience Early Career Researcher initiative.