**Peer Review File**

**Manuscript Title:** A wet heterogeneous mantle creates a habitable world in the Hadean

## Reviewer Comments & Author Rebuttals

**Reviewer Reports on the Initial Version:**

**Referee #1:**

The paper may be made easier to follow.

(1) I agree that CO2 is not very soluble in shallow mafic to ultramafic magmas. So a massive CO2 atmosphere is reasonable and has generally been assumed. But the water concentration is low. MORB forms water blubbles at ~250 bar (2.5 km) so that might be guess for partial water pressure.

(2) Thus, table of present, end Hadean and end magma ocean global CO2 and water reservoirs would help greatly.

(3) I do not see logic behind high Mg magmas. If we totally melt pyrolite mantle we have about 1/3 basaltic component. The basaltic component had to be somewhere during and at the end of the magma ocean. If it is in dense lower mantle phases so state. Cite Geoph Davies. He suggested this was one way to get thin Hadean ocean crust that subducts and into the Archean. One does not get Icealnd crust that is hard to subduct.

(4) The modern ultrafast ridge axis provides some calibration. The hydrothermal flux is then about 40 W/m^2, not greatly lower than runaway greenhouse conditions. There is magma len s and the mush expels melt. This is basaltic magma.

(5) What is equivalent of subduction. The CO2-rich solid cannot get too hot at too shallow dpths. It would also help to discuss equivalent of arc magma more.

Norm Sleep

**Referees #2**

Miyazaki and Korenaga: A new mode of geodynamics in the Hadean facilitates the emergence of early life

Marc Hirschmann

This is an interesting paper that addresses one of the most puzzling aspects of the functioning of the deep Earth carbon cycle and its relationship to tectonic and climactic evolution. We expect magma oceans to vent much of the bulk Earth carbon to the surface, but yet today and apparently from an early time, much of that carbon is in the mantle. The paper suggests that this is owing to rapid recycling of carbon in a more-vigorous hyper-plate tectonic regime in the early Hadean. This idea was suggested first by Sleep and Zahnle (2001), which I was disappointed to see is not cited here. However, Sleep and Zahnle were rather vague about how this rapid recycling may occur, whereas Miyazaki and Korenaga focus on a particular hypothesis, accompanied by calculations, that take the problem several steps further.

This paper is potentially publishable in Nature, as it may provide an answer or a partial answer to the key question of how Earth made the surprisingly rapid transition from the magma ocean stage to the more "normal" world evidenced in Earths' oldest rocks (and zircons). However, at present it is lacking some key explanations and calculations and so I find it unconvincing. Possibly with significant revision, it may reach the intended goal.

There are four principle parts of the presentation that are lacking, and need to be improved.

1. The model presented is predicated on the assumption that plate tectonics begins operating almost immediately following crystallization of the magma ocean. Of course, we don't know this to be so and other geodynamicists have concluded that different tectonic regimes prevailed in these early times. However, in the context of the present model, a particular challenge must be addressed. The approach taken here assumes a parameterized convection model linked to heat flow, and an essential aspect of the model, required to reach the paper's conclusions, is that this early plate tectonic regime operated ~40 times faster than modern rates. This assertion does not give any insight into whether plate tectonics actually works in this regime. The paper argues that under these circumstances, mean subducted oceanic lithosphere age is 5 Ma. But how does this work? What are the negative buoyancy forces driving subduction for 5 Ma lithosphere? And how would plate tectonics operate if those buoyancy forces are not strong? Without some calculation that this could work, the model is not convincing.

2. The focus here is that plate manufacture is rapid, that lithosphere formation is retarded owing to a lack of mantle dehydration, and that the ultramafic character of much of the sea floor forms an efficient CO2 sink. But an essential factor, the survival of CO2 in subducting lithosphere is not addressed. How do we know whether C is actually subducted, rather than stripped from the downgoing plate? It is controversial whether C is subducted into convecting mantle today (e.g., Kelemen and Manning, 2015), but wholly speculative as to whether it would be for a hotter (i.e., younger) plate subducting into a hotter mantle. The faster presumed subduction rates may counteract these problems, and so….maybe. But the onus is on the authors to demonstrate so with a pertinent calculation.

3. The model presented here is launched from a previous paper by Miyazaki and Korenaga arguing that following magma ocean crystallization, the mantle consists of a highly heterogeneous mix of depleted peridotite salted with Fe-enriched domains. Here, they assert that this depleted peridotite has a Mg# of 95 and therefore has a high melting temperature, resulting in little crust production and in inefficient mantle dehydration, resulting in thin lithosphere. Several problems arise:

(a) Whereas the earliest crystallizing products of a magma ocean may have a Mg# of 95, later products would not be similarly extreme. The assumption that large fractions of the mantle would be this end-member composition needs to be justified with some kind of mass balance. After all, the average mantle must remain close to a Mg# of 89. And what are the proportions and compositions of the Fe-rich domains?
(b) There is little reason to believe that the solidus temperature of peridotite can be extrapolated as a simple function of Mg#. Rock with such extreme composition will not likely have pyrolite mineralogy. It seems that the source-model from the earlier paper has no associated mineral information? If so, the application of the simple solidus parameterization given here is speculative.
(c) The model assumes that the highly depleted Mg#95 peridotite remains sufficiently hydrous to have low viscosity because it does not melt enough to lose appreciable H2O. However, the paper acknowledges that such a lithology will melt to a small degree. It asserts without any quantification that the extent of melting would be too small to dehydrate the residue. In its present form, it is more of a speculation than a constraint. It needs a calculation. What melt fractions are expected? What is the applicable partition coefficient (at 1600 °C)? This requires saying something about the mineralogy. If the mineralogy is dominantly olivine, then the effective partition coefficient is a small fraction of that applicable to fertile peridotite and merely 0.1% melting would be sufficient for near-complete dehydration. If the mineralogy is more "normal", then the melt fraction would have to be no more than about 1% for the residue significant H2O.

4. An important feature of the paper, leading to the conclusion of high carbon sequestration and hot spring activity conducive to prebiotic chemistry, is the assumption that the earliest seafloor consisted chiefly of ultramafic peridotite. This, it is said, is owing to the low crust production from highly depleted magma ocean residues. However, it is not made clear how this is reconciled with the requirement that the average mantle must have remained (nearly) the same composition as modern peridotite (actually, a little more fertile, as the continents had not yet been extracted). What is the role of the Fe-enriched domains in crust production? If these domains were widely sampled by "ridges", then they should have produced a great deal of extra crust. Stolper and Asimow (2007) showed that compared to homogeneous mantle, dividing the mantle into depleted and enriched domains produces about the same amount of crust, provided the two approximate thermal equilibrium. Even if they aren't in thermal equilibrium, the amount of crust produced by the enriched domains should be substantial for a mantle potential temperature of 1600 °C.

What suppresses the participation of the Fe-rich domains in melt production? Is it that they are so large and widely dispersed, that they make large amounts of crust but in very restricted portions of the sea-floor? Is it supposed that the more Fe-rich domains that formed from late-stage magma ocean crystallization were initially

hidden in the lower mantle? What is their average length scale at the point of their formation, and how rapidly did they get stirred back in to make "average" mantle? In the modern mantle, convective stirring homogenizes subducted oceanic crust with the rest of the mantle within a few hundred million years (e.g., Kellogg and Turcotte, 1990). If convective stirring during the time of interest was ~40 times more vigorous, then one would expect that quite rapidly the mantle would become a streaky marble cake that produced a lot of crust. Does the model have some mechanism for preventing the Fe-rich domains from participating in melting? And, returning to an earlier point, by mass balance, what fraction of the mantle resides in these Fe-rich domains? Without addressing these questions, the inference of a peridotite-dominated melting regime seems to derive from a selective description of the system.

If a revised manuscript can quantitatively address the questions above and still fit within the confines of a Nature ms., it could be very interesting.

**Author Rebuttals to Initial Comments:**

**Response to Referee #1 (Norm Sleep):**

1. "I agree that $CO_2$ is not very soluble in shallow mafic to ultramafic magmas. So a massive $CO_2$ atmosphere is reasonable and has generally been assumed. But the water concentration is low. MORB forms water bubbles at ~250 bar (2.5 km) so that might be guess for partial water pressure."

   → We agree that MORB forms water bubbles at 250 bar or 2.5 km below the sea level,and this fact is consistent with the solubility model adopted in this study. Equation (2) suggests that water dissolves into silicate magma up to 1.1 wt% at 250 bar. Under the mid-ocean ridges, volatiles including water concentrate to a small amount of melt as themantle partially melts. Considering that MORB includes 100–750 ppm of water and thatMORB produces 1 % of partial melt, water concentration in the partial melt exceeds the saturation level of 1.1 wt%. Water is expected to form bubbles at modern mid-ocean ridges, whereas water content in the magma ocean remains below 1 wt% and thus belowthe saturation limit. Efficient degassing does not take place during the most of the magma ocean stage, but the same degassing mechanism as mid-ocean ridges takes place in the final stage of magma ocean solidification. We clarified this by adding following sentencesin Methods:

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   P. 12

   When the melt-dominated layer disappears, the upper part of the mantle, while be- ing rheologically solid, remains partially molten. When upward melt percolation is faster than solid-state convection, volatiles contained in the partially molten layer are expected to be released to the atmosphere (Figure 1b). As partial melt approaches the surface and solidifies, volatiles concentrate in the residual melt phase. Volatiles eventually exceed the saturation limit, and thus volatiles in the near-surface region would have degassed efficiently.

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2. "Thus, table of present, end Hadean and end magma ocean global $CO_2$ and water reservoirs would help greatly."

   → Thank you for the suggestion. A table summarizing the evolution of volatile budget is now added to the Supplementary Information.

3. “I do not see logic behind high Mg magmas. If we totally melt pyrolite mantle we have about 1/3 basaltic component. The basaltic component had to be somewhere during and at the endof the magma ocean. If it is in dense lower mantle phases so state. Cite Geoph Davies. He suggested this was one way to get thin Hadean ocean crust that subducts and into the Archean. One does not get Icealnd crust that is hard to subduct.”

→ The differentiation mechanism considered here is not by the partial melting of the mantle, but it is rather induced by the segregation of liquidus mineral, which is bridgmanite in the deep mantle conditions, from the remaining magma ocean. We have clarified the mechanism of fractionation in the main text, and the compositions of two components considered in this study, high-Mg# matirx and iron-rich blobs, are now presented in the Supplementary Information.

Because the sedimentation of bridgmanite is driving differentiation, the high-Mg# matrix has higher MgO and $SiO_2$ contents than the pyrolitic mantle, whereas the complementary reservoir, Fe-rich blobs, becomes enriched in FeO and depleted in $SiO_2$. It is noted that such a fractionation mechanism does not take place in the present, and the iron-rich blobs do not resemble the present-day basaltic crust.

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P. 6

We propose that rapid recycling is possible with a chemically heterogeneous mantle, which results from the fractional solidification of a magma ocean (Miyazaki and Ko- renaga, 2019). The differentiation of a magma ocean is driven by the settling and sedimentation of a liquidus mineral, bridgmanite, at the base of the melt-dominated layer. Consequently, high-Mg# (defined as molar Mg/(Mg+Fe)×100) and $SiO_2$-richmaterials would constitute the lower mantle, whereas the upper mantle would be com- prised of residual iron-rich dense materials (Figure 3a). Our scaling analysis suggests that such a gravitationally unstable structure is subject to small-scale Rayleigh-Taylor instabilities with a length scale shorter than 100 km (Miyazaki and Korenaga, 2019). At the end of solidification, iron-rich materials would thus have existed as small blobs embedded in a high-Mg# pyroxenite matrix (Figure 3a)

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Supplementary Information P. 2

The fractional crystallization of a magma ocean produces a structure dominated by the high-Mg# matrix with small iron-rich blobs (Figure 3a). Miyazaki and Korenaga estimated that the ternary composition of the high-Mg# matrix is on average MgO 41.7 wt%, FeO 4.4 wt%, and $SiO_2$ 53.9 wt%, whereas that of iron-rich blobs is MgO 41.0 wt%, FeO 20.6 wt%, and $SiO_2$ 38.3 wt%. The Mg# of the two components are ~94 and ~77, respectively, resulting in contrasting densities and melting tempera- tures. It is noted that iron-rich blobs make up ~25% of the heterogeneous mantle, and the average composition of the mantle is consistent with the pyrolitic mantle.

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4. "The modern ultrafast ridge axis provides some calibration. The hydrothermal flux is then about 40 W/m$^2$, not greatly lower than runaway greenhouse conditions. There is magma len sand the mush expels melt. This is basaltic magma."

   → We agree that the hydrothermal flux would be higher that the present day, but the globalheat flux is substantially lower than the heat flux by hydrothermal circulation. Also, the solar luminosity in the Hadean was likely ~30% lower than the present, and thus the geothermal heat flux needs to be larger than 100 W/m$^2$ to reach the runaway greenhouse limit.

5. "What is equivalent of subduction. The $CO_2$-rich solid cannot get too hot at too shallow depths. It would also help to discuss equivalent of arc magma more."

   → We agree that some fraction of carbonates would decompose during the subduction, andwe now discuss this issue in the following sentences:

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P. 20–21 (Methods)

Some amount of carbonates created by hydrothermal alteration may decompose and return to the hydrosphere during subduction. The main source of loss is expected to be dissolution of carboantes to serpentine-driven fluid. The estimate of carbon being removed from theplate remains controversial for the present (Dasgupta and Hirschmann, 2010; Kelemen and Manning, 2015), but the Hadean plates are expected to contain ~6 times more carbonates than the present-day oceanic crust, and thus it is unlikely for all carbonates to be dissolved. When the top 500 m of oceanic crust contains ~10 wt% of water, the maximum amount of$CO_2$ that could dissolve into serpentinite-driven fluid is $4.4\times10^{17}$ kg, which is less than 1%of total carbonates in oceanic crust. Here, we assume that serpentinize releases water at ~3 GPa and the solubility of carbonates to fluid is $10^4$ ppm (Kelemen and Manning, 2015). Moreover, some fraction of the extracted carbon would be stored in mantle lithosphere and continental crust (Kelemen and Manning, 2015), the latter of which would have been created by the melting of hydrated subducted materials. Therefore, not all recycled carbon would return to the hydrosphere. Even if we assume that 50% of the subducted carbon returns to the hydrosphere, the sequestration of carbon would still be completed within ~260 Myr. It is noted that decarbonation reactions may not be efficient under a hotter mantle as well. Carbonates releases $CO_2$ at a hotter temperature, but even under a hotter subduction geotherm, the top of oceanic crust would be cold enough to stabilize carbonates (Figure S8). The subduction geotherm, however, would depend on various factors including the dip angle, and phase equilibria under different mineral and melt assemblage would also be necessary for more detailed discussion.

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**Response to Referee #2 (Marc Hirschmann):**

1. "The model presented is predicated on the assumption that plate tectonics begins operating almost immediately following crystallization of the magma ocean. Of course, we don't know this to be so and other geodynamicists have concluded that different tectonic regimes prevailedin these

early times. However, in the context of the present model, a particular challenge must be addressed. The approach taken here assumes a parameterized convection model linked to heat flow, and an essential aspect of the model, required to reach the paper's conclusions, is that this early plate tectonic regime operated ~40 times faster than modern rates. This assertion does not give any insight into whether plate tectonics actually works in this regime. "

→ We realized that conditions for plate tectonics were not discussed in the previous manuscript.We added the following sentence in the main text and a new paragraph in Methods to discuss the likelihood of the Hadean plate tectonics based on the scaling law of Korenaga(2010):

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P. 7

Stress necessary to bend plates is smaller with thinner depleted lithospheric man- tle, whereas stress due to the negative buoyancy of plates increases under a higher potential temperature, so the operation of plate tectonics is plausible (see Methods).

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P. 19 (Methods)

For plate tectonics to operate, the negative buoyancy of the cold lithosphere should be large enough to bend subducting plates. It is noted that the term plate plate tectonicshere refers to a mode of mantle convection that allows for the recycling of the surfacelayer, and plate kinematics could be different from modern-style plate tectonics. The criteria for plate tectonics is presented in Korenaga (2010) as

$$\Delta\eta_L \leq 0.25Ra^{1/2} \equiv \Delta\eta_{L,crit}, \qquad (1)$$

where $\Delta\eta_{L,crit}$ denotes the critical value for the operation of plate tectonics. Forboth chemically homogeneous and heterogeneous mantles, the lithospheric viscosity contrast is smaller than the critical value (Figure S5), and thus plate tectonics is possible in the Hadean. Because water oceans were likely present at the surface,we consider that thermal cracking weakened the stiff lithosphere (Korenaga, 2007) to reduce the value of effective friction coefficient to $\mu$=0.02–0.03. The existenceof oceans is the key to plate tectonics, because, if water oceans were absent in the Hadean, the value of $\mu$ increases to ~0.8, resulting in a large lithospheric viscosity contrast. In such a case, the Hadean Earth may have operated under the stagnant lidmode, which lacks the recycling of the surface.

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"The paper argues that under these circumstances, mean subducted oceanic lithosphere age is5 Ma. But how does this work? What are the negative buoyancy forces driving subduction for 5 Ma lithosphere? And how would plate tectonics operate if those buoyancy forces are notstrong? Without some calculation that this could work, the model is not convincing.

→ For the chemically heterogeneous mantle, plates are likely to be negatively buoyant be- cause crust formed by the melting of iron-rich blobs would have a density higher than 3500

kg/m$^{-3}$ and would act as a weight inside oceanic crust. We added the following sentences in the main text to clarify this issue. For a pyrolitic mantle, we have consideredthe buoyancy criterion which is indicated by a dotted line in Figure 3C. Plates become negatively buoyant after 20 Myr of formation for the present-day mantle, and thus for apyrolitic mantle, the buoyancy of plates is a limiting factor only when it is hotter than 1590 ˚C.

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P. 8

Because the depleted mantle is drier and thus stiffer, thicker lithosphere results in slower convection than the present-day mantle (Korenaga, 2006; Figure 3c). Also, the presence of thick buoyant crust would require a longer cooling time for plate to be negatively buoyant (Davies, 1992), which would further limit the plate velocity. The scaling of plate tectonic convection (Korenaga, 2010) suggests that, for a pyroiticmantle, average plate velocity would remain the same as the present at a potential temperature of 1600 ˚C. In contrast, the depleted lithospheric mantle of chemically heterogeneous mantle is thin enough to allow for rapid plate motion, and its crust isonly marginally chemically buoyant because of crust produced from iron-rich blobs, which has a density greater than 3500 kg m$^{-3}$ (Table S2) and would act as a weight inside oceanic crust.

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2. "The focus here is that plate manufacture is rapid, that lithosphere formation is retarded owingto a lack of mantle dehydration, and that the ultramafic character of much of the sea floor forms an efficient $CO_2$ sink. But an essential factor, the survival of $CO_2$ in subducting lithosphere is not addressed. How do we know whether C is actually subducted, rather than stripped from the downgoing plate? It is controversial whether C is subducted into convecting mantle today(e.g., Kelemen and Manning, 2015), but wholly speculative as to whether it would be for a hotter (i.e., younger) plate subducting into a hotter mantle. The faster presumed subduction rates may counteract these problems, and so .maybe. But the onus is on the authors to demonstrate so with a pertinent calculation."

→ We agree that some fraction of subducted carbonates would return to the surface during the subduction, and we now discuss this issue in the following paragraph:

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P. 20–21 (Methods)

Some amount of carbonates created by hydrothermal alteration may decompose and return to the hydrosphere during subduction. The main source of loss is expected to be dissolution of carboantes to serpentine-driven fluid. The estimate of carbon being removed from the plate remains controversial for the present (Dasgupta and Hirschmann, 2010; Kelemen and Manning, 2015), but the Hadean plates are expected to contain ~6 times more carbonates than the present-day oceanic crust, and thus itis unlikely for all carbonates to be dissolved. When the top 500 m of oceanic crust contains ~10 wt% of water, the maximum amount of $CO_2$ that could dissolve into serpentinite-driven fluid is $4.4\times10^{17}$ kg, which is less than 1% of total carbonates in oceanic crust. Here, we assume that serpentinize releases water at ~3 GPa and the solubility of carbonates to fluid is $10^4$ ppm (Kelemen and Manning, 2015). More- over, some fraction

of the extracted carbon would be stored in mantle lithosphere and continental crust (Kelemen and Manning, 2015), the latter of which would have been created by the melting of hydrated subducted materials. Therefore, not all recycled carbon would return to the hydrosphere. Even if we assume that 50% of the sub- ducted carbon returns to the hydrosphere, the sequestration of carbon would still becompleted within ~260 Myr. It is noted that decarbonation reactions may not be effi- cient under a hotter mantle as well. Carbonates releases $CO_2$ at a hotter temperature,but even under a hotter subduction geotherm, the top of oceanic crust would be cold enough to stabilize carbonates (Figure S8). The subduction geotherm, however, would depend on various factors including the dip angle, and phase equilibria under different mineral and melt assemblage would also be necessary for more detailed discussion.

3. "The model presented here is launched from a previous paper by Miyazaki and Korenaga argu- ing that following magma ocean crystallization, the mantle consists of a highly heterogeneousmix of depleted peridotite salted with Fe-enriched domains. Here, they assert that this depleted peridotite has a Mg# of 95 and therefore has a high melting temperature, resulting in little crust production and in inefficient mantle dehydration, resulting in thin lithosphere. Several problems arise:

   Whereas the earliest crystallizing products of a magma ocean may have a Mg# of 95, later products would not be similarly extreme. The assumption that large fractions of the mantle would be this end-member composition needs to be justified with some kind of mass balance. After all, the average mantle must remain close to a Mg# of 89. And what are the proportionsand compositions of the Fe-rich domains?"

   → We thank the reviewer for raising this important point. We realized that our earlier assumption of high-Mg# matrix being peridotitic was incorrect and that the situation required a more careful thermodynamic analysis, which we now present in the revised manuscript. Overall, this newly added thermodynamic analysis has provided us a more complete picture of the likely geodynamic regime in the Hadean. Whereas this picture is different from our original speculation in a few important details, the main conclusions of our earlier manuscript still hold (see also our response to comments #4–8). To respond tocomment #3, we have added the following paragraphs in the Supplementary Information:

   Supplementary Information P. 2

   The fractional crystallization of a magma ocean produces a structure dominated by the high-Mg# matrix with small iron-rich blobs (Figure 3a). Miyazaki and Korenaga estimated that the ternary composition of the high-Mg# matrix is on average MgO 41.7 wt%, FeO 4.4 wt%, and $SiO_2$ 53.9 wt%, whereas that of iron-rich blobs is MgO 41.0 wt%, FeO 20.6 wt%, and $SiO_2$ 38.3 wt%. The Mg# of the two components are ~94 and ~77, respectively, resulting in contrasting densities and melting tempera- tures. It is noted that iron-rich blobs make up ~25% of the heterogeneous mantle, and the average composition of the mantle is consistent with the pyrolitic mantle.

   The solidus of such a high-Mg# matrix is expected to be higher than that of a py- rolitic

mantle, and we estimate the solidus temperature using the Rhyolite-MELTS model (Gualda et al., 2012). To predict the mineralogy of high-Mg# matrix, the $Al_2O_3$ and CaO contents are estimated using a partitioning model described in the following paragraph. Combining the results of the partitioning model with the ternary composition, we obtain an estimate of MgO 39.5 wt%, FeO 4.1 wt%, $SiO_2$ 51.3 wt%, $Al_2O_3$ 3.5 wt%, and CaO 1.5 wt% for the high-Mg# matrix, which results in a mineralassemblage of 70 wt% pyroxene, 25 wt% olivine, 5 wt% spinel at 1400 ˚C and 1.2 GPa.For Fe-rich blobs, a composition of MgO 37.8 wt%, FeO 19.0 wt%, $SiO_2$ 35.3 wt%, $Al_2O_3$ 3.7 wt%, and CaO 4.1 wt% is predicted, resulting in a mineral assemblage of 84 wt% olivine, 8 wt% of merwinite, 6 wt% of spinel, and 2 wt% of periclase. The melt fraction of the two components are shown in Figure S2.

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→ The high-Mg# matrix is inherited from the layer created during magma ocean by the settling and sedimentation of bridgmanite. The composition of this sedimented layer indeed varies with depth, and the value of Mg# is estimated to range between 92 and 95.Because this layer is expected to be well mixed by the Rayeligh-Taylor instability, we usethe mean value of ~94 to represent its composition.

4. “There is little reason to believe that the solidus temperature of peridotite can be extrapo- lated as a simple function of Mg#. Rock with such extreme composition will not likely have pyrolite mineralogy. It seems that the source-model from the earlier paper has no associatedmineral information? If so, the application of the simple solidus parameterization given here is speculative.”

→ As mentioned in our reply to comment #3, we now calculate the melting of the high-Mg# matrix using the Rhyolite-MELTS model, and the initial depth of melting is corroboratedwith the pMELTS model. The mineralogy presented in the Supplementary Information is used to calculate the solidus and liquidus, and its results are described in Figure S2. TheMELTS model suggests that the solidus temperature is indeed not as high as 1600 ˚C, but because the high-Mg# matrix is predominantly made of pyroxene, the melting and dehydration of the heterogeneous mantle would be different from the case of an olivine-rich pyrolitic mantle. We modified the following paragraph to explain the difference:

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P. 7

The chemically heterogeneous mantle is predicted to experience limited dehydrationat mid-ocean ridges. Modeling of magma ocean solidification suggests that the pyrox- enite matrix has an average Mg# of 94 and constitutes ~75% of the mantle (Miyazakiand Korenaga, 2019). The high-Mg# pyroxenite yields a higher solidus than a ho- mogeneous pyrolitic mantle, and with a mantle potential temperature of 1600 ˚C, the Rhyolite-MELTS and pMELTS models (Ghiorso et al., 2002; Gualda et al., 2012) pre-dict that the initial depth of melting is ~4 GPa with 0.1 wt% of water (Figure S2a). As the mantle undergoes fractional melting, the pyroxenite matrix becomes mostly dehydrated at ~2 GPa and forms a depleted lithospheric mantle of 40 km thickness(Figure S2b). (...) Pyrolite, on the other hand, starts to melt at a depth deeper than 10 GPa, and because it is rich in olivine, which is less compatible with water than pyroxene, a pyrolitic mantle would dehydrate faster than pyroxenite. Crust and de- pleted

lithospheric mantle are thus estimated to be as thick as ~26 km and ~145 km, respectively, at a mantle temperature 250 K hotter than the present (Figures 3b, S3).

5. "The model assumes that the highly depleted Mg #95 peridotite remains sufficiently hydrousto have low viscosity because it does not melt enough to lose appreciable $H_2O$. However, the paper acknowledges that such a lithology will melt to a small degree. It asserts without any quantification that the extent of melting would be too small to dehydrate the residue. In its present form, it is more of a speculation than a constraint. It needs a calculation. What melt fractions are expected? What is the applicable partition coefficient (at 1600 °C)? This requires saying something about the mineralogy. If the mineralogy is dominantly olivine, then the effective partition coefficient is a small fraction of that applicable to fertile peridotite andmerely 0.1% melting would be sufficient for near-complete dehydration. If the mineralogy is more "normal", then the melt fraction would have to be no more than about 1% for the residuesignificant $H_2O$."

→ As we mentioned above, we realized that the high-Mg# matrix has a high $SiO_2$ content and is composed of ~70% pyroxene. We have added the following sentences to explain how the melting and dehydration of the pyroxenite would proceed:

P. 16–17 (Methods)

The difference in plate velocity between the chemically homogeneous and heteroge- neous mantles arises from $h_m$. Because a hotter mantle with the same composition would melt by a larger degree, the value of $h_m$ for a pyrolitic mantle would be thickerthan the present. To predict the thickness of dehydrated lithosphere, a fractional melting model is constructed based on the melting model of Katz et al. (2003). As- suming that a fraction of the partial melt escapes to the surface with each incrementof new melting (see Supplementary), the depleted mantle would thicken to ~140 km(4.5 GPa) for a mantle potential temperature of 1600 °C, from ~60 km (2 GPa), which is the thickness for the present-day depleted lithospheric mantle (Figure S3).On the other hand, the value of $h_m$ for the chemically heterogeneous mantle is pre- dicted to be ~40 km based on the Rhyolite-MELTS models (Gualda et al., 2012), andthe initial depth of melting has been checked with the pMELTS model (Ghiorso et al., 2002). Because the high-Mg# pyroxenite starts to melt at a shallower depth thana pyrolitic homogeneous mantle, and also because the partition coefficient of water between pyroxene and melt is larger than that between olivine and melt, we estimatethat the mantle would not be completely dehydrated until 2 GPa. A value of 0.02 is adopted for water partitioning between the pyroxenite and melt, and 0.005 for par-titioning between pyrolite and melt (Aubaud et al., 2004). It is noted that iron-rich blobs would melt at a depth deeper than 10 GPa (Figure S2c), but the melting of blobs only creates locally depleted regions and thus is unlikely to affect the overall value of $h_m$.

6. "An important feature of the paper, leading to the conclusion of high carbon sequestration and hot spring activity conducive to prebiotic chemistry, is the assumption that the earliest seafloor consisted chiefly of ultramafic peridotite. This, it is said, is owing to the low crust production from

highly depleted magma ocean residues. However, it is not made clear how thisis reconciled with the requirement that the average mantle must have remained (nearly) the same composition as modern peridotite (actually, a little more fertile, as the continents had not yet been extracted). What is the role of the Fe-enriched domains in crust production? If thesedomains were widely sampled by "ridges", then they should have produced a great deal of extra crust. Stolper and Asimow (2007) showed that compared to homogeneous mantle, dividing the mantle into depleted and enriched domains produces about the same amount of crust, provided the two approximate thermal equilibrium. Even if they aren't in thermal equilibrium, the amount of crust produced by the enriched domains should be substantial for a mantle potential temperature of 1600 ˚C."

→ We agree with the reviewer that the chemically heterogeneous mantle would produce about the same amount of crust with a homogeneous pyrolitic mantle. Indeed, with a mantle potential temperature of 1600 ˚C, iron-rich blobs are expected to melt up to ~40%. To discuss the fate of crust formed from iron-rich blobs, we added the following sentences in the main text and in the Supplementary:

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P. 7–8

Iron-rich blobs would also melt during their upwelling, but because they are of small scale, their melting would not contribute to producing a uniformly thick depleted lithospheric mantle. (...) In contrast, the depleted lithospheric mantle of chemically heterogeneous mantle is thin enough to allow for rapid plate motion, and its crust isonly marginally chemically buoyant because of crust produced from iron-rich blobs, which has a density greater than 3500 kg $m^{-3}$ (Table S2) and would act as a weight inside oceanic crust.

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Supplementary Information P. 4

With a mantle potential temperature of 1600 ˚C, the high-Mg# pyroxenite matrix would experience ~27% melting, whereas iron-rich blobs would melt up to ~43% when blobs maintain a thermal equilibrium with the pyroxenite matrix (Figure S2c).Iron-rich blobs are predominantly consisted of olivine, and the partial melt would be further enriched in iron. The Rhyolite-MELTS model (Gualda et al., 2012) predicts that such a crust has a composition of MgO 16.0 wt%, FeO 22.9 wt%, $SiO_2$ 39.2 wt%,$Al_2O_3$ 9.2 wt%, and CaO 12.6 wt%, resulting in an Mg# of 56 and a crustal density higher than 3500 kg $m^{-3}$ (Table S2). The melt density of partial melt from iron-richblobs is 3000 kg $m^{-3}$, which is still lower than the density of the pyroxenite matrix, so the iron-rich melt would migrate upward.

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P. 18 (Methods)

Fe-rich blobs start to melt at a deeper depth, so its crust would have an average thickness of ~11 km. The crust of the chemically heterogeneous mantle would not be uniform, but the average crustal thickness is predicted to be ~15 km, ~70% of which originates from Fe-rich blobs. It is noted that the density of partial melt fromiron-rich

blobs is estimated to be 3000 kg $m^{-3}$, which is still lower than the high- Mg# matrix. Partial melt from iron-rich blobs would thus migrate upwards within the high-Mg# matrix. However, the density difference between iron-rich melt and the matrix is approximately four times smaller than that between partial melt from the matrix and the matrix itself, so these two kinds of partial melt are expected to migrate upward without much mixing, and majority of the iron-rich crust is likely tobe situated below the high-Mg# crust.

7. "What suppresses the participation of the Fe-rich domains in melt production? Is it that theyare so large and widely dispersed, that they make large amounts of crust but in very restrictedportions of the sea-floor? Is it supposed that the more Fe-rich domains that formed from late-stage magma ocean crystallization were initially hidden in the lower mantle? What is their average length scale at the point of their formation, and how rapidly did they get stirredback in to make "average" mantle? In the modern mantle, convective stirring homogenizes subducted oceanic crust with the rest of the mantle within a few hundred million years (e.g., Kellogg and Turcotte, 1990). If convective stirring during the time of interest was ~40 times more vigorous, then one would expect that quite rapidly the mantle would become a streaky marble cake that produced a lot of crust. Does the model have some mechanism for preventing the Fe-rich domains from participating in melting? And, returning to an earlier point, by mass balance, what fraction of the mantle resides in these Fe-rich domains? Without addressing these questions, the inference of a peridotite-dominated melting regime seems to derive from a selective description of the system."

→ As we discussed above, we realized that Fe-rich blobs would melt and create patches of heavy crust. Considering that iron-rich domain constitutes 25% of the mantle, the iron- rich crust would constitute more than a quarter of crustal component. The density of produced crust, however, is higher than 3500 kg/$m^3$, and thus would be heavier than crust produced from the high-Mg# pyroxenite and also than ambient mantle. Therefore,iron-rich crust may not be widely exposed to the surface, and the area of iron-rich crust would be limited to a small portion of the surface.

→ Timescale for mixing between the high-Mg# pyroxenite, iron-rich blobs and crust depends on a number of parameters. We have added the following sentences to discuss this issue:

P. 10

Small-scale heterogeneities would eventually be homogenized with high-Mg# pyrox- enite as convective mixing proceeds, but homogenization likely takes a few tens to hundreds million years to complete (Davies, 2006).

Supplementary Information P. 4–5

The preexisting lithological heterogeneities (high-Mg# matrix and iron-rich blobs) as well as the newly generated heterogeneities (crustal products and solid residues) would eventually be homogenized as the mantle evolves, but timescale for mixing dependson many uncertain parameters (Tackley, 2015). For example, the subducted iron-rich crust would be denser than ambient mantle, so it may stay longer in the lower mantlewithout

being homogenized with the entire mantle (Davies, 2006). It is not unrealistic to expect that the initial heterogeneity likely survived until the end of the Hadean.

**Reviewer Reports on the First Revision:**

**Referee #1:**

The paper has been fixed in accord with the comments. It is OK with me.

Norm Sleep

**Referee #2:**

2nd Review of Miyazaki and Korenaga "A new mode of geodynamics in the Hadean facilitates the emergence of early life" for Nature

Marc Hirschmann

This is my second review of this ms. It is unfortunate that it was not written more promptly, but when asked by Nature editors to take on this review, I replied that I would not have time for it until mid-February, and agreed to the review only with that understanding.

The revised manuscript is substantially different from the original, though the basic theme remains the same – that the heterogeneous mantle descended from crystallization of the magma ocean had special properties that lent themselves to effectively drawing down of the thick CO2 atmosphere expected on the earliest Earth. However, in the new version, those special properties, and in particularly, the hypothesized petrologic nature of the heterogeneities, are entirely different from the original. In the original, one key lithology was highly depleted pyrolite, but now that is considered to be a high Mg pyroxenite, produced from bridgemanite cumulates during magma ocean crystallization. In the original, the contribution of the more Fe-rich ("Fe-rich blobs") to hadean oceanic crust was ignored. Now it is considered essential to provide the negative buoyancy that might drive early vigorous plate tectonics even though the plates are hot and young. But despite these momentous changes, the authors argue that the essential nature of this special heterogeneous mantle had the same effect- bringing about the rapid onset of a hyper-plate tectonic regime that effectively sequestered carbon back into the mantle

This paper is full of interesting ideas and, as I wrote in my original review, addresses a fundamental problem about how the Earth evolved- the fate of the carbon that almost certainty was initially degassed to the surface but apparently returned to the mantle early in Earth history. Too few are aware of the importance of this problem, and even fewer have worked on possible solutions. However, the ideas and processes discussed in the paper remain rather loosely argued. It is far from convincing that the scenario described is likely, though I don't know that it is unlikely either.

Below I add some specific commentary. I apologize to the editors, but I don't have a strong recommendation for or against publication. I think that the paper is timely and will stimulate a lot of new research, but I also think that parts of it are highly vulnerable to criticism.

I will not review this ms. a third time. Based on reviewers input and/or another round of revisions, I leave it to the editors or others to make a decision.

Specific points:
1. An essential assumption for calculation of CO2 uptake by hadean oceanic crust is that the upper 500 meters is entirely derived from the depleted "Mg pyroxenite" mantle source rather than from the "iron blob" mantle source. The justification for this is that the latter is less dense, and so that it is supposed that when the two ascend together from the mantle, they erupt in some kind of unmixed stratified fashion. There really is no reason to

assume this and it's not even clear what mechanism might allow it. Do the authors envision that one lithology erupts volcanically, whereas the other only makes sills or other intrusive rocks? Why would this be, as the density of their melts in Table S2 is less than the density of the crystallized crust from either lithology? Do they envision that the denser magmas in the crust are uneruptable and also unmixable? More likely, the different sources would contribute variably to individual volcanic eruptions varying in time and space, making for a volcanic pile that is heterogeneous.

At a minimum, I think that a revised ms. should recalculate the potential $CO_2$ draw down based on the alternative assumption that the two lithologies contribute to the upper 500 m layer of crust in the same proportions as they are erupted.

2. Based on comments from the initial review, the revised manuscript now considers the preservation of C in the downgoing subducting slab. The essential argument seems to be that Hadean subducted crust was so C-rich that any loss processes would have been ineffective, however, much about the specific discussion seems ad hoc and stitched together. Most of the discussion is about the possible efficacy of dissolution reactions in hadean subducted crust. Apart from the fact that this discussion seems to misunderstand how these work (the text considers $H_2O$ coming from hydrated subducted crust, whereas the chief mechanism for carbonate dissolution in subducted crust is $H_2O$ coming from underlying serpentinized subducted lithosphere), this misses the much more important consideration of the distinct thermal and structural architecture of Hadean subduction as opposed to modern. Here, the text only refers to a supplementary figure (S8), saying the temperatures aren't high enough to decompose carbonate. But Fig. S8 is problematic.

The starting point for Fig S8 is the thermal structure of the modern South Chile subduction zone, which the authors assert is a hot endmember in modern subduction. However, Fig. S8 seems to have the thermal structure of South Chile located incorrectly – the source is Syracuse et al., which has the South Chile slab surface temperature of 276 °C at 30 km depth, whereas Fig. S8 has it at about 150 °C. Further, the South Chile ridge is by no means the hottest endmember on the modern Earth – the Cascadia subduction zone is 100 °C hotter, and this is likely relevant to the Hadean given the very young lithosphere that is subducting there. Fig. S8 further assumes that Hadean subduction would be 200 °C hotter than the S. Chile, simply based on differences in mantle potential temperature. This may or may not be accurate, but it doesn't take into account the distinct Hadean lithosphere thermal structure and architecture (about which much is made elsewhere in the paper) or kinematics that the paper supposes drive the early rapid plate tectonic regime. But most importantly, Fig. S8 only considers the shallow portion of the subducting system (shallower than the so-called decoupling point at ~80 km) and therefore does not address the (carbonated eclogite/pyroxenite) melting reactions that would be encountered when the slab surface is rapidly heated at pressures above 3 GPa.

A revised paper should take into account the temperature profiles of the slab surface temperature to at least 100 km, and compare them to solidi of carbonated eclogite and hydrous carbonated eclogite. Arguments about the large amount of carbon in the subucted crust don't really matter. If the solidus is crossed, then the crust won't deliver the carbon to significant depth.

I don't know if carbon would be effectively subducted during the Hadean, but I conclude that the discussion added here does not clarify if it does or does not and therefore the current manuscript does not really address whether carbon recycling into the Hadean mantle is efficient or not.

3. An important part of the paper is the argument that $CO_2$ was fixed into oceanic crust more effectively in the Hadean. In the original ms., the seafloor was supposed to be ultramafic and capable of drawing down 200 bars $CO_2$ in 10 Ma. In the revised ms, the seafloor is mafic, and the calculation suggests that 200 bars are drawn down in 135 Ma. The chief argument now is not that the Hadean seafloor was a superior "sponge", but rather that the high $pCO_2$ promoted more pervasive reaction.

There are a few features of this that aren't explained very well. First, at the outset of the ingassing process, the $CO_2$ partial pressure is high and the surface temperature is above 100 °C (Fig. S6). Although I can see how this would result in very effective surface weathering, how would significant C penetrate into the "oceanic" crust without an ocean? Do the authors envision that some different type of hydrothermal circulation would occur? Second, once temperatures do fall below the boiling temperature, what kind of chemical dynamics are expected in that hot ocean below a thick $CO_2$ atmosphere? The ms. emphasized that $CO_2$ uptake on the seafloor would be more vigorous because $pCO_2$ would be higher, but what would be the carbonate compensation depth in such an

ocean, would the concentration of CO2 in seafloor seawater really be so high? Would there be significant authigenic carbonates precipitated on the sea floor, and would these be a substantial carrier of recycled carbon?

4. Paragraph beginning at line 47 – I should have caught this one in the first round of review - The idea presented here is about the importance of trapped liquid in storing H2O and C in the crystallizing magma ocean. The paragraph, as it stands, states that previous treatments of volatile behavior during magma ocean crystallization have ignored trapped liquid, but this isn't true. This specific point was the main thrust of Hier-Majumder and Hirschmann (2017, G3), which should be credited and cited.

**Author Rebuttals to First Revision:**

**Response to Referee #2 (Marc Hirschmann):**

1. "An essential assumption for calculation of $CO_2$ uptake by hadean oceanic crust is that the upper 500 meters is entirely derived from the depleted 'Mg pyroxenite' mantle source rather than from the "iron blob" mantle source. The justification for this is that the latter is less dense, and so that it is supposed that when the two ascend together from the mantle, they erupt in some kind of unmixed stratified fashion. There really is no reason to assume this and it's not even clear what mechanism might allow it. Do the authors envision that one lithology erupts volcanically, whereas the other only makes sills or other intrusive rocks? Why would this be, as the density of their melts in Table S2 is less than the density of the crystallized crust from either lithology? Do they envision that the denser magmas in the crust are uneruptable and also unmixable? More likely, the different sources would contribute variably to individual volcanic eruptions varying in time and space, making for a volcanic pile that is heterogeneous.

   At a minimum, I think that a revised ms. should recalculate the potential $CO_2$ draw down based on the alternative assumption that the two lithologies contribute to the upper 500 m layer of crust in the same proportions as they are erupted."

→ The lithology of the upper crust does not have a large influence on $CO_2$ uptake by the seafloor becauseboth high-Mg# and Fe-rich crusts contain high MgO and CaO concentrations. We modified the paragraph below to clarify this issue:

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**P. 21**

The crust of the chemically heterogeneous mantle would be a mixture of those derived from high-Mg# pyroxenite and Fe-rich blobs, and both components have a large capacity of storing $CO_2$. Because the formation of carbonates is thermodynamically favorable for Ca- and Mg-silicates under a high $CO_2$ concentration (Sleep et al., 2001), we assume that Ca- and Mg-silicates are used to form carbonates. The crust formed from high-Mg# pyroxenite and Fe-rich blobs both contain ~10 wt% of CaO (Table S2), so when all Ca-silicates are carbonated, the upper crust would store up to $3.5\times10^{19}$ kg or ~7 bar of $CO_2$. This is 2.7 times of the present-day Earth, where the top 500 m of basaltic crust contains ~3.0 wt% of $CO_2$, equivalent to $1.3\times10^{19}$ kg or

~2.5 bar of $CO_2$ (Alt and Teagle, 1999). Mg-silicates, which mostly exist as olivine, orthopyroxene, or clinopyroxene (Table S2), are also prone to carbonation (Kelemen et al., 2011), among which olivine has the highest rate of carbonation. The crust of the heterogeneous mantle contains 16– 22% of MgO, depending on the relative proportion of high-Mg# and Fe-rich source crusts, so even with a lower bound estimate, the upper crust could contain up to $8.0\times10^{19}$ kg or ~15.4 bar of $CO_2$. This suggests that the seafloor could

contain up to ~22 bar of $CO_2$ at one time. Fe-silicates are not considered as a sink for $CO_2$ because they are more likely to be oxidized to produce $H_2$.

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2. "Based on comments from the initial review, the revised manuscript now considers the preservation of C in the downgoing subducting slab. The essential argument seems to be that Hadean subducted crust was so C-rich that any loss processes would have been ineffective, however, much about the specific discussion seems ad hoc and stitched together. Most of the discussion is about the possible efficacy of dissolution reactions in hadean subducted crust. Apart from the fact that this discussion seems to misunderstand how these work (the text considers $H_2O$ coming from hydrated subducted crust, whereas the chief mechanism for carbonate dissolution in subducted crust is $H_2O$ coming from underlying serpentinized subducted lithosphere), this misses the much more important consideration of the distinct thermal and structural architecture of Hadean subduction as opposed to modern. Here, the text only refers to a supplementary figure (S8), saying the temperatures aren't high enough to decompose carbonate. But Fig. S8 is problematic."

→ The water content in the underlying lithosphere is expected to be limited as we now discuss in thefollowing paragraph:

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**P. 23**

The loss of carbon may also take place by the dissolution of carbonates to fluid released by serpentine. The Hadean plates, however, are expected to contain ~9 times more carbonates thanthe present-day oceanic crust, and thus it is unlikely for all carbonates to be dissolved. When thetop 500 m of oceanic crust contains ~10 wt% of water, the maximum amount of $CO_2$ that could dissolve into serpentinite-driven fluid is $4.4\times10^{17}$ kg, which is less than 1% of total carbonates in oceanic crust. Here, we assume that serpentinize releases water at ~3 GPa and the solubility of carbonates to fluid is $10^4$ ppm (Kelemen and Manning, 2015). The amount of water in the underlying crust and mantle is predicted to be limited due to high confining pressure (Korenaga,2017; Miller et al., 2021). Considering that the top 30 km includes 0.1 wt% of water, the additional dissolution of $CO_2$ would be less than 1% of total carbonates in the upper crust. Therefore, the amount of recycled carbon through dissolution is expected to be negligible in the Hadean.

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"The starting point for Fig S8 is the thermal structure of the modern South Chile subduction zone, which the authors assert is a hot endmember in modern subduction. However, Fig. S8 seems to have the thermalstructure of South Chile located incorrectly – the source is Syracuse et al., which has the South Chileslab surface temperature of 276°C at 30 km depth, whereas Fig. S8 has it at about 150°C. Further, the South Chile ridge is by no means the hottest endmember on the modern Earth – the Cascadia subduction zone is 100°C hotter, and this is likely relevant to the Hadean given the very young lithosphere that is subducting there. Fig. S8 further assumes that Hadean subduction would be 200°C hotter than the S. Chile, simply based on differences in mantle potential temperature. This may or may not be accurate, but it doesn't take into account the distinct Hadean lithosphere thermal structure and architecture (about which much is made elsewhere in the paper) or kinematics that the paper supposes drive the early rapid plate tectonic regime. But most importantly, Fig. S8 only considers the shallow portion of the subducting system(shallower than the so-called decoupling point at ~80 km) and therefore does not address the (carbonated eclogite/pyroxenite) melting reactions that would be encountered when the slab surface is rapidly heated at pressures above 3 GPa.

A revised paper should take into account the temperature profiles of the slab surface

temperature to at least 100 km, and compare them to solidi of carbonated eclogite and hydrous carbonated eclogite. Arguments about the large amount of carbon in the subucted crust don't really matter. If the solidus is crossed, then the crust won't deliver the carbon to significant depth.

I don't know if carbon would be effectively subducted during the Hadean, but I conclude that the discussion added here does not clarify if it does or does not and therefore the current manuscript does not really address whether carbon recycling into the Hadean mantle is efficient or not."

→ We realized that our earlier attempt of estimating the Hadean geotherm was too crude, so we have conducted additional convection modeling to calculate geotherms for the Hadean subduction zones and added the following paragraph to discuss the stability of carbonate:

**P. 21–22**

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Carbonates created by hydrothermal alteration would mostly be delivered to the deep mantle under a rapid plate motion. The loss of carbonates in the plate is determined by two factors:

(1) the stability of carbonates under a hotter deeper region and (2) the solubility of carbon to water released from the decomposition of hydrous minerals (Kelemen and Manning, 2015). The stability of carbonates within oceanic crust depends on the $P$-$T$ path of subducting plates, and carbonates are considered to be stable under majority of the modern subduction settings (Dasgupta and Hirschmann, 2010). This is also likely to be the case for the Hadean subduction zones. Although the surface temperature and mantle potential temperature are both hotter by ~200 K,

the $P$-$T$ path of subducting slab would remain similar to the present-day young subduction zones (Figure S4). For the chemically heterogeneous mantle, the rapid plate velocity would allow the slab to be delivered to the deeper region without substantial heating from the surrounding mantle. A pyrolitic homogeneous mantle would have slower plate velocity, but because oceanic plates areolder and thus colder, the slab temperature at the deeper region would be similar to that ofthe heterogeneous mantle. The actual geotherm could vary among different subduction settings even with the same age and plate velocity (Syracuse et al., 2010), so the temperature of some slab surfaces may reach the solidus of carbonate melting (Dasgupta et al., 2004). On average, however, the subducting slab is expected to remain colder than the carbonate solidus (Figure S4), recycling only a fraction of subducting carbonates back to the surface. Moreover, some fraction ofthe extracted carbon would be stored in mantle lithosphere and continental crust (Kelemen and Manning, 2015), the latter of which would have been created by the melting of hydrated subducted materials. Modeling the thermal structure of subduction zones is still subject to large uncertainties as a number of important assumptions are involved, and in this study, we tentatively estimate that 50% of subducted carbonates return to the surface for both pyrolitic and heterogeneous mantles (Figure 4). Whereas the timescale of carbon sequestration would be longer for less efficient carbonate subduction, it would always be shorter by an order of magnitude for the case of the heterogeneous mantle.

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3. “An important part of the paper is the argument that $CO_2$ was fixed into oceanic crust more effectively in the Hadean. In the original ms., the seafloor was supposed to be ultramafic and capable of drawing down 200 bars $CO_2$ in 10 Ma. In the revised ms, the seafloor is mafic, and the calculation suggests that 200 bars are drawn down in 135 Ma. The chief argument now is not that the Hadean seafloor was a superior “sponge”, but rather that the high $pCO_2$ promoted more pervasive reaction.

There are a few features of this that aren’t explained very well. First, at the outset of the ingassing process,the $CO_2$ partial pressure is high and the surface temperature is above 100 ˚C (Fig. S6). Although I can see how this would result in very effective surface weathering, how would significant C penetrate into
the “oceanic” crust without an ocean? Do the authors envision that some different type of hydrothermal circulation would occur?”

→ We modified the following sentences to clarify that the surface temperature is below the water boilingtemperature given the high atmospheric pressure:

**P. 5**

This exceeds a threshold of 18 atm to form water oceans when the atmosphere contains ~200 barof $CO_2$ (Abe, 1993), so the presence of ocean is thus plausible immediately after the solidificationof a magma ocean. The surface temperature exceeds ~100 ˚C due to greenhouse effect, yet liquidwater is stabilized by high atmospheric pressure. At the same time, the mantle would still contain a large fraction of water and is expected to be hydrated, lowering the viscosity (Hirth and Kohlstedt, 1996; Jain et al., 2019) of the Hadean mantle.

“Second, once temperatures do fall below the boiling temperature, what kind of chemical dynamics are expected in that hot ocean below a thick CO2 atmosphere? The ms. emphasized that $CO_2$ uptake onthe seafloor would be more vigorous because $pCO_2$ would be higher, but what would be the carbonate compensation depth in such an ocean, would the concentration of $CO_2$ in seafloor seawater really be so high? Would there be significant authigenic carbonates precipitated on the sea floor, and would these be asubstantial carrier of recycled carbon?”

→ We modified the following sentences to address this issue:

**P. 20–21**

The basis for efficient carbonation is that the Hadean seafloor was supersaturated with respect tocarbonate minerals. The modern oceans become undersaturated below 3500–5000 m (Peterson, 1966; Andersson, 2014), but the carbonate compensation depth was likely deeper in the Hadean owing to two main factors (Andersson, 2014): (1) a higher surface temperature due to greenhouse effect and (2) an initially shallower ocean because of the greater water content of the mantle.The amount of water at the surface is estimated to have been less than 10% of the presentday in the Hadean (Figure 2), so ocean depth was likely shallower than 500 m. Even with an atmospheric pressure of ~200 bar, pressure at the seafloor would be lower than ~250 bar. Higher temperatures and lower pressures inhibit the dissolution of carbonates (Andersson, 2014) and thus promote carbonate alteration of the oceanic crust. It is noted that a lower pH due to a thicker $CO_2$ atmosphere would not be a limiting factor for carbonation. The relative proportion of carbonate ion to the total carbon amount decreases significantly with a lower pH, but

carbonate ion concentration would still be larger than the present, or at least remain the same, because the total amount of carbon in the seawater is substantially larger. The actual carbonate ion concentration remains uncertain due to limited experimental data, but the carbonation of oceanic crust is expected to be efficient as long as carbonate ion is present and dissolution is inhibited.

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4. "Paragraph beginning at line 47 – I should have caught this one in the first round of review - The idea presented here is about the importance of trapped liquid in storing $H_2O$ and C in the crystallizing magma ocean. The paragraph, as it stands, states that previous treatments of volatile behavior during magma ocean crystallization have ignored trapped liquid, but this isn't true. This specific point was the main thrust of Hier-Majumder and Hirschmann (2017, G3), which should be credited and cited."

→ We now cite and acknowledge the work by Hier-Majumder and Hirschmann (2017) in the main text.

**Reviewer Reports on the Second Revision:**

**Referee #1**

I have limited internet access today so I will be brief.

(1) Hot runaway atmosphere was opaque. There were sulphur and chloride species at intermediate levels. Radiation was at ~300K from water cloud tops. The available heat from the moon-forming impact and tides took crudely 10 million years to escape. So the thermal boundary layer at the top of the magma ocean also had the heat flux from the top of the atmosphere. The temperature contrast at the top of the magma ocean adjusted for this heat flux.

Basically mantle material had to pass through the thermal boundary layer to cool. Each batch of the mantle had to pass (multiple times) through the thermal boundary layer. Exchange of volatiles with air and silicate occurred within the thin layer. So equilibrium occurred at P and T of the base of the atmosphere. Water now degasses at 250 bars at the ridge axis. One ocean of Earth with no topographic relief is 250 bars. So I am somewhat skeptical of very little water being at the surface at the end of degassing and start toward clement Earth. But it is worth exploring the possibility.

(2) The surface of the magma ocean starts to freeze and there is transition to solid lid near the end of the runaway greenhouse. It is very hard to get rheology and degassing for this time. There may be sufficient caveats already in the paper. Thin layers of very low viscosity magma could repeatedly erupt to the top and freeze. A thick cold lid could form this way. The material eventually sank. There is very little chemical differentiation this way. The mantle could overcool. Korenega's models have nonmonotonic cooling. My two cents.

(3) Arc magma from fully carbonatized crust should come in. Kate Kiseeva has done experiments. One can get peralkaline magmas. There is excess K and high pH with cooling. At hydrothermal and clement conditions brines are alkaline. The mineral aegergine KFe(III)Si2O6 forms. This sucks oxygen out of the magma. Graphite can form. Hydrocarbonates can form on cooling. Sodium silicate is soluble. See Lambert paper on stabilizing ribose this way. So more biochem can be done.

XXX So I think subduction of highly carbonatized oceanic crust is inevitable. Mantle heterogenieties are likely. I do not like very little surface ocean, but that is me. There biological conditions might be made more friendly to the biologist. If that is not done, it should be stated that this is out of the scope.

My name should go to the authors.

Norm Sleep
Thanks you

**Author Rebuttals to Second Revision:**

## Response to Referee #1 (Norm Sleep):

1. "Hot runaway atmosphere was opaque. There were sulphur and chloride species at intermediate levels. Radiation was at ~300 K from water cloud tops. The available heat from the moon-forming impact and tides took crudely 10 million years to escape. So the thermal boundary layer at the top of the magma ocean also had the heat flux from the top of the atmosphere. The temperature contrast at the top of the magma ocean adjusted for this heat flux."

   → We added the following paragraph to clarify that the greenhouse effect is considered when estimating the surface temperature and the heat flux:

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**P.28**

The surface temperature, $T_s$, is a function of the atmospheric mass and composition and the net outgoing infrared radiation, and the outgoing radiation, $F_{out}$, is described as the sum of absorbedsunlight and convective heat flux (Zahnle et al., 1988; Solomatov 2007):

$$F_{out} = (1 - A)F_{\odot} + 0.089\frac{k(T_m - T_s)}{d}Ra^{1/3}, \quad (1)$$

where $A$ is the albedo, $F_{\odot}$ is solar flux (values discussed in a later section), $k$ is thermal conduc- tivity, $T_m$ is mantle potential temperature, $d$ is the depth of the melt-dominated layer, and $Ra$ isthe Rayleigh number of the melt-dominated layer. This value is set as a boundary condition ($F_{\uparrow}$ at $\tau = 0$) in the atmospheric model.

**P.45**

We assume that the early Earth atmosphere contains 1 bar of $N_2$ with a varying mixture of $CO_2$ and $H_2O$, which are determined based on the partitioning of volatiles between a magma ocean and the atmosphere. Sulfur and chloride volatiles may have also contributed to the greenhouse effect (Johnson et al., 2008), but its effect is likely to be smaller compared to that created by a thick atmosphere of $CO_2$ and $H_2O$.

Basically mantle material had to pass through the thermal boundary layer to cool. Each batch of the mantle had to pass (multiple times) through the thermal boundary layer. Exchange of volatiles with air andsilicate occurred within the thin layer. So equilibrium occurred at *P* and *T* of the base of the atmosphere. Water now degasses at 250 bars at the ridge axis. One ocean of Earth with no topographic relief is 250 bars. So I am somewhat skeptical of very little water being at the surface at the end of degassing and start toward clement Earth. But it is worth exploring the possibility."

→ We agree that the mantle would have rapidly degassed after the solidification of the mantle surface.

We modified the following sentences to describe how water ocean mass evolved during the Hadean:

**P.10–11**

On the other hand, hydrous minerals are vulnerable to dehydration upon subduction, so a rapid plate motion would result in a net increase in surface water through degassing at ridges. Given ahigher processing rate than the present-day mantle, the Hadean mantle would have dried out in ~500 Myr (see Methods), and an initially small ocean mass would have increased to the present-day level or higher by the beginning of the Archean.

1. "The surface of the magma ocean starts to freeze and there is transition to solid lid near the end of the runaway greenhouse. It is very hard to get rheology and degassing for this time. There may be sufficient caveats already in the paper. Thin layers of very low viscosity magma could repeatedly erupt to the top and freeze. A thick cold lid could form this way. The material eventually sank. There is very little chemicaldifferentiation this way. The mantle could overcool. Korenega's models have nonmonotonic cooling. My two cents."

→ While chemical differentiation during magma ocean solidification is expected to be significant, we agree that episodic eruptions after the solidification of the mantle surface would not contribute to differentiation. We added the following paragraph in the method to discuss transition from magma ocean to solid-state convection.

P.31

It is noted that a small amount of additional degassing by episodic eruptions may take place during the transition from magma ocean to solid-state convection. Our model assumed that the Rayleigh-Taylor instability efficiently cools the solid-dominated layer (Maurice et al., 2017; Miyazaki and Korenaga, 2019), and the layer maintains an adiabatic thermal profile within the timescale of magma ocean solidification. Cooling, however, could be inhibited by tidal heating (Zahnle et al., 2015), and such heat may be released by episodic eruptions in addition to convective heat flux after the solidification of the mantle surface. Consequently, water degassing could be promoted, and ocean mass may increase rapidly. Yet, the interplay between tidal heating, cooling by the Rayleigh-Taylor instability, and melt percolation remains unconstrained, and future workis required to quantify the degree of degassing during this period. Episodic eruptions, however, would not affect the overall mode of mantle dynamics. Erupted melt would be recycled to the mantle by plate tectonics, and the rate of carbon sequestration is expected to remain unchangedfrom our model results.

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2. “Arc magma from fully carbonatized crust should come in. Kate Kiseeva has done experiments. One can get peralkaline magmas. There is excess K and high pH with cooling. At hydrothermal and clement conditions brines are alkaline. The mineral aegergine $KFe(III)Si_2O_6$ forms. This sucks oxygen out of the magma. Graphite can form. Hydrocarbonates can form on cooling. Sodium silicate is soluble. See Lambert paper on stabilizing ribose this way. So more biochem can be done.”

→ Although we moved the relevant paragraph to the supplementary, we expanded on the possibility ofbiochemistry at the Hadean seafloor:

P.10

Given that rocks constituting the LCHF are rich in olivine and have low Si content, a wide exposure of pyroxenite may have created a similar environment to the LCHF at most of seafloor in the earlyHadean. Alkailne conditions may also develop at an environment resembling hydrothermal vents, and combined with the high dissolution rate of olivine, abiotic synthesis of ribose could occur (Lambert et al., 2010). Therefore, the emergence of primordial life may have happened globally owing to the seafloor rich in olivine and orthopyroxene.

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3. “So I think subduction of highly carbonatized oceanic crust is inevitable. Mantle heterogenieties are likely.I do not like very little surface ocean, but that is me. There biological conditions might be made more friendly to the biologist. If that is not done, it should be stated that this is out of the scope.”

→ Following the comment by the editor and the referee, we modified the title and

the first paragraph toreduce the emphasis on implications for life.