Constraining the Volume of Earth's Early Oceans With a Temperature-Dependent Mantle Water Storage Capacity Model

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Abstract The water content in Earth's mantle today remains poorly constrained, but the bulk water capacity in the solid mantle can be quantified based on experimental data and may amount to a few times the modern surface ocean mass (OM). An appreciation of the mantle water storage capacity is indispensable to our understanding of how water may have cycled between the surface and mantle reservoirs and changed the volume of the oceans through time. In this study, we parameterized high pressure-temperature experimental data on water storage capacities in major rock-forming minerals to track the bulk water storage capacity in Earth's solid mantle as a function of temperature. We find that the mantle water storage capacity decreases as mantle potential temperature ($T_p$) increases, and its estimated value depends on the water storage capacity of bridgmanite in the lower mantle. 1.86–4.41 OM with a median of 2.29 OM for today ($T_p = 1600$ K), and 0.52–1.69 OM with a median of 0.72 OM for the early Earth's solid mantle (for a $T_p$ that was 300 K higher). An increase in $T_p$ by 200–300 K results in a decrease in the mantle water storage capacity by 1.19$^{+0.9}_{-0.36}$–1.56$^{+1.1}_{-0.22}$ OM. We explored how the volume of early oceans may have controlled sea level during the early Archean (4–3.2 Ga) with some additional assumptions about early continents. We found that more voluminous surface oceans might have existed if the actual mantle water content today is > 0.3–0.8 OM and the early Archean $T_p$ was ≥1900 K.

Plain Language Summary At the Earth's surface, the majority of water resides in the oceans, while in the interior, major rock-forming minerals can incorporate significant amounts of water as hydroxyl groups (OH), likely forming another reservoir of water inside the planet. The amount of water that can be dissolved in Earth’s mantle minerals, called its water storage capacity, generally decreases at higher temperatures. Over billion-year timescales, the exchange of water between Earth's interior and surface may control the surface oceans' volume change. Here, we calculated the water storage capacity in Earth's solid mantle as a function of mantle temperature. We find that water storage capacity in a hot, early mantle may have been smaller than the amount of water Earth's mantle currently holds, so the additional water in the mantle today would have resided on the surface of the early Earth and formed bigger oceans. Our results suggest that the long-held assumption that the surface oceans' volume remained nearly constant through geologic time may need to be reassessed.

1. Introduction

Water was delivered to the Earth during accretion or soon afterward (Morbidelli et al., 2000). This was followed by vigorous outgassing of possibly oxidized volcanogenic gases, including H$_2$O (Hirschmann, 2012), from the early Earth's mantle, which may have led to the formation of primordial oceans (Elkins-Tanton, 2011). Sometimes after the onset of plate tectonics, mantle rehydration by subduction (Iwamori, 2007; Rüpe et al., 2004; van Keken et al., 2011) began to change the relative water contents of the surface and internal reservoirs. Extensively preserved eustatic sea-level records indicate that the continental freeboard has been approximately constant throughout the Phanerozoic (541 Ma to the present) (Miller et al., 2005). However, the subaqueous and subaerial proportions of the Earth’s surface may have been quite different before the Phanerozoic, especially in the early Archean, approximately from the Eoarchean to the Paleoarchean (4–3.2 Ga). Though they are sparse, isotopic records from the early Archean may be used to indirectly constrain the size of the early
To quantify the $P$-$T$ effects on water storage capacities in major NAMs of Earth’s mantle, we compiled and parameterized high-pressure mineral physics data on water storage capacities (Tables S1–S3; Figure 1) and partition coefficients of the NAMs (Table S4). Hydrogen enters these NAMs through crystal defects and forms hydroxyl groups. The water storage capacity in a mineral under $H_2O$-saturated conditions can be expressed as (see Appendix A):

$$
\ln\left(c_{H_2O}\right) = \ln\left(\frac{1}{2n}\right) + a + \frac{n}{2} \ln f_{H_2O} + \frac{b + c \cdot P + d \cdot X_{Fe}}{T}
$$

(1)

Previous studies showed that surface water may have been continuously transported beyond the volcanic front to contribute to the deeper mantle’s possible water-rich regions (Hacker, 2008; van Keken et al., 2011). The exact value of the present-day global net water flux into the deep mantle is still under debate, especially in terms of uncertainties associated with the contribution of (nonmagmatic) updip transport of water (e.g., Bercovici & Karato, 2003) and the hydration (serpentinization) of oceanic mantle (e.g., Korenaga, 2017). The current estimates on the present-day value are on the order of $10^{11}$–$10^{12}$ kg/year (Korenaga et al., 2017; Schmidt & Poli, 2013; van Keken et al., 2011). Furthermore, the onset and efficiency of early subduction, and hence the total net water flux in the past, remains even less constrained (e.g., van Hunen & Moyen, 2012). If the recycled water flux were larger than the outgassed water flux over the age of the Earth, then possibly several tens of percent to a few times the Earth’s present-day ocean mass (OM) could have been returned into the deep mantle (van Keken et al., 2011).

Numerous geodynamical studies have investigated the coupled evolution of Earth’s interior and surface oceans in hydrous mantle convection, implying that the volume of the surface oceans evolves with time over billion-year timescales (e.g., Crowley et al., 2011; a 2008; van Keken et al., 2017; Nakagawa & Iwamori, 2019; Price et al., 2019; Sandu et al., 2011). However, most of the existing geodynamical models did not take into account the mineral physics constraints on mantle water storage; some assumed that the solid mantle is a water reservoir with infinite storage capacity. In reality, Earth’s solid mantle has a finite water storage capacity (e.g., Bercovici & Karato, 2003). Melting of mantle rocks and hence the release of water will occur when the mantle water content exceeds its storage capacity (the maximum amount of water that mantle mineral assemblages can hold structurally) near its solidus. The coevolution of water in Earth’s interior and surface reservoirs may have been further regulated by the bulk water storage capacity in the solid mantle; however, this particular perspective remains virtually unexplored.

In mineral physics, water storage capacities in major rock-forming minerals of Earth’s mantle have been studied individually by high pressure ($P$) and temperature ($T$) experiments and/or ab initio molecular dynamic simulations (Tables S1–S3). In particular, due to the strong negative effect of temperature on water storage capacities in nominally anhydrous minerals (NAMs), the mantle water storage capacity would have increased over geologic time as a result of secular planetary cooling. Although these individual water storage capacities have been measured over ranges in temperature and pressure that encompass much of the present-day and early mantle conditions, a detailed, quantitative estimate of the bulk mantle water storage capacity is not yet available. This study estimates the bulk water storage capacity in Earth’s solid mantle at different mantle potential temperatures ($T_p$) and demonstrates that the bulk solid mantle water storage capacity was smaller in the hotter mantle of the early Earth relative to that of the present-day. In addition, we apply this temperature-dependent water storage capacity model to constrain the volume of the surface oceans and explore its implications on long-term sea-level change.

2. Methods

2.1. Water Storage Capacities in Olivine, Wadsleyite, and Ringwoodite

To quantify the $P$-$T$ effects on water storage capacities in major NAMs of Earth’s mantle, we compiled and parameterized high-pressure mineral physics data on water storage capacities (Tables S1–S3; Figure 1) and partition coefficients of the NAMs (Table S4). Hydrogen enters these NAMs through crystal defects and forms hydroxyl groups. The water storage capacity in a mineral under $H_2O$-saturated conditions can be expressed as (see Appendix A):

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\ln\left(c_{H_2O}\right) = \ln\left(\frac{1}{2n}\right) + a + \frac{n}{2} \ln f_{H_2O} + \frac{b + c \cdot P + d \cdot X_{Fe}}{T}
$$

(1)
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with \( T \) in kelvin (K), \( P \) in gigapascals (GPa), \( f_{H_2O} \) (water fugacity) in GPa, and \( X_{Fe} \) (iron content) in mole fraction. The parameters \( a, b, c, d \), and \( n \) are obtained from a nonlinear robust regression to the compiled experimental data for olivine, wadsleyite, and ringwoodite. We quantified the water storage capacities in other abundant NAMs in the upper mantle and transition zone using their water partition coefficients (see Section 2.3).

In selecting experimental data to include in the fitting, we have established the following criteria: 1) At least one liquid phase has to be present in the recovered phase assemblage to ensure that the coexisting NAMs were most likely saturated with water (when the water concentration equals the water storage capacity). 2) Only water storage capacities measured by direct quantitative analytical methods (Fourier transform infrared spectroscopy [FTIR], secondary ionization mass spectrometry [SIMS], and elastic recoil detection analysis [ERDA]) were used. In contrast, those based on more qualitative estimation methods (e.g., water content estimated from lattice parameters) were excluded. More details on data selection are described in Appendix B.

2.2. Water Storage Capacity in Bridgmanite and Other Lower Mantle NAMs

Experimental constraints on water storage capacities in lower mantle NAMs are inconsistent and insufficient to fit the thermodynamic model presented above. The synthesis of large, high-quality hydrous single crystals of lower mantle phases and measurements of their water contents will likely remain technically difficult for a long time to come.

Figure 1. Water storage capacities in (a) olivine, (b) wadsleyite, and (c) ringwoodite at high pressure and temperature. The colored open circles are experimentally determined water storage capacity data from Tables S1–S3. The colored lines correspond to the fits to the data, with the regression parameters provided in Table S5. Symbol and line colors indicate water storage capacities in log ppm wt. (olivine) or wt.% (wadsleyite and ringwoodite). In (a), the black dotted lines indicate the present-day geotherm (\( T_p = 1600 \) K) and the early Archean geotherm (\( T_p = 1900 \) K). In (b), the star is a theoretical estimate of water storage capacity in wadsleyite at 1573 K and 15.5 GPa, based on a charge balance calculation (Kudoh et al., 1996). In (b and c), the dashed rectangular boxes indicate the temperature ranges of wadsleyite or ringwoodite stability in pyrolite for \( T_p = 1600 \) K and \( 1900 \) K. In (c), the natural hydrous ringwoodite inclusion containing \( \sim 1.4 \) wt.% \( H_2O \) (Pearson et al., 2014) is indicated by an arrow. Its water content corresponds to formation at up to \( \sim 1690 \) K, consistent with its cold origin in a subducting slab.
This study focuses on the temperature effect on solid mantle storage capacity, so we investigated a temperature-dependent scenario: different values for bridgmanite’s water storage capacity are calculated based on the temperature-dependent water storage capacity in ringwoodite (Figure 1c) and the experimentally determined partition coefficient between ringwoodite and bridgmanite \(D_{\text{rw/bdg}} = 15 \pm 8; \text{Inoue et al., 2010}\). The partition coefficient values used here are in good agreement with the \textit{ab initio} simulation performed by Hernández et al. (2013), where they found a partition coefficient of about 15 for the Fe-bearing system. This scenario resembles a pyrolitic mantle where ringwoodite and bridgmanite coexist. Bridgmanite makes up more than half the mass of the mantle, so uncertainties in bridgmanite’s water storage capacity are by far the most important consideration in evaluating constraints on whole-mantle storage capacity. Therefore, we performed a series of Monte Carlos simulations to examine the combined uncertainties of the thermodynamic models for olivine, wadsleyite, and ringwoodite, and the water storage capacity in bridgmanite calculated from the partition coefficient (Section 2.4). We also estimated the bulk water storage capacity associated with each experimental constraint on bridgmanite (Section 3.2) and demonstrated that our stated uncertainty range encompasses the full range of water storage capacities reported by previous experimental studies on bridgmanite (see data compilations in Table S6, based on Kaminsky, 2018 and Fu et al., 2019).

2.3. Phase Equilibria of Earth’s Solid Mantle and Its Bulk Water Storage Capacity

Using the thermodynamic code HeFESTo and the self-consistent thermodynamic data set contained therein (Stixrude & Lithgow-Bertelloni, 2011), phase proportions for the pyrolite mantle were computed along a series of geothermal adiabats at various mantle potential temperatures \(T_p\) from 1550 K to 1950 K, with consideration of the influence of phase transformations on the mantle’s thermal structure. Melting was not included in the phase equilibria computation. The simplified bulk composition of pyrolite (Workman & Hart, 2005) (in mole %) includes six chemical components: \(\text{SiO}_2\) (38.71%), \(\text{MgO}\) (49.85%), \(\text{FeO}\) (6.17%), \(\text{CaO}\) (2.94%), \(\text{Al}_2\text{O}_3\) (2.22%), and \(\text{Na}_2\text{O}\) (0.11%), which form 21 distinct mantle phases. An equilibrium phase diagram of Earth’s pyrolite mantle was computed up to 27 GPa (Figure 2), and phase assemblages above the anhydrous solidus curve (Herzberg et al., 2000) were truncated.

In calculating the Earth’s solid mantle bulk water storage capacity, the water storage capacities in individual mantle minerals at each pressure and temperature were evaluated and summed. The water storage capacity models for olivine, wadsleyite, ringwoodite, and bridgmanite are described in Sections 2.1 and 2.2. The water storage capacities in the rest of the abundant mantle silicates \(j\), including orthopyroxene, clinoxyroxene, garnet, and high-pressure clinoenstatite, were estimated based on their water partition coefficients \(D_{\text{H}_2\text{O}}^{\text{ij}}\) relative to a major silicate phase \(i\):

\[
\epsilon_{\text{H}_2\text{O}}^i = \epsilon_{\text{H}_2\text{O}}^j / D_{\text{H}_2\text{O}}^{\text{ij}}(T, P, X_{\text{Fe}}, X_{\text{Al}})
\]
The partitioning behavior of water is a function of $T$, $P$, $X_{Fe}$, and $X_{Al}$. Here, we consider temperature, pressure, and compositional dependences for the water partition coefficients of orthopyroxene, clinopyroxene, and garnet relative to olivine, which have been parameterized based on previous studies (Hauri et al., 2006; Keppler & Bolfan-Casanova, 2006):

$$
D_{ij}^{H_2O} = \frac{c_{i}^{H_2O}}{c_{j}^{H_2O}} = \frac{A_{i}}{A_{j}} \cdot \frac{f_{i}^{H_2O}}{f_{j}^{H_2O}} \cdot \exp \left( -\frac{\Delta H_{i}^{\text{bar}} - \Delta H_{j}^{\text{bar}}}{RT} \right) + \left( \Delta V_{i}^{\text{solid}} - \Delta V_{j}^{\text{solid}} \right) \cdot P \right)$$

(3)

The water storage capacities in the literature compilations (Hauri et al., 2006; Keppler & Bolfan-Casanova, 2006) used in the partition coefficient calculations were based on uncorrected experimental data (see Appendix B), but they are self-consistent and hence provide the $P-T$ dependences of the partition coefficients. Here, to obtain their water partition coefficients at different $P-T$ conditions and Al contents, we used the previously published and tabulated parameters for the water storage capacities in olivine, orthopyroxene, clinopyroxene, and garnet (Table S4) in Equation 3. The water storage capacities in orthopyroxene, clinopyroxene, and garnet used here were based on the water storage capacity in olivine and the partition coefficients calculated from Equations 2 and 3. The water partition coefficient of high-pressure clinoenstatite relative to olivine can be approximated as a constant ($D_{ol/hpcpx}^{H_2O} \approx 0.8$; Withers et al., 2007).

The total solid mantle water storage capacity was then taken as the sum of the water storage capacities in individual mantle minerals based on the relative abundances of the thermodynamically stable phases that are present at that $P-T$ condition:

$$
c_{H_2O} = \left( c_{i}^{H_2O} \right)_{i} \cdot \left( X_{i} + \sum_{j} \left( X_{j} \cdot D_{ij}^{H_2O} \right) \right)
$$

(4)

where $X_{ij}$ is the mole fractions of minerals $i$ (olivine and its polymorphs) and $j$ (other mantle minerals) in pyrolite. Water storage capacity profiles as a function of depth were quantified along a series of geotherms and integrated to produce total water storage capacities of the pyrolitic mantle as a function of mantle potential temperature.

The nominally anhydrous phase assemblage of the pyrolite mantle between 3 and 27 GPa computed from HeFESTo (Figure 2) is consistent with the experimentally determined phase relations for a CMAS-pyrolite with 2 wt.% H$_2$O (Litasov & Ohtani, 2002), where all hydrous minerals are unlikely to be stable along the geotherms of the ambient mantle, especially in the early Archean when $T_p$ was 200–300 K higher (Herzberg et al., 2010). The effects of water on melting (Hirschmann et al., 2009) and solid-state phase transformations (Frost & Dolejš, 2007), likely to be relatively small under nominally anhydrous conditions, were not considered in the computation.

2.4. Uncertainties in Mantle Water Storage Capacity From Monte Carlo Sampling

The temperature-dependent mantle water storage capacity model described above involves a number of input parameters with associated uncertainties, assumed to be normally distributed, including the experimentally determined partition coefficient between ringwoodite and bridgmanite (Inoue et al., 2010) and fitted regression parameters (Table S5).

To obtain a statistically significant estimate of mantle water storage capacity and investigate the propagation of the uncertainties associated with all parameters, we used a Monte Carlo sampling method to explore the full parameter space. Each Monte Carlo simulation is based on $10^4$ combinations of randomly sampled parameters from their normal distributions. Parameters for upper mantle and transition zone minerals are given in Table S5. For bridgmanite, $D_{rw/bdg} = 15 \pm 8$ (Inoue et al., 2010). The mantle water storage capacity reported for each potential temperature corresponds to the median of one representative set of $10^4$ simulations. Two-sided uncertainties in mantle water storage capacity at each $T_p$ were used, corresponding to the 5th and 95th percentiles of the distribution; the interquartile range (IQR) of each distribution, between the 25th and 75th
2.5. Isostatic Equilibrium and Flooding of Early Continents

The estimated water storage capacity in the early Archean mantle and the possible range of present-day mantle water contents, were then used to calculate minimum volumes of the early Earth’s oceans (Section 3.3). The implications for flooding of the continents were investigated, with the isostatic balance between continents and oceans parameterized as (Flament et al., 2008; Korenaga et al., 2017; Figure S4):

\[
\rho_{\text{w}} \cdot (h + d_0) + \rho_{\text{oc}} \cdot h_{\text{oc}} + \rho_{\text{ol}} \cdot h_{\text{ol}} + \rho_{\text{om}} \cdot h_{\text{om}} = \rho_{\text{cc}} \cdot \max(0, h) + \rho_{\text{cl}} \cdot h_{\text{cl}}
\]

where \( h \) is the sea level difference relative to the present-day value and \( d_0 \) is the depth of mid-ocean ridges. \( \rho_i \) and \( h_i \) denote the densities and thicknesses, respectively, of the various components \( i \) in the isostatic balance, namely water (\( w \)), oceanic crust (\( \text{oc} \)), depleted oceanic lithospheric mantle (\( \text{ol} \)), oceanic asthenospheric mantle (\( \text{om} \)), continental crust (\( \text{cc} \)), and continental lithospheric mantle (\( \text{cl} \)).

The present-day hypsometry curve (continental elevation relative to sea level) was approximated by fitting the ETOPO1 Global Relief Model (Amante & Eakins, 2009), where \( \sim 27.5\% \) of the present-day continents are submerged under water. The vertical scale of hypsometry was defined as a variable to describe its likely reduction due to the weaker lithosphere in the Archean (Rey & Coltice, 2008). Both cases with and without reduction in the hypsometry were tested (Section 3.4). The bathymetry curve was calculated based on Flament et al. (2008), and the details are described in Appendix C.

The densities and thicknesses of oceanic crust and depleted mantle (\( \rho_{\text{oc}}, \rho_{\text{ol}}, h_{\text{oc}}, \text{ and } h_{\text{ol}} \)) can be parameterized as functions of mantle potential temperature, based on a decompression melting model (Korenaga, 2006) in which a hotter mantle in the Archean would generate thicker oceanic crusts (\( h_{\text{oc}} \)) and depleted mantle residues (\( h_{\text{ol}} \)). The density of oceanic asthenospheric mantle (\( \rho_{\text{om}} \)) can also be parameterized as a function of mantle potential temperature (Korenaga et al., 2017):

\[
\rho_{\text{om}}(T_p) = \rho_{\text{om}}(T_p, 0) \left(1 - \alpha \cdot (T_p - T_{p,0})\right)
\]

where \( \rho_{\text{om}}(T_{p,0}) \) is 3,300 kg/m\(^3\) and the thermal expansion coefficient \( \alpha \) is \( 3.5 \times 10^{-5} \) K\(^{-1}\) (Bouhifd et al., 1996). The density and thickness of continental crust are assumed to be independent of potential temperature and are taken to be \( \rho_{\text{cc}} = 2,890 \) kg/m\(^3\) (Korenaga et al., 2017) and \( h_{\text{cc}} = 34 \) km (Huang et al., 2013), respectively. The average density of continental crust is assumed to be constant through time based on recent studies suggesting that the Archean continental crust was dominated by felsic rocks (e.g., Greber & Dauphas, 2019; Guo & Korenaga, 2020; Ptáček et al., 2020). However, the possibility of a more mafic early continental crust, which would have been denser, cannot be ruled out (e.g., Tang et al., 2016). The average Archean continental lithospheric mantle was likely highly depleted compared to its oceanic counterpart (Servali & Korenaga, 2018). To describe how buoyant the continents would have been in the early Archean, we introduced a buoyancy correction, \( \Delta \rho^A \), similar to Korenaga et al. (2017). The density \( \rho^A \) is hence expressed as \( \rho_{\text{om}}(T_p) + \Delta \rho^A \). The compensation depth is taken as the base of the continental lithosphere (200 km), which is used to determine the thicknesses of the oceanic asthenospheric mantle (\( h_{\text{om}} \)) and continental lithosphere (\( h_{\text{cl}} \)) in the model (Figure S4).

With the final topographic profile governed by oceanic and continental buoyancy, bathymetry, and hypsometry, the integral of the ocean volume over the submerged area was calculated. The convergence of sea level \( (h) \) was obtained by iteratively computing the ocean volume at various \( h \) until the desired ocean volume at the relevant mantle potential temperature \( T_p \) was reached, using the bisection method with a step size in \( h \) of \( 10^{-4} \) km. The ocean volume of the present-day Earth was benchmarked to be \( 1.45 \times 10^{21} \) kg, which agrees
with the ocean volume calculated from ETOPO1 (OM = $1.335 \times 10^{21}$ kg) (Eakins & Sharman, 2010) within <12%.

3. Results and Discussion

3.1. Parameterization of the Water Storage Capacity Model in Olivine, Wadsleyite, and Ringwoodite

The five parameters $a$, $b$, $c$, $d$, and $n$ were fitted to the data on water storage capacity in olivine (Table S1) using a robust regression with bisquare weighting of the residuals. Constants $a$, $c$, and $d$ were found to be statistically insignificant ($p$-values > 0.05; no significant compositional dependence within the range $\text{Mg#} = \frac{X_{\text{Mg}}}{X_{\text{Mg}} + X_{\text{Fe}}} \cdot 100 = 81 - 100$, and pressure affects water storage capacity only through $f_{\text{H}_2\text{O}}$), which yields the equation (Figure 1a; Table S5):

$$\ln\left(c_{\text{H}_2\text{O}}\right) = \frac{n}{2} \cdot \ln\left(f_{\text{H}_2\text{O}}\right) + \frac{b}{T}$$

(7)

The fitted value of $n$ may represent the average number of hydroxyls through multiple substitution mechanisms over the wide range of pressure and water concentration for olivine (Ferot & Bolfan-Casanova, 2012; Otsuka & Karato, 2011). Temperature dependence of the water storage capacity in olivine ($b/T$) can be attributed to the decrease in the activity of the $\text{H}_2\text{O}$ component in the aqueous fluid or silicate melt with increasing temperature (Mibe et al., 2002; Stalder et al., 2001).

Similarly, the five parameters were fitted to the data on water storage capacity in wadsleyite and ringwoodite (Tables S2 and S3), and $n$, $c$, and $d$ were found to be statistically insignificant ($p$-values > 0.05; i.e., no significant water fugacity, pressure, or compositional dependence). Therefore, with the fits of $a$ and $b$ from a robust regression (Figures 1b and 1c; Table S5), the water storage capacities in wadsleyite and ringwoodite can be expressed in a simplified form of Equation 1:

$$\ln\left(c_{\text{H}_2\text{O}}\right) = a + \frac{b}{T}$$

(8)

Under pressures and temperatures beyond upper mantle conditions (>5 GPa), aqueous fluids and silicate melts are completely miscible and become one single liquid phase (Mibe et al., 2002). Increasing miscibility between silicate melts and hydrous fluids may provide a physical explanation for the lack of statistical significance of the water fugacity term in Equation 8 (when $f_{\text{H}_2\text{O}}$ can no longer be approximated by the equation of state of pure water at these conditions), and leaves three degrees of freedom in the system MgO–SiO$_2$–H$_2$O. Even at a fixed pressure and temperature (two degrees of freedom), the water storage capacity can only be strictly defined and unique if one additional coexisting phase is present. In practice, however, measurements of the water storage capacities in wadsleyite and ringwoodite remain approximately the same regardless of the speciation, even in the presence of an additional buffering phase (Litasov et al., 2011). Therefore, experimental data in the literature may be used to fit the model (Equation 8) regardless of the experiments' buffering phases.

Since wadsleyite and ringwoodite are stable over a narrower range of pressure than olivine, no significant pressure effect was observed experimentally (Demouchy et al., 2005). Water storage capacities in Fe-bearing wadsleyite (Mg# = 87–96) were shown to be comparable to those in Fe-free wadsleyite (Litasov & Ohtani, 2008). Temperature is the only variable shown experimentally to directly affect water storage capacities in both wadsleyite and ringwoodite (Demouchy et al., 2005; Litasov & Ohtani, 2008; Ohtani et al., 2000). Although thermodynamic interpretations of the fitted parameters $n$, $a$, and $b$ remain challenging, the water storage capacity parameterizations (Equations 7 and 8; Table S5) describe the experimental observations adequately (Figure 1; Tables S1–S3).
3.2. A Comparison of Bridgmanite Water Storage Capacity Between the Temperature-Dependent Mantle Water Storage Capacity Model and Experiments

To model the temperature-dependent water storage capacity in bridgmanite, we used the partition coefficient of water between ringwoodite and bridgmanite. Both the mineral physics experiments of Inoue et al. (2010) and the first-principles simulation of Hernández et al. (2013) suggest \( D_{rw/bdg} \approx 15 \pm 8 \). In the model, this choice of \( D_{rw/bdg} \) leads to 370\((^{1240}_{180})\) ppm wt. (IQR = 270–570 ppm wt.) in bridgmanite at \( T_p = 1600 \) K and 170\((^{570}_{95})\) ppm wt. (IQR = 110–270 ppm wt.) at \( T_p = 1900 \) K. With 10 ppm wt. each in calcium silicate perovskite and ferroperriclas, the bulk water storage capacity in the lower mantle is 290\(^{1440}_{70}\) ppm wt. at \( T_p = 1600 \) K (IQR = 210–440 ppm wt.) and 130\(^{1440}_{70}\) ppm wt. (IQR = 90–210 ppm wt.) at \( T_p = 1900 \) K.

Existing measurements on the water storage capacity in bridgmanite are still in disagreement (Table S6), with quantitative reports ranging from less than a few ppm wt. (Bolfan-Casanova et al., 2003), to a few hundred ppm wt. (e.g., Meade et al., 1994; Litasov et al., 2003 [Al-free]; Inoue et al., 2010; Fu et al., 2019 [FTIR]), to 1,000–1,500 ppm wt. (Litasov et al., 2003 [Al-bearing]; Fu et al., 2019 [SIMS]), to >2,000 ppm wt. (Murakami et al., 2002).

Inoue et al. (2010), referred to hereafter as “In10,” synthesized coexisting mineral assemblages of ringwoodite with 0.63–0.76 wt.% \( \text{H}_2\text{O} \) and bridgmanite with 300–700 ppm wt. \( \text{H}_2\text{O} \) at 23–23.2 GPa and 1873 K, with a starting composition of \((\text{Mg}_{0.8}\text{Fe}_{0.2})_2\text{SiO}_4\) plus 15.8 wt.% \( \text{H}_2\text{O} \). The water contents in the ringwoodite phase have reached the storage capacity of ringwoodite (-0.68 wt.%, predicted from our ringwoodite model in Section 3.1), while a significant amount of water still remained in the bridgmanite phase; both phases are expected to have reached their storage capacities with 15.8 wt.% \( \text{H}_2\text{O} \) in the starting bulk composition. Use of the bridgmanite storage capacity based on In10 has the significant advantage of constraining the temperature dependence through use of the partition coefficient and a temperature-dependent model for ringwoodite.

Two earlier studies by Meade et al. (1994) (Me94) and Litasov et al. (2003) (Li03) reported slightly lower water storage capacities in the Mg-endmember of bridgmanite MgSiO\(_3\); in Me94, the FTIR measurements showed 105\((^{25}_{)}\) ppm wt. \( \text{H}_2\text{O} \) at 27 GPa and 2103 K; in Li03, the FTIR measurements showed 104\((^{14}_{)}\) ppm wt. \( \text{H}_2\text{O} \), while the SIMS measurements showed 300\((^{100}_{)}\) ppm wt. \( \text{H}_2\text{O} \), in bridgmanite synthesized at 25 GPa and 1500 K. Unlike other silicate phases such as olivine, wadsleyite, and ringwoodite (Tables S1–S3), there is an evident discrepancy between the FTIR and SIMS measurements for bridgmanite. The prevalence and cause of this discrepancy between the two analytical methods remain unclear. The most recent study by Fu et al. (2019) (Fu19) showed a similar discrepancy and reported a water concentration in bridgmanite of 596–731 ppm wt. (FTIR) and 1,005–1,031 ppm wt. (NanoSIMS), where (Al,Fe)-bearing bridgmanite single crystals were synthesized from a hydrous melt containing 6.7 wt.% water at 24 GPa and 2073 K. Compared with the Mg-endmember, the presence of (Al,Fe) may have slightly enhanced the water storage capacity in bridgmanite through a variety of possible substitution mechanisms (Walter et al., 2015), as similar effects have been observed in olivine (Hauri et al., 2006; Withers et al., 2011).

As discussed above, the majority of the existing measurements on water storage capacity in bridgmanite, except Murakami et al. (2002) (Mu02) and Bolfan-Casanova et al. (2003) (Bo03), are in agreement with the predicted range at \( T_p = 1600 \) K (190–1,570 ppm wt.). Bo03 reported that bridgmanite could contain only a few ppm wt., near or less than the detection limit, while Mu02 reported 1,900–2,400 ppm wt. \( \text{H}_2\text{O} \) in their (Al,Fe)-bearing samples. Both the extremely low (<10 ppm wt.) and high (>2,000 ppm wt.) water storage capacities have been suggested to result from the presence of tiny inclusions of hydrous phases in their mineral assemblages (Fu et al., 2019; Schmandt et al., 2014). In Bo03, brucite, Mg(OH)\(_2\), and ringwoodite with 0.14–0.21 wt.% water were formed along with dry bridgmanite (1–2 ppm wt.) from a bulk \( \text{H}_2\text{O} \) content of 6 wt.% that was likely undersaturated. The water content in its ringwoodite phase was well below the expected storage capacity based on our model (~1.59 wt.%), also suggesting that the sample was synthesized in water-undersaturated conditions. Brucite and ringwoodite in Bo03 may have devoured the water in the system and made the amount of water dissolved in their bridgmanite samples much smaller than the storage capacity of bridgmanite coexisting only with hydrous melt (Fu et al., 2019). In Mu02, the high water content based on FTIR may include a contribution from the brucite phase, which is associated with a sharp peak at 3,680 cm\(^{-1}\) that is present in their FTIR spectra (Fu et al., 2019; Schmandt et al., 2014). The formation of na-
noscale, intergranular inclusions of the hydrous phase remains the main challenge in synthesizing hydrous bridgmanite. As yet, only Fu19 successfully measured the water capacity in high-quality, inclusion-free (Al,Fe)-bearing bridgmanite single crystals (596–731 ppm wt. from FTIR and 1,005–1,031 ppm wt. from NanoSIMS). This measurement may be more robust than previous studies because it avoids the primary source of uncertainty arising from the presence of intergranular hydrous inclusions (Fu et al., 2019). Though no temperature dependence can be derived directly from the Fu19 study based on their bridgmanite single crystals, it does imply that the water storage capacity in bridgmanite is within the ranges of our model results for bridgmanite within the 5th and 95th percentiles (e.g., 190–1,610 ppm wt. at $T_p = 1600$ K; 75–740 ppm wt. at $T_p = 1900$ K).

We also estimated the bulk water storage capacities that Earth’s lower mantle would have based on each experimentally measured bridgmanite water storage capacity (Table S6). The top of the lower mantle would have a storage capacity of 70–110 ppm wt. and 70–280 ppm wt. based on the MgSiO$_3$ samples from Me94 and Li03, respectively; 310–680 ppm wt. and 890–1,300 ppm wt. based on (Al,Fe)-bridgmanite single crystals in Fu19 and (Al,Fe)-bridgmanite in complex assemblages in Li03, respectively; 1,250–1,740 ppm wt. based on (Al,Fe)-bridgmanite with inclusions of hydrous phases from Mu02. If the absolute water contents in bridgmanite from In10 are used, the bulk lower mantle storage capacity is 210–620 ppm wt. $H_2O$. However, some of these bridgmanite samples were synthesized at temperatures that are much lower than a realistic geotherm, so these values are, to some extent, upper bounds. Even without the Monte Carlos simulations, our model results (Figure 3) are already consistent with the majority of bulk mantle water storage capacities derived from experimental data on bridgmanite and are slightly lower than those based on the (Al,Fe)-bearing samples embedded in complex mineral assemblages reported in Mu02 and Li03. With the Monte Carlos simulations, the estimated uncertainties of our model results between the 5th and 95th percentiles (e.g., 190–1,610 ppm wt. at $T_p = 1600$ K) encompass the full range of the existing measurements of bridgmanite’s water storage capacity (Figures 4a and 4b; Table S6).
3.3. Temperature-Dependent Mantle Water Storage Capacity

We computed the mantle water storage capacity at each P-T point based on the relative abundances of minerals and their water storage capacities up to 27 GPa (Figure 2), and calculated water storage capacity profiles along the different geotherms, then integrated them throughout the entire mantle (Figure 3). The water storage capacity in an upper mantle mineral assemblage increases significantly with pressure, but at the base of the upper mantle it is still about 7 times smaller than that of the transition zone (1,210 ppm wt. at 13.5 GPa and 1800 K, vs. 8,060 ppm wt. at 14.0 GPa and 1800 K).

For the entire Earth's mantle, we obtained a storage capacity of $2.12^{+0.43}_{-0.43}$ OM (OM = $1.335 \times 10^{21}$ kg H$_2$O) at present with $T_p = 1600$ K (IQR = 2.07–2.65 OM), and 0.72$^{+0.07}_{-0.20}$ OM in the early Archean with $T_p = 1900$ K (IQR = 0.62–0.90 OM) (Figures 4, S1, S2; Table 1). While the uncertainties (5th–95th percentiles) are somewhat large due to the uncertainties in bridgmanite, the negative temperature effect is very robust because the uncertainties at different $T_p$ are highly correlated, and the median bulk water storage capacity of Earth's mantle increases significantly with de-

Table 1

| $T_p$ (K) | 5th percentile | 25th percentile | Median 50th percentile | 75th percentile | 95th percentile |
|-----------|----------------|-----------------|------------------------|----------------|----------------|
| 1550      | 2.37           | 2.62            | 2.86                   | 3.30           | 5.36           |
| 1600      | 1.86           | 2.07            | 2.29                   | 2.65           | 4.41           |
| 1650      | 1.48           | 1.66            | 1.85                   | 2.16           | 3.65           |
| 1700      | 1.19           | 1.35            | 1.52                   | 1.79           | 3.05           |
| 1750      | 1.03           | 1.18            | 1.34                   | 1.58           | 2.73           |
| 1800      | 0.83           | 0.96            | 1.10                   | 1.32           | 2.32           |
| 1850      | 0.66           | 0.78            | 0.90                   | 1.09           | 1.98           |
| 1900      | 0.52           | 0.62            | 0.72                   | 0.90           | 1.69           |
| 1950      | 0.41           | 0.50            | 0.59                   | 0.74           | 1.47           |

$^{+1}$ OM = $1.335 \times 10^{21}$ kg H$_2$O.
creasing mantle potential temperature (secular cooling). For example, a lower storage capacity in bridgemanite would decrease the calculated bulk water storage capacity at present, but it would similarly decrease the calculated bulk water storage capacity in the early Archean (Figure S3). During the early Archean, the higher temperatures would have caused significantly lower water storage capacities in most rock-forming NAMs, narrowed the width of the transition zone, and possibly caused the breakdown of wadsleyite into garnet and ferropericlase, which store much less water (Figures 2 and 3). These temperature effects result in a significantly lower water storage capacity for the mantle of the young planet: the storage capacity in an early, hotter mantle ($T_p = 1800–1900 \text{ K}, \Delta T_p = +200–300 \text{ K}$) is $1.19^{+0.16}_{-0.22} \times 10^{-2}$ OM smaller than at present.

A critical source of uncertainty is the pressure-temperature effect on the water storage capacities in lower mantle NAMs. The pressure-temperature effect on these water storage capacities, as well as their water partition coefficients above mid-mantle pressures (>30 GPa), have not been constrained experimentally. The temperature effect would likely give rise to a smaller bulk water storage capacity in the lower mantle when $T_p$ is higher, as seen in other NAMs. Conversely, we may conjecture that the storage capacities will increase at higher pressures, as this is the case for olivine and pyroxene for which have been studied over a significant range of pressures (Figures 3 and 4). Such a pressure dependence for wadsleyite and ringwoodite is not evident in their regressions, perhaps because those phases are not stable over a sufficient range of pressures. If the pressure effect surpasses the temperature effect at greater depth, our upper bound on the bulk water storage capacity may need to be revised upwards.

Another important source of uncertainty is that water storage capacities are not strictly additive for complex rocks. The pyrolite mantle is likely to have a lower storage capacity than the sum of its individual minerals, as demonstrated experimentally (e.g., Ardia et al., 2012; Ferot & Bolfan-Casanova, 2012; Tenner et al., 2012). Our current model could be improved if experiments that were conducted with complex mineral assemblages were used. Unfortunately, there is not enough literature data on the $P$-$T$ effects on the actual storage capacity of the full complex pyrolite assemblage. At some conditions, such as in the lower mantle, experiments with complex assemblages are simply not available, in which case the storage capacities in individual minerals have to be used. Thus, these calculated water storage capacities may only represent an upper bound on the storage capacity profile of the upper mantle, transition zone, and possibly lower mantle as well.

Furthermore, a variety of hydrous phases, such as phase D (Frost & Fei, 1998; Shieh et al., 1998), phase H (Nishi et al., 2014; Ohtani et al., 2014), δ-phase (Yuan et al., 2019), and possibly hydrous silica (Lin et al., 2020) as well as free aqueous fluid (Tschauner et al., 2018), carried by or derived from cold subducted slabs, may provide additional water storage in the present-day mantle. However, these phases are stable only at relatively low temperatures, so most of them may only remain thermally stable for less than 300 Ma after subduction (van der Meer et al., 2010) before slab materials become thermally equilibrated with the surrounding mantle. The additional water storage capacity from hydrous phases would also be limited by the total amount of water available in the cold subducting slab. Petrological analyses suggest that the fraction of Earth’s deep mantle comprised of recycled crust is ~20% (Sobolev et al., 2007; Xu et al., 2008). If subducted slab materials are cold enough to retain water for a maximum of 300 Ma (van der Meer et al., 2010) and subduction has been operating for ~4,000 Ma, then ~300 Ma/4,000 Ma = 7.5% of this subducted material remains cold enough to stabilize hydrous materials ($7.5\% \times 20\% = 1.5\%$ of the total deep mantle). If slabs contain an average of 2 wt.% water, with an upper bound of 1/3 of this water retained beyond the volcanic arc to be subducted (van Keken et al., 2011), this would amount to 0.3 OM stored in the present-day mantle in hydrous phases of subducted slabs. This upper bound is significantly lower than the storage capacity of NAMs in the present-day mantle and would have been considerably lower in the hotter early mantle, so their presence in the mantle today only increases the difference between the storage capacities of the early and modern Earth. Although the residence time for water in hydrous minerals (<300 Ma; van der Meer et al., 2010) is short, and the storage capacity in hydrous minerals is relatively small compared with that in NAMs, these slab-derived hydrous minerals and their breakdown reactions are key intermediates of the deep water cycle toward long-term stable water storage in the NAMs of Earth’s mantle.

In this study, we focus on the early period of the Archean Eon (4–3.2 Ga) that includes the Eoarchean (4–3.6 Ga) and the Paleoarchean (3.6–3.2 Ga). Rock records from this period are scanty or lacking; Earth’s mantle had cooled enough to remain mostly solid, and water delivery from impacts had waned, making the total
water budget in the Earth more stable. The ambient mantle \( T_p \) for the early Earth is thought to have been higher than in the present-day mantle (\( T_p = 1600 \) K), though estimates of the mantle \( T_p \) in the early Archean (4–3.2 Ga) vary from \( \Delta T_p = +200 \) to \( \Delta T_p = +300 \) K, depending on conflicting interpretations of petrological constraints (Aulbach & Arndt, 2019; Condie et al., 2016; Ganne & Feng, 2017; Herzberg, 2019; Herzberg et al., 2010). The classical heat flow scaling predicts that mantle temperature decreases monotonically but would lead to a thermal runaway if the present-day Urey ratio is adopted; alternatively, a different thermal history model presented by Korenaga and others (e.g., Herzberg et al., 2010; Korenaga, 2006) suggests that the Earth's thermal evolution switched from initially warming up to cooling down around 3 Ga. Other resolutions to the thermal runaway include different modes of tectonics or a weaker temperature effect on viscosity. All of the studies discussed above maintain, however, that the mantle potential temperature in the early Archean (4-3.2 Ga) was 1800–2000 K (\( \Delta T_p = +200–300 \) K).

### 3.4. Minimum Volume of Earth's Early Archean Oceans

The present-day mantle water content is not necessarily equal to its storage capacity. The total amount of water in the oceans plus the mantle may have remained approximately constant since the early or mid-Archean; exchange between the ocean and mantle reservoirs redistributed water over geological time (van Keken et al., 2011). If the mantle today holds more water than its water storage capacity during the early Archean (e.g. 0.72_{0.20}^{+0.97} OM at \( T_p = 1900 \) K, IQR = 0.62–0.90 OM), more voluminous surface oceans must have existed in the past. If 1 or 2 OM water are stored in the mantle today, the early Earth with a water-saturated mantle at \( T_p = 1900 \) K should have had surface oceans with a minimum volume of 1.27_{0.09}^{+0.21} OM (IQR = 1.10–1.38 OM) or 2.27_{0.20}^{+0.56} OM (IQR = 2.10–2.38 OM), respectively (Figure 5). The exact prediction depends on the amount of water in the mantle today and the early Archean mantle \( T_p \).

Alternatively, with 0.5 OM stored in the mantle today, the minimum volume of the early Archean oceans is 0.77_{0.20}^{+0.48} OM (IQR = 0.60–0.88 OM), smaller than the surface oceans today. In this particular scenario, the volume of the early oceans becomes less constrained without a detailed knowledge of the water flux. If a more voluminous Archean ocean can be excluded through an improved understanding of the geologic record in the future, this would indicate that the bulk water content of the present-day mantle is lower than its early water storage capacity 0.72_{0.20}^{+0.97} OM at \( T_p = 1900 \) K.

To place constraints on the volume of the early oceans, the bulk water content in the present-day Earth's mantle is a key piece of information. One standard method to estimate the mantle H\(_2\)O content is to use the
roughly constant $\text{H}_2\text{O}/\text{Ce}$ ratio in oceanic basalts and the Ce concentration in the bulk silicate Earth (BSE). Estimates of the actual mantle water content today based on $\text{H}_2\text{O}/\text{Ce}$ ratios in oceanic basalts typically span 0.5–2 OM (Dauphas & Morbidelli, 2014; Hirschmann, 2018). The latest estimate with a rigorous error analysis by Hirschman (2018) implies that the present-day water content is between 0.55 and 0.95 OM, which is at the lower end of this range. However, the assumption underlying this $\text{H}_2\text{O}/\text{Ce}$ method, that all geochemical reservoirs with different $\text{H}_2\text{O}/\text{Ce}$ have been sampled by the full spectrum of oceanic basalts that have been analyzed, may not hold if there exist large regions of the mantle with $\text{H}_2\text{O}/\text{Ce}$ ratios higher than the bounds constrained from a typical mid-ocean ridge basalt or ocean island basalt source. Recent studies have found some modern localities with elevated $\text{H}_2\text{O}/\text{Ce}$ ratios, such as the islands of Bermuda (Mazza et al., 2019), and suggest that some deeper water-enriched mantle reservoirs with distinct $\text{H}_2\text{O}/\text{Ce}$ may not be represented in the suite of oceanic basalts that have been analyzed previously. In particular, Bermuda may sample the water-enriched transition zone underneath it, in which wadsleyite and ringwoodite can theoretically store up to a few weight percent of water in their structures. Geophysical and geochemical constraints also suggest that the transition zone is at least locally water-enriched, based on the water concentration measured in a diamond-hosted hydrous ringwoodite inclusion (Pearson et al., 2014), as well as geophysical inferences from electrical conductivity (Kelbert et al., 2009), viscosity (Fei et al., 2017), and possibly shear wave velocities (Schulze et al., 2018; Wang et al., 2019). If modern localities like Bermuda with elevated $\text{H}_2\text{O}/\text{Ce}$ ratios represent a significant fraction of the mantle, such as the transition zone, the current best estimate based on $\text{H}_2\text{O}/\text{Ce}$ (0.55–0.95 OM) may be an underestimate. However, it is perhaps unlikely that the hundreds of oceanic basalt localities that have been studied in the literature (compared to only a few with elevated $\text{H}_2\text{O}/\text{Ce}$ ratios) somehow fail to sample the majority of the mantle. Therefore, the upper bound on the present-day mantle water content likely does not exceed 2 OM; at the same time, a present-day water content of <0.5 OM would be contradictory to observations of deep water-enriched reservoirs (e.g., Mazza et al., 2019) and the predicted global flux of $\text{H}_2\text{O}$ to the deep mantle over geological time (e.g., Korenaga et al., 2017; van Keken et al., 2011).

Some estimates of the bulk water content in the present-day Earth’s mantle span 10–50 OM from cosmochemical arguments (e.g., Abe et al., 2000; Dreibus & Wänke, 1989; Righter & Drake, 1999). The cosmochemical constraints are often based on the expected water contents of the primitive planetesimals that formed the Earth. The efficiency of water delivery and retention during accretion, however, remains highly controversial. Based on this temperature-dependent mantle water storage capacity model, the possible range for present-day mantle water storage capacity (with $T_p = 1550–1650$ K) is 1.85–2.86 OM (medians), and at most 5.36 OM (the 95th percentile at 1550 K), with the possible addition of up to 0.3 OM stored in hydrous phases. Therefore, the Earth’s solid mantle is physically unable to accommodate more than 5–6 OM today. If the total amount of water that was delivered to the Earth was tens of OM, the excess water may have been either devolatilized and lost upon impact, lost to space, or partitioned into the core during or shortly after the end of accretion (Wu et al., 2018).

Here, we conservatively consider only the minimum surface ocean volume under the assumptions that the early mantle was water-saturated and the total water budget of the mantle plus ocean was constant through time. This results in a lower bound on early ocean volume because these assumptions are unlikely to be strictly correct. Though it has been observed that some primordial water was retained in the Earth’s mantle (Hallis et al., 2015), processes associated with magma ocean solidification and overturn would have degassed the primordial mantle significantly and made its actual water content much lower than its storage capacity (e.g., Elkins-Tanton, 2008; Tikoo & Elkins-Tanton, 2017). Recent models based on noble gas constraints suggest that the early Earth may have been dominated by net degassing from the interior, with net regassing (re-hydration) not becoming prevalent until sometime later (Marty et al., 2019; Parai & Mukhopadhyay, 2018), though the exact timing of the switch is model-dependent, possibly sensitive to continental growth (Guo & Korenaga, 2020). In addition, we assumed that the total water budget of the Earth system became stable by the early or mid-Archean (Morbidelli et al., 2000; Zahnle et al., 2019) and remained approximately constant to the present, with no significant $\text{H}_2\text{O}$ loss to space. The validity of this assumption strongly depends on atmospheric composition (Wordsworth & Pierrehumbert, 2013), and the hydrogen escape rate might have been significant throughout the Archean (Zahnle et al., 2019) if the early Archean atmosphere was $\text{N}_2$ poor and/or $\text{H}_2$ rich. If the early mantle were undersaturated with water and/
or if a considerable amount of water was lost to space since that time, the early surface oceans would have been even larger than these predicted volumes. Alternatives to water having resided on the surface include possible reservoirs of water in the deep Earth, such as the core or partial melts in the mantle. However, these possibilities are less likely. The water content of the core remains controversial (e.g., Clesi et al., 2018; Hirose et al., 2019; Iizuka-Oku et al., 2017; Li et al., 2020; Okuchi et al., 1997; Terasaki et al., 2012; Yuan & Steinle-Neumann, 2020), but it is most likely not decreasing over time, making it an unlikely source of an increase in mantle water. Possible hydrous partial melts in the early mantle would have likely been too buoyant to remain gravitationally stable at depth (Mookherjee et al., 2008) and would have erupted to release their water onto the surface. To maintain neutral buoyancy of a hydrous melt layer at the top of the transition zone, even along the present-day geotherm, its water content would be limited to ~8 wt.% (Sakamaki et al., 2006). With a 10 km thick melt layer above the transition zone (Bercovici & Karato, 2003), the additional water storage capacity contributed from this stable hydrous melt layer would amount to ~1 OM. Seismic signatures of another possible melt layer at the base of the transition zone have only been reported in a few localities around the globe (e.g., Schmandt et al., 2014), so that the possible contribution from this layer should be several orders of magnitude smaller. In addition, it has been suggested that a neutrally buoyant hydrous layer could be present at the core-mantle boundary (Sakamaki et al., 2006), but there is no direct seismic evidence of this on a global scale. Even if it exists, most of its water should have had been lost to the core, which has a greater affinity for hydrogen (O’Rourke et al., 2019).

### 3.5. Implications for the Flooded Fraction of Early Continents

Besides the volume of early oceans, the nature and tempo of continental growth play a critical role in the modeling of the early Archean surface environment, which involves a number of parameters without direct observational constraints, including the nature of early continental and oceanic plates as well as surface topography (Section 2.4). In particular, the total amount of emergent continents during the early Archean could have been anywhere between ~10% and 100% (e.g., Korenaga, 2018). The parameter space of the sea level model has been explored in the earlier studies (Flament et al., 2008; Korenaga et al., 2017), without consideration of the finite mantle water storage capacity. However, it remains difficult to assess the degree to which different sets of parameters can satisfy the observational constraints on the early Archean Earth. Therefore, unlike for the volume of the early oceans, we did not perform a full uncertainty analysis on the combined effects of these parameters (e.g., Seales & Lenardic, 2020). In this study, the primary goal is to explore how our estimated volumes of the early oceans may have influenced early Archean sea level. Therefore, we present three representative cases with a continental configuration of (1) 100% of its present-day size \( f_c = 1 \) and height \( f_h = 1 \) and \( f_t = 1 \); (2) \( f_c = 0.3 \) and \( f_h = 1 \); and (3) \( f_c = 1 \) and \( f_h = 0.3 \) (Figure 6; Tables 2 and 3). For example, given the geochemical estimates of 1 OM stored in the mantle at present and an early Archean \( T_p = 1900 \) K, with (1) \( f_c = f_h = 1 \), the early continents were likely at least 92\(^{+6}\)\(^{-4}\)% flooded (IQR = 89%–94%); with (2) \( f_c = 0.3 \) and \( f_h = 1 \), at least 64\(^{+22}\)\(^{-58}\)% flooded (IQR = 33%–79%); and with (3) \( f_c = 1 \) and \( f_h = 0.3 \), 100\(^{+3}\)\(^{-10}\)0% flooded (IQR = 98%–100%). More specifically, the boundaries between less flooding and more flooding relative to present-day sea level are shown as solid black lines in Figures 6a–6i. For the early Archean Earth at \( T_p = 1900 \) K with \( f_c = f_h = 1 \), the early continents would have been more flooded than today if the present-day mantle water content is > 0.4^{+0.5}\(_{-0.5}\) OM (IQR = 0.3–0.5 OM); with \( f_h = 0.3 \), \( f_c = 1 \), \( f_t = 1 \), > 0.8^{+0.9}\(_{-0.2}\) OM (IQR = 0.7–1.0 OM); and with \( f_h = 1 \), \( f_c = 0.3 \), > 0.4^{+1.0}\(_{-0.2}\) OM (IQR = 0.2–0.5 OM).

In general, increasing the volume of early continental crust or decreasing its topography (height) increases the flooded fraction for a given \( T_p \). If the early oceans were more than twice as voluminous as in the present day with an early Archean \( T_p = 1900 \) K, the continents were possibly covered entirely by water, with only a small number of subaerial landmasses protruding above sea level due to hot spot volcanism and/or oceanic plateaus. In contrast, if we assume a present-day mantle water content of 0.5–1 OM or less, the minimum volume of the early oceans may be smaller than that of the modern ocean (Figure 6). In these scenarios, the majority of the early continents would have been above sea level. It has been argued recently (Korenaga et al., 2017) that the greater buoyancy of continental lithosphere relative to oceanic lithosphere was enhanced in the early Archean (e.g., \( \Delta\rho_{A} = -5 \) kg/m\(^3\); see Section 2.4), which suggests that having an early
Archean sea level similar to present-day requires a larger volume of surface oceans, even if early continents are 100% of their present-day size.

This model’s major caveat is that plate tectonics, with oceanic and continental plates of different densities, is assumed to have been operating during the early Archean, which may or may not have been the case (e.g., Brown et al., 2020; Cawood et al., 2018; Korenaga, 2013). Alternatively, the early crust could have been a more uniform lithosphere with a single density (e.g., Moore & Webb, 2013), which would have inhibited the positive topography in isostatic equilibrium, hence requiring a smaller ocean volume to achieve the same flooded fraction of the early Earth’s surface.

Figure 6. Minimum flooded fraction of early Archean continents as a function of $T_p$ and present-day mantle water content from isostatic modeling. Three explorative cases with an early Archean continental configuration of $f_c = 1$ and $f_t = 1$, $f_c = 0.3$ and $f_t = 1$, and $f_c = 1$ and $f_t = 0.3$ are plotted in (a–c), (d–f), and (g–i), respectively. With a greater volume of surface oceans at $T_p \geq 1900$ K for the early Archean mantle and >0.3–0.8 OM (medians) water stored in the mantle today, the early continents may have been excessively flooded. The exact fraction of submerged continents is dependent on model parameters. The black solid lines correspond to the flooded fraction of present-day continents (~27.5%), delineating the boundary between necessarily more flooding than at present and possibly less flooding.
Most of the model results suggest a largely flooded early Archean Earth (Table 2). Though extremely sparse, the geologic records from the early Archean (before 3.2 Ga) are consistent with a global surface ocean with little or no land (a “water world”). A number of recent isotope studies on Earth’s early surface environment (hydrogen isotopes: Pope et al., 2012; Kurokawa et al., 2018; oxygen isotopes: Bindeman et al., 2018; Johnson & Wing, 2020) suggest more voluminous early oceans with largely flooded continents. The stable isotope signatures are consistent with observations that almost all large igneous province volcanism was subaqueous before the Archean (e.g., Arndt, 1999; Kump et al., 2007). Additionally, Earth’s earliest thick and laterally extensive platform sedimentary successions were deposited only in the late Archean (Windley, 1995), which could not have been created without continents becoming exposed at that time.

A largely water-covered early Earth would have had a lower albedo than at present, which may have moderated surface temperatures during this period of reduced solar influx (Rosing et al., 2010) when early life may have been emerging. It might also have induced the deep hydration of primitive oceanic lithosphere via thermal cracking and subsequent serpentinization, helping to facilitate the onset of plate tectonics (Korenaga, 2007). Additionally, Earth’s earliest thick and laterally extensive platform sedimentary successions were deposited only in the late Archean (Windley, 1995), which could not have been created without continents becoming exposed at that time.

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However, interpretations for these early geologic records may not be unique. If excessively flooded early continents and deeper oceans can be excluded through an improved understanding of the geologic record in the future, then the present-day mantle water content would have to be < 0.3–0.8 OM, and most likely much less than this range since the early mantle was likely water-undersaturated.

### Table 2
Minimum Flooded Fraction of Early Archean Continents at \( T_p = 1900 \) K

| Present-day mantle water content (OM) | \( f_c^b \) | \( f_t^c \) | 5th percentile (%) | 25th percentile (%) | Median 50th percentile (%) | 75th percentile (%) | 95th percentile (%) |
|--------------------------------------|------------|------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| 0.5                                  | 1          | 1          | 84                | 74                | 54                | 19                | 0                 |
| 0.3                                  | 1          | 0.3        | 19                | 17                | 15                | 12                | 0                 |
| 1                                    | 1          | 1          | 95                | 94                | 92                | 89                | 11                |
| 0.3                                  | 1          | 0.3        | 86                | 79                | 64                | 33                | 6                 |
| 1                                    | 1          | 0.3        | 100               | 100               | 100               | 98                | 0                 |
| 1                                    | 1          | 0.3        | 99                | 99                | 99                | 98                | 93                |
| 0.3                                  | 1          | 0.3        | 98                | 97                | 97                | 96                | 70                |
| 1                                    | 0.3        | 1          | 100               | 100               | 100               | 100               | 100               |

\( a \) Results with possibly less flooded early Archean continents relative to present-day (~27.5%) are printed in italic. \( b \) \( f_c = 1 \) represents 100% of the present-day size of the continents. \( c \) \( f_t = 1 \) represents 100% of the present-day height of the continents.

### Table 3
Maximum Present-Day Mantle Water Content Permitted for Constant Freeboard \( f_c \) at \( T_p = 1900 \) K (OM)

| \( f_c^b \) | \( f_t^c \) | 5th percentile | 25th percentile | Median 50th percentile | 75th percentile | 95th percentile |
|------------|------------|----------------|-----------------|------------------------|----------------|----------------|
| 1          | 1          | 0.2            | 0.3             | 0.4                    | 0.5            | 0.9            |
| 0.3        | 1          | 0.6            | 0.7             | 0.8                    | 1.0            | 1.7            |
| 0.3        | 0.3        | 0.1            | 0.2             | 0.3                    | 0.5            | 1.3            |

\( a \) Constant continental freeboard indicates that the submerged fraction of the continents is ~27.5%. \( b \) \( f_c = 1 \) represents 100% of the present-day size of the continents. \( c \) \( f_t = 1 \) represents 100% of the present-day height of the continents.

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### 4. Conclusions

We fitted a thermodynamic model to the experimental data on water storage capacities in olivine, wadsleyite, and ringwoodite and quantified the pressure-temperature effects. We also calculated the possible water storage capacity in bridgmanite at different \( T_p \) based on the partition coefficient of water between ringwoodite and bridgmanite. Combined with the mantle phase assemblages and their mineral proportions calculated from HeFESTo, we established a temperature-dependent water storage capacity model for the Earth’s solid mantle.

The bulk water storage capacity of the Earth’s solid mantle was significantly affected by secular cooling due to its constituent minerals’ tempera-
ture-dependent storage capacities. The mantle’s water storage capacity today is 1.86–4.41 OM, with a median of 2.29 OM \((T_p = 1600 \text{ K})\). This value is consistent with the geochemical estimates of the present-day water content (0.5–2 OM). For the early Earth’s solid mantle \((T_p = 1900 \text{ K})\), the water storage capacity was only 0.52–1.69 OM, with a median of 0.72 OM. The uncertainties in our bulk water storage capacity estimates of Earth’s mantle are dominated by uncertainties in the water storage capacities of lower mantle NAMs, especially bridgmanite, which is an important target for future experimental studies.

Nevertheless, due to correlations in the uncertainties at different \(T_p\), we robustly find that the early (hotter) mantle’s water storage capacity would have been significantly lower than at present, by \(\pm 0.90 \pm 0.16\)–\(\pm 1.19 \pm 0.22\) OM. If the actual water content of today’s mantle is greater than its storage capacity was in the early Archean, then the early Archean oceans must have been larger than at present, possibly causing excessive flooding of the surface. Alternatively, if in the future an improved understanding of the early Archean geological record rules out the possibility of larger oceans, then the present-day mantle water content is constrained to be < 0.3–0.8 OM, and likely much less than this range.

Appendix A: Thermodynamic Model of Water Storage Capacities in NAMs

Water is dissolved in NAMs as hydroxyl groups (OH\(^{-}\)), and the water storage capacity is primarily controlled by the amount of point defects and at which sites they substitute with OH\(^{-}\) groups. The thermodynamics of water storage capacity can be generalized by considering the following reaction (Keppler & Bolfan-Casanova, 2006):

\[
\frac{n}{2} \text{H}_2\text{O} + \frac{n}{2} \text{O}^{2-} = \left(\text{OH}^{-}\right)_n
\]

for which the equilibrium constant is:

\[
K = \frac{a_{\text{OH}^{-}}}{f_{\text{H}_2\text{O}} n^{1/2} a_{\text{O}^{2-}}^{-n/2}}
\]

where \(a_{\text{O}^{2-}}\) and \(a_{\text{OH}^{-}}\) are the activities of the unprotonated oxygens and hydroxyls, respectively; \(f_{\text{H}_2\text{O}}\) is the water fugacity; and \(n\) is the fugacity exponent. Assuming that the unprotonated oxygens have an activity of approximately unity and remain constant throughout the reaction \((a_{\text{O}^{2-}} \sim 1)\) and that OH\(^{-}\) is a dilute species, and its activity equals its concentration \((c_{\text{OH}^{-}}) \sim n \cdot c_{\text{OH}^{-}}\), the hydroxyl concentration in NAMs can be expressed as:

\[
c_{\text{OH}^{-}} = \frac{1}{n} f_{\text{H}_2\text{O}} n^{1/2} K
\]

For a given Mg endmember mineral, the equilibrium constant \(K\) can be described by:

\[
\Delta G^* \{P, T\} = -RT \ln K = \Delta H^* - T \Delta S^* + \Delta V^*(P - P_0)
\]

where \(\Delta G^*\) is the Gibbs free energy of hydration, \(\Delta H^*\) and \(\Delta S^*\) are the hydration enthalpy and entropy, respectively, at a reference pressure \(P_0\), \(\Delta V^*\) is the volume change in the solid upon hydration, and \(R\) is the universal gas constant.

The Gibbs free energy of the concentration of hydroxyl defects (i.e., water storage capacity) in NAMs, \(\Delta G\), is a function of the solid solution composition (e.g., the mole fraction of iron, \(X_{\text{Fe}}\)). In ideal (Mg, Fe) solid solutions with minor amounts of Fe\(^{2+}\), the Gibbs free energy of hydration may be approximated as a MacLaurin series (Schmalzried, 1995; Zhao et al., 2004):
\[ \Delta G(P, T, X_{Fe}) \sim \Delta G^* (P, T) + X_{Fe} \Delta G^* (P, T) + \text{higher order terms} \]  \hspace{1cm} (A5)

where \( X_{Fe} \Delta G^* (P, T) \) represents the deviation of the Gibbs free energy of hydration from that of the Mg endmember due to the presence of iron. For a small amount of Fe, as in mantle silicates, the higher-order terms may be neglected. By combining Equations A3–A5, we can express the water concentration \( c_{OH^-} \) in moles as:

\[ c_{OH^-} = \frac{1}{n} \exp \left( \frac{\Delta S^*}{R} \right) \exp \left( -\frac{\Delta H^*}{RT} \right) \exp \left( \frac{\Delta V^* (P - P_0)}{RT} \right) \exp \left( -\Delta G^* (P, T) \frac{X_{Fe}}{RT} \right) f_{H_2O}^{n/2} \]  \hspace{1cm} (A6)

\[ c_{H_2O} = \frac{1}{2} c_{OH^-} \]  \hspace{1cm} (A7)

which, under water-saturated conditions, is equal to the water storage capacity. Equation A6 can be further simplified into Equation 1 of the main text in wt.% or ppm wt.:

\[ \ln (c_{H_2O}) = \ln \left( \frac{1}{2n} \right) + a + \frac{n}{2} \ln (f_{H_2O}) + \frac{b + c \cdot P + d \cdot X_{Fe}}{T} \]  \hspace{1cm} (A8)

where \( a, b, \) and \( c \) are constants related to the changes in entropy, enthalpy, and volume of the hydration reaction, respectively, and \( d \) is related to the compositional effect. At mantle-relevant conditions where \( P \gg P_0 \) (typically 1 bar) becomes negligible (i.e., \( P - P_0 \approx P \)). The water fugacity was calculated by parameterizing the modified Redlich-Kwong equation of state (Frost & Wood, 1997; Holland & Powell, 1991; Otsuka & Karato, 2011).

**Appendix B: Selection and Correction of Experimental Data on Water Storage Capacities in NAMs**

It is possible that one of our criteria for selecting experimental data on water storage capacity, the presence of a liquid phase, is not sufficient to ensure that water storage capacities in NAMs have been established, especially at higher pressures when the liquid phase is hydrous melt if the NAMs of interest coexisted with water-undersaturated melt. However, the water contents in the coexisting melt are often not reported in literature. Thus, we did not exclude any data set based on water content in the melt.

Moreover, for wadsleyite and ringwoodite, we excluded experiments at temperatures less than 1673 K. At lower temperatures than this, their water concentrations approach the structural limit for water storage capacity (Kudoh et al., 1996); that is, there are no/few available unprotonated oxygen atoms, which likely causes the water storage capacity to become temperature-independent and deviate from the trend predicted from the thermodynamic model (Demouchy et al., 2005).

Quantitative determination of water storage capacities in NAMs has often made use of FTIR spectroscopy. Earlier FTIR measurements were based on two empirical calibrations of the strength of frequency-dependent OH absorption bands that are generic to all water-/hydroxyl-bearing minerals (Libowitzky & Rossman, 1997; Paterson, 1982), which bear uncertainties as high as 30%–50% and are inconsistent with one another. In addition, these infrared absorption calibrations may cause unsystematic errors in water concentration determination with the use of unpolarized light, and the associated uncertainty remains difficult to assess. Several independent and more accurate infrared absorption calibrations were later established specifically for OH in olivine, including Bell et al. (2003) (cross-calibrated with \(^{15}\)N nuclear reaction analysis) and Withers et al. (2012) (cross-calibrated with ERDA). The Bell et al. (2003) calibration revises the water measurements upward based on the Paterson (1982) or Libowitzky and Rossman (1997) calibrations by a factor of 2–4, and the Withers et al. (2012) calibration gives water concentrations of \( \sim 2/3 \) of the value from
the Bell et al. (2003) calibration. To ensure the compiled water storage capacity measurements are internally consistent, we have corrected all the FTIR measurements for olivine to the Withers et al. (2012) calibration. Though no independent infrared absorption calibration has been established specifically for OH in either wadsleyite or ringwoodite, it has been shown that the Paterson (1982) calibration underestimates water concentrations in wadsleyite and ringwoodite by ∼55% relative to the ERDA technique (Bolfan-Casanova et al., 2018). Here, we corrected those water concentrations in wadsleyite and ringwoodite determined by FTIR based on the Paterson (1982) calibration by this amount.

Some water storage capacity measurements considered in this study were made using another method, SIMS, which was often calibrated against standards whose water concentrations were determined from FTIR. It has been shown that the SIMS measurements are typically in good agreement with the ERDA measurements (an absolute method for water measurement), at least for olivine (Bolfan-Casanova et al., 2018), so SIMS data in the compilation remain uncorrected. It would be ideal if the regressions were performed with the sub-data set of either FTIR measurements or SIMS measurements exclusively, to avoid any potential systematic error from mixed-use of measurements obtained by two different techniques. At the moment, both FTIR and SIMS measurements are, however, required to encompass the P-T conditions of both the present-day and early mantle. The possible errors arising from the mixed-use of the two techniques are included in the variances of the regression coefficients, which have been incorporated in the Monte Carlo simulation (Table S4).

Appendix C: Parametrization of the Earth’s Hypsometric Curve

To reconstruct the bathymetry curve (ocean floor topography from the ridge axis), a hybrid area-age distribution of the seafloor was assumed, evolving from a rectangular distribution to a triangular distribution as a linear function of χ = Aoc / Acc, where χ is the ratio of Archean (Aoc) to present-day (Acc) continental area. The age-area distribution of the ocean floor can be parametrized as (Flament et al., 2008):

\[ \frac{dA}{d\tau} = \nu \left( 1 - \frac{\tau}{\tau_{\text{max}}} \right) \]  

\[ \tau_{\text{max}} = \frac{2 \cdot A_{\text{cc}}}{2 - \chi} \cdot \nu \]  

where \( \nu \) is the global rate of seafloor spreading, \( A_{\text{oc}} \) is the surface area of Earth’s oceanic crust (\( A_{\text{oc}} = A_{\text{Earth}} - A_{\text{cc}} \)), and \( \tau \) and \( \tau_{\text{max}} \) are the age of the oceanic floor and its maximum value, respectively. It has been suggested that the observed triangular distribution of the present-day seafloor age is a deviation from the theoretical rectangular distribution due to the presence of continents (Coltice et al., 2013).

A classical half-space cooling model was used to describe the subsidence of oceanic plates, and the depth to the ocean floor from the sea level (d) can be parameterized as (Korenaga et al., 2017):

\[ d(\tau, T_p) = d_0(T_p) + b(T_p) \sqrt{\tau} \]  

\[ b(T_p) = b_0 \frac{T_p}{T_{p,0}} \]  

where the zero-age depth of mid-ocean ridges \( d_0(T_p) \) and the subsidence rate \( b(T_p) \) depend on mantle potential temperature \( T_p \) (with modern value \( T_{p,0} = 1627 \text{ K} \)), and \( b_0 = 0.323 \text{ km/yr} \) (Korenaga & Korenaga, 2008). In fact, the volumes of ocean basins (from ocean floor to sea level) are not only associated with ridge spreading rate but also influenced by dynamic topography (Gurnis, 1992). The global rate of seafloor spreading is taken as its present-day value, \( \nu = v_0 \sim 3 \text{ cm/yr} \) (Rowan & Rowley, 2017), consistent with the recent paleomagnetic evidence for modern-like plate motion velocities at 3.2 Ga (>2.50 cm/yr, Brenner et al. [2020]) as well as the scaling law for plate velocity adopted in Korenaga (2013) and Korenaga.
et al. (2017) (cf. the slower plate tectonic scenario in Korenaga et al., 2017). The bathymetry model described here remains kinematic but serves as a first-order estimation on time scales of $>10^9$ years (Conrad, 2013).

Data Availability Statement

The data sets on water storage capacities in NAMs used in the calculations are compiled from the literature, which are available within the Zenodo repository (located via https://doi.org/10.5281/zenodo.4110827), or Tables S1–S3 in the supporting information. The thermodynamic code HeFESTo used in the calculations is available on GitHub (https://github.com/stixrude/HeFESToRepository), or within the Zenodo repository (located via https://doi.org/10.5281/zenodo.4115959).

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