



**HAL**  
open science

# Mantle rain toward the Earth's surface: A model for the internal cycle of water

Denis Andrault, Nathalie Bolfan-Casanova

## ► To cite this version:

Denis Andrault, Nathalie Bolfan-Casanova. Mantle rain toward the Earth's surface: A model for the internal cycle of water. *Physics of the Earth and Planetary Interiors*, 2021, pp.106815. 10.1016/j.pepi.2021.106815 . hal-03418806

**HAL Id: hal-03418806**

**<https://uca.hal.science/hal-03418806v1>**

Submitted on 8 Nov 2021

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License

# Mantle rain towards the Earth's surface: a model for the internal cycle of water

Denis Andrault<sup>1,\*</sup> and Nathalie Bolfan-Casanova<sup>1</sup>

<sup>1</sup>Laboratoire Magmas et Volcans, CNRS-OPGC-IRD, Université Clermont Auvergne, France

\* Correspondence to denis.andrault@uca.fr

## Abstract

The internal or deep water cycle controls the volume of the oceans at the surface of the Earth. The advent of subduction 2-3 billion years ago initiated the transport of water back to the Earth's interior. With one ocean mass injected into the deep mantle over the last 2-3 billion years, some mantle regions must have become saturated and thus turned into a deep source of water. The mantle transition zone (MTZ) between 410 and 660 km depths is unlikely to be a source of hydrous melt, because its minerals can integrate several thousand ppm of water. On the contrary, the low-velocity layer (LVL) lying above the 410 km-discontinuity is one such source. As proposed by the "Transition-Zone-Water-Filter Model", the LVL is ubiquitously formed by the global uplift of the hydrous MTZ as a counter flow of subduction of slabs into deeper regions. The seismic signature of the LVL is compatible with the presence of 0.5 to 1% melt. This melt is produced by dehydration melting during upwelling of the mantle transition zone (MTZ) containing 2200(300) ppm wt H<sub>2</sub>O, which corresponds to 0.6 ocean mass stored today in the MTZ. Hydrous silicate melt can be gravitationally stable just above the 410 km discontinuity. We propose, here, that at the upper limit of the LVL it becomes buoyant, especially where the mantle is particularly hot and/or hydrous. Once it becomes buoyant, the melt can percolate rapidly upwards through the mantle. As a consequence, the olivine-bearing mantle (OBM) could be almost saturated in water, due to the presence of upwelling hydrous melt. On its path, the melt may be responsible for the seismic low-velocity zones at mantle depths of between 80 and 150 km. It could also be a source for refertilisation of the lithospheric mantle. Based on this model, there should be ~1.0 oceanic mass (OM) stored in the upper mantle today. Secular cooling of the mantle implies an increased capacity of the OBM minerals to store water. The related decrease of oceans' mass at the Earth's surface is estimated to ~20% per one billion years.

## Introduction

Water is present at the surface of our planet, which made the development of life possible. Cosmochemical arguments indicate that the amount of initial water in the planet could have been equivalent to several oceanic masses (1 OM = 1.4.10<sup>21</sup> kg H<sub>2</sub>O) depending on the source (Marty, 2012; Piani et al., 2020). Regardless of the origin of water on Earth, the deep water cycle determines how much of the accreted water resides at the surface and how much is trapped in the planet's interior. In this article, we develop a model based on the water storage capacity of mantle minerals, which brings new constraints on the distribution of water in the Earth's mantle.

Modern plate tectonics has been operating for the last 2-3 billion years (Ga), during which water has been continuously introduced into the mantle at subduction zones. This regassing flux is particularly difficult to assess, since the different layers of a slab have heterogeneous water contents and the degree of serpentinization can vary as a function of the geological setting. Recent estimates converge to 1 to 2 10<sup>12</sup> kg.yr<sup>-1</sup> of water transported to the mantle at subduction zones, which equates to 0.7 to 1.4 times the OM every Ga. Still, most of the water is thought to escape from the slab, percolate into the mantle wedge and trigger arc volcanism. Only 30% to 40% of the initial slab water could be efficiently carried down to the deep mantle, resulting in a flux of 0.3 to 0.8 10<sup>12</sup> kg.yr<sup>-1</sup> H<sub>2</sub>O or 0.2 to 0.5 OM/Ga (Rupke et al.,

2004; van Keken et al., 2011). With increasing depth, the hydrous minerals that are stable at the Earth's surface break down upon heating and their water content can eventually be incorporated as H point defects in nominally anhydrous minerals (NAMs) (Ohtani et al., 2018; Padrón-Navarta and Hermann, 2017). This evolution is facilitated by an increasing H-solubility in NAMs with increasing pressure. At depths greater than 410 km, the formation of wadsleyite and ringwoodite increases hydrogen solubility by about one order of magnitude (Bolfan-Casanova et al., 2000; Ferot and Bolfan-Casanova, 2012; Inoue, 1994; Ohtani et al., 2000). We note that a high water storage capacity does not necessarily imply a high water content. Still, the discovery of the first terrestrial ringwoodite, which contains 1.2 wt% H<sub>2</sub>O, in a deep diamond, is a proof that water can reach the transition zone (Pearson et al., 2014).

Today, there is no consensus on the water content in the mantle transition zone (MTZ) with budgets varying from 0.3 to 3 OM (corresponding to ~1000 ppm to 1 wt% H<sub>2</sub>O). A number of arguments have been proposed for or against high water contents based on different approaches: (i) elasticity measurements related to observed velocity jumps at 410 km depth invoke water contents that vary between 0.3 and 0.6 wt% H<sub>2</sub>O (Buchen et al., 2018; Mao et al., 2008). (ii) Electrical conductivity yields controversial results from negligible water contents (Yoshino et al., 2008) to 0.1-0.2 wt% (Huang et al., 2005). (iii) It has also been proposed that 1-2 wt% H<sub>2</sub>O are necessary to explain the viscosity contrast at 660 km depth, as inferred from the effect of water content on dislocation mobility in bridgmanite and ringwoodite (Fei et al., 2017). Of course, the water content of the MTZ does not have to be homogeneous. Hydrogen has a slow diffusion rate in the solid state (Demouchy, 2010; Hae et al., 2006), which can, however, be counterbalanced by a long residence time of a slab in the MTZ (Fukao and Obayashi, 2013).

A crucial geological setting for the internal cycle of water is the global low velocity layer (LVL) above the 410 km discontinuity (Revenaugh and Sipkin, 1994; Tauzin et al., 2010; Vinnik and Farra, 2007). It is unanimously interpreted as being due to dehydration melting (Revenaugh and Jordan, 1991; Song et al., 2004; Tauzin et al., 2010) and this is a strong indication that the transition zone is hydrous. Melting triggered by dehydration of the rising mantle occurs if the water content in the MTZ exceeds the water solubility of the olivine-bearing mantle (OBM). The LVL displays an average shear-wave velocity drop ( $\delta V_s/V_s$ ) of about 4-5%, as reported in previous works, compared to the overlying OBM. Based on the experimental relationship between the  $V_s$  drop in hydrous peridotites and the amount of melt produced, the melt content in the LVL was constrained between 0.5 to 1.0 vol. % (Freitas et al., 2017; Xiao et al., 2020). The Transition Zone Water Filter model (Bercovici and Karato, 2003) is used as a mean of explaining the presence and fate of the melt in the LVL. It involves a continuous production of melt in the LVL associated with a water cycle around the transition zone, functioning in a closed system with only one input: water from the slab. The hypothesis is that, once formed atop the 410 km discontinuity, the dense melt travels laterally in the LVL until it freezes in contact with the cold slab and gets subducted (e.g. (Leahy and Bercovici, 2007)). The water would be released again after crossing the 660 km boundary and would diffuse up to the MTZ because water incorporation in bridgmanite is low (Bolfan-Casanova et al., 2003; Liu et al., 2021). Seismic low velocity zones below the 660-km discontinuity indicate inefficient penetration of water into the lower mantle (Durand et al., 2017; Liu et al., 2016; Schmandt et al., 2014). However, it should be relatively difficult to subduct the LVL melt because it is lighter than the MTZ (Matsukage et al., 2005; Sakamaki et al., 2006). In addition, the sustainability of a closed deep water cycle with water entering from the slab is questionable: if 0.2 to 0.5 OM were to be injected into the deep mantle every Ga (see above), the water content in the MTZ should have increased by 0.4-1.5 OM and hence the ocean level decreased of the same amount since the beginning of plate tectonics 2-3 Ga ago. This is obviously too extreme and incompatible with previous proposals of a 10-20% decrease in ocean mass since the Archean, based on hydrogen isotopes (Lecuyer et al., 1998; Pope et al., 2012) and geodynamic modeling (Flament et al., 2013).

At the other end of the deep water cycle, mid-ocean ridges are the main recognized geological setting where water is transferred from the mantle to the crust (in this study, water outgassing by arc

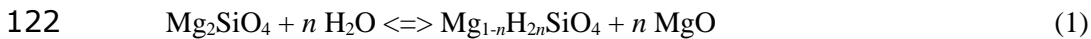
96 volcanism is not considered in the mantle budget because we only consider the flux entering the mantle  
 97 beyond the formation of arcs, which is 0.3 to 0.8  $10^{12}$  kg.yr<sup>-1</sup> H<sub>2</sub>O, as proposed by van Keken et al.,  
 98 2011). The rate of basalt production is  $\sim 20$  km<sup>3</sup>.yr<sup>-1</sup> (Cogne and Humler, 2006), corresponding to  $\sim 1.2$   
 99  $10^{11}$  kg.yr<sup>-1</sup> of water assuming 2000 ppm water in basalts (Michael, 1988). This is at least twice less  
 100 water than the lowest estimate of the water flux that is efficiently injected into the deep mantle by  
 101 subduction. The difference of at least 0.2-0.7  $10^{12}$  kg H<sub>2</sub>O.yr<sup>-1</sup> points out to the need of considering other  
 102 possible sources of outgassing.

103 In this article, the strategy is to use quantitative constraints arising from mineral physics data, such  
 104 as the water storage capacity of minerals, the effect of partial melting on seismic properties, the effect  
 105 of composition (FeO and H<sub>2</sub>O) on melt density, to develop a new model of the internal cycle of water.  
 106 We explain how the LVL can play a major role in extracting water from the major reservoir constituted  
 107 by the MTZ. The amount of water transported by the melt counterbalances the incoming water flow  
 108 associated with the entry of slabs into the MTZ, resulting in a stable water balance for the MTZ. We  
 109 investigate the conditions that can make buoyant a water-bearing melt formed in the LVL. Finally, we  
 110 describe the possible implications of such "mantle rain" and compare them with various geological  
 111 observations.

112

### 113 **Maximum water solubility of the olivine-bearing mantle**

114 The upper mantle is constituted of nominally anhydrous minerals (NAMs), which can  
 115 accommodate water in their lattice in the form of OH defects. The major upper mantle phase, olivine,  
 116 can incorporate more than 1000 ppm by weight of H<sub>2</sub>O at high upper mantle pressures. The maximum  
 117 water solubility in olivine is a function of pressure, temperature and oxygen fugacity. When the  
 118 solubility limit is overpassed, an H<sub>2</sub>O-rich phase appears, which can be a water-rich fluid at pressures  
 119 lower than a couple of GPa or a water-bearing silicate melt at higher pressures (Kessel et al., 2015; Mibe  
 120 et al., 2007). The phase equilibrium between the H<sub>2</sub>O-bearing melt and the H-saturated olivine yields  
 121 the following relations:



$$123 \quad [\text{OH}]^{\text{olivine}} = f_{\text{H}_2\text{O}}^n \exp(\Delta G^{\text{Ol}}/\text{RT}) \quad (2)$$

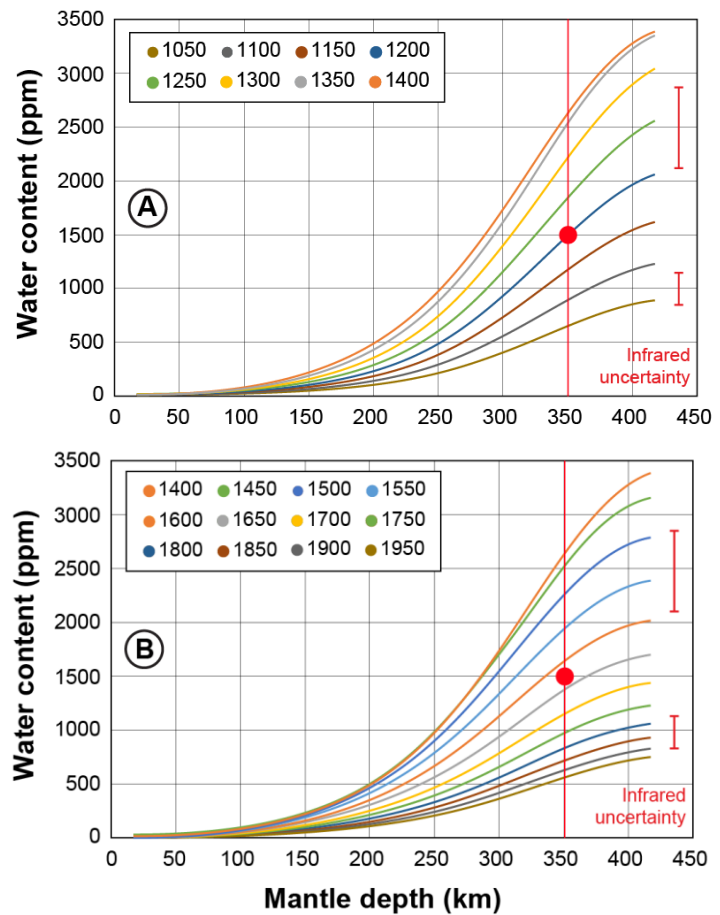
124  $f_{\text{H}_2\text{O}}$  is the water fugacity (see details in [Suppl. Material](#)) and  $\Delta G^{\text{Ol}}$  ( $\Delta H^{\text{Ol}} - T\Delta S^{\text{Ol}} - P\Delta V^{\text{Ol}}$ ) the Gibbs  
 125 free energy of the formation of H-defects in olivine,  $n$  is the exponent describing the incorporation  
 126 mechanism. A best fit to the available experimental data (see (Ferrot and Bolfan-Casanova, 2012) and  
 127 references therein) yields the following values (assuming that H is substituting for Mg, hence  $n=1$ ): (i)  
 128 formation enthalpy  $\Delta H^{\text{Ol}} = 47$  kJ/mol, which compares well with previous reports of 37.1 kJ/mol (Bali  
 129 et al., 2008) and 50 kJ/mol (Zhao et al., 2004), (ii) formation entropy  $\Delta S^{\text{Ol}}$  increasing from 75 to 92  
 130 J/molK when pressure increases from 1 to 12 GPa, as described in (Bali et al., 2008; Smyth et al., 2006),  
 131 and (iii) constant activation volume at  $\Delta V^{\text{Ol}} = 10.6$  cm<sup>3</sup>/mol (Kohlstedt et al., 1996; Mosenfelder et al.,  
 132 2006). Note that the hypothesis of site substitution of H in Mg ( $n=1$ ) or Si ( $n=2$ ) site is necessary to  
 133 extract the thermodynamic parameters from the solubility data, however, as demonstrated by (Yang,  
 134 2016) both sets of ( $n, \Delta V$ ) for  $n=1$  or  $n=2$  equally describe the data. Then, the maximum water-solubility  
 135 limit in olivine can be calculated at all P-T conditions relevant to the upper mantle using Suppl-Eq. 1  
 136 ([Suppl. Material](#)) addressing the value of  $f_{\text{H}_2\text{O}}$  in Eq. 2.

137 We can also calculate the H-content in the other mantle minerals coexisting with olivine under  
 138 water-saturation conditions, using the published partitioning coefficients of H between olivine (Ol),  
 139 enstatite (Opx), clinopyroxene (Cpx) and garnet (Gt). We adopt a H partition coefficients of 1 between  
 140 olivine and garnet  $D^{\text{water}}_{\text{ol/gt}}$  (Ardia et al., 2012; Novella et al., 2014), a  $D^{\text{water}}_{\text{opxx/ol}}$  decreasing from 2 to  
 141 0.9 between Opx and olivine with increasing pressure from 2.5 to 9 GPa (Ferrot and Bolfan-Casanova,

142 2012) and a  $D^{water}_{cpx/opx}$  of 2 between Cpx and Opx (Demouchy et al., 2017). Finally, the well-known  
 143 modal fractions of each of these phases in a peridotite-type upper mantle (Irifune and Isshiki, 1998)  
 144 enables the calculation of the maximum water-solubility limit of the bulk olivine-bearing mantle (OBM)  
 145 at any depth, following a given adiabatic temperature gradient (Fig. 1).

146 For a present-day mantle potential temperature ( $T_P$ ) of  $\sim 1650$  K (Katsura et al., 2010), the  $H_2O$   
 147 solubility limit of the OBM increases with depth from a few tens of ppm at the surface to  $\sim 1700$  ppm  
 148 wt at the 410 km seismic discontinuity. The effect of temperature is major, with a solubility limit  
 149 decreasing by a factor of  $\sim 4$  for  $T_P$  increasing from 1500 to 1950 K. It implies an increase of the water-  
 150 storage capacity of the mantle with secular cooling that we will discuss later in more detail (see also  
 151 (Dong et al., 2021)). We note that this calculation does not include possible effects associated with an  
 152 increased water solubility limit in the first 100 km of the lithosphere (Green et al., 2014; Mierdel et al.,  
 153 2007). In addition, the present model assumes conditions at which  $H_2O$  is the major hydrogen species  
 154 in the melt (Druzhbin et al., 2021).

**Fig. 1: Water-saturation limit of a peridotite-type mantle.** Colored curves present the water saturation limits of the mantle as a function of depth along an adiabatic temperature gradient prevailing for mantle potential temperatures ( $T_P$ ) from (A) 1050 to 1400 K and (B) 1400 to 1950 K (derived from (Ferot and Bolfan-Casanova, 2012)). A hydrous melt (or fluid) occurs at water contents above the saturation limit. At a given mantle depth, the saturation limit first increases with temperature up to  $T_P \sim 1400$ K and then decreases with further T increase. The latter effect is due to silicate dissolution in the melt and, therefore, a decrease in the activity of water in the hydrous silicate-melt at high temperature (see (Bali et al., 2008) and text). Uncertainty in the determination of the water content using infrared is  $\pm 15\%$  (Ferot and Bolfan-Casanova, 2012). Red dot corresponds to 1500 ppm  $H_2O$  in the mantle at 350 km depth (see text).

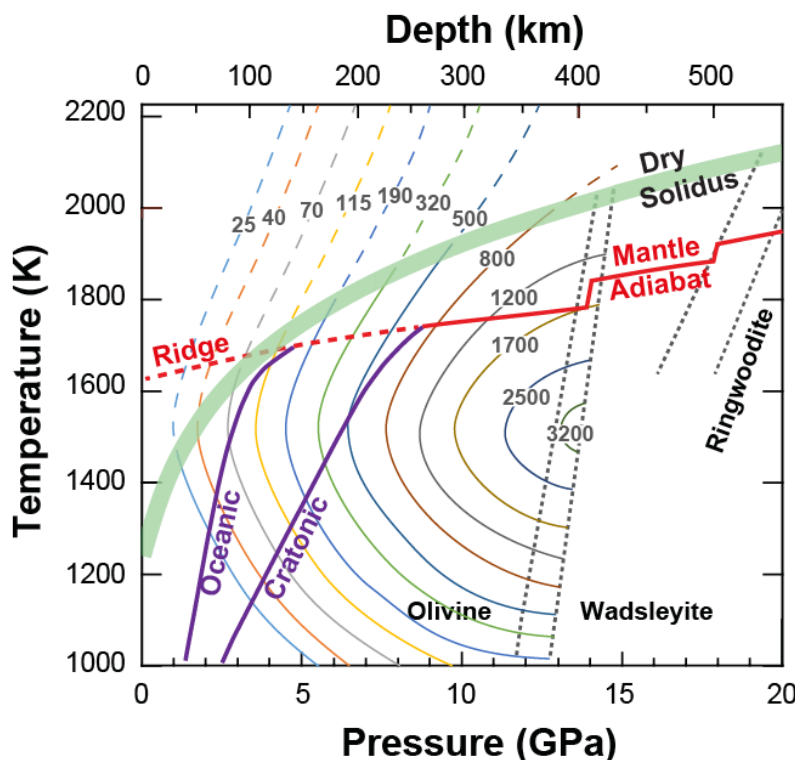


156

## 157 $H_2O$ -saturated solidus of upper mantle

158 The solubility curves of Fig. 1 can be inverted into  $H_2O$ -saturated solidii (Fig. 2), considering that  
 159 a melt appears when, at any P-T condition above the second critical point of water, the water solubility  
 160 limit of the OBM is overpassed. The “500 ppm  $H_2O$ -saturated solidus”, for instance, is the P-T profile  
 161 at which a water content above 500 ppm produces an incipient amount of melt saturated in  $H_2O$ . Due to  
 162 the formalism of Eq. 2 (see Suppl. Material), the calculated  $H_2O$ -saturated solidii present a complex  
 163 shape in the P-T diagram. In particular, they do not fall parallel to the dry-solidus (here we use that from

164 (Andrault et al., 2018), but the same observation would also stand for other works). Reasons for this are  
 165 intrinsic to the thermodynamical properties of the minerals and hydrous melt. If we consider, for  
 166 example, 1500 ppm of water in the mantle at 350 km depth (red dot in Fig. 1), such water content is  
 167 higher than the water saturation limit, and would therefore yield the formation of a aqueous fluid, at  
 168 temperatures below 1200 K (Fig. 1A) and hydrous melt above 1625 K (Fig. 1B). In contrast, the NAMs  
 169 integrate 1500 ppm in the temperature range between 1200 and 1625 K. This yields a loop shape H<sub>2</sub>O-  
 170 saturated solidus in Fig. 2. We note that the effect of water on the  
 171 thermodynamic parameters of the solid and melt may also induce  
 172 mantle melting at temperatures just below the dry-solidus (Aubaud  
 173 et al., 2004). In this case, however, neither the melt nor the solid  
 174 residue would be saturated in H<sub>2</sub>O. We do not report this effect in  
 175 Fig. 2.



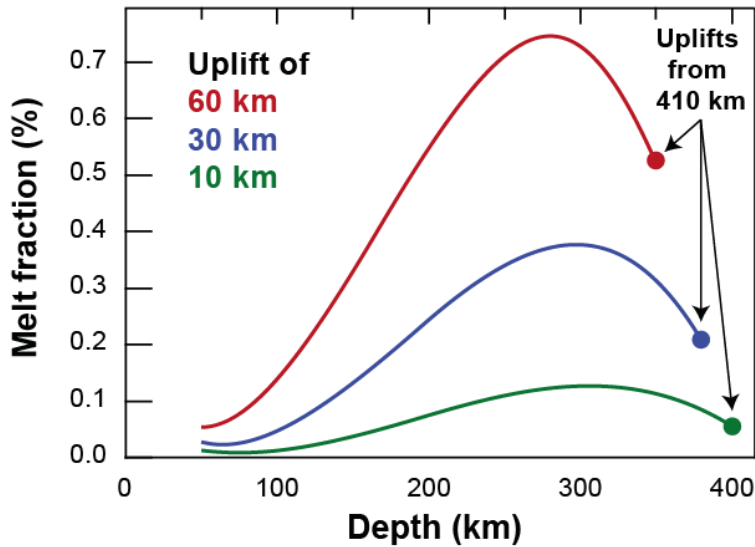
**Fig. 2: The H<sub>2</sub>O-saturated mantle solidii** for water contents between 25 and 3200 ppm H<sub>2</sub>O. The colored lines with associated water contents arise from the maximum water solubility displayed in Fig. 1. An H<sub>2</sub>O-saturated melt appears on the left side of these curves (because water solubility decreases upon decompression) for H<sub>2</sub>O-contents equal or superior to the reported number. We also report (i) the dry-solidus of chondritic-type mantle (green thick line (Andrault et al., 2018)) and (ii) the adiabatic temperature profile corresponding to  $T_P=1600-1650$  K (Katsura et al., 2010). This figure does not consider the possible effect of a higher H<sub>2</sub>O solubility limit in the lithosphere (Green et al., 2014; Mierdel et al., 2007; Sarafian et al., 2017). Uncertainty on water contents as in Fig. 1.

176

### 177 Continuous dehydration melting in the uprising mantle

178 A water content higher than the maximum water solubility induces the coexistence of water-  
 179 saturated melt and solid residue. At a depth of 410 km, the OBM residue (produced by dehydration  
 180 melting) should contain about 1700(200) ppm-wt H<sub>2</sub>O (Fig. 2). This is much higher than the water  
 181 content in the shallow source of mid ocean-ridge basalts (MORB), which contains between 50 and 250  
 182 ppm-wt H<sub>2</sub>O at around 100 km depth (Michael, 1988). An appealing option to solve this discrepancy is  
 183 that, during upwelling, the mantle H<sub>2</sub>O-content decreases along with the water storage capacity limit  
 184 (Ferot and Bolfan-Casanova, 2012) (Fig. 1). Hence, the mantle would remain partially molten at all  
 185 depths (Hirschmann et al., 2005), until, at shallow depths, it finally reaches the value invoked for the  
 186 MORB source. This seems incompatible with electrical conductivity measurements that do not favor a  
 187 partially molten upper mantle between 180 and 350 km depth (Freitas et al., 2019; Toffelmier and  
 188 Tyburczy, 2007). At least it implies that the fraction of melt should remain small. Based on the water-  
 189 saturation profiles (Fig. 1), we calculate that the uprising by e.g. 10, 30 or 60 km from 410-km depth of  
 190 the water-saturated mantle residue would produce ~0.07, ~0.25 or ~0.58 % of melt (colored dots in Fig.  
 191 3), respectively, assuming that the hydrous melt contains 6.5 wt% of water (see Table 1 and text below).

192 The same calculation can be performed for the uplift of the mantle residue from any mantle depth. The  
 193 maximum amount of melt produced by an uplift of e.g. 30 km is ~0.37 wt%. This calculation shows that  
 194 the amount of melt can be very small at any mantle depth if the melt that is regularly produced by  
 195 dehydration melting is rapidly segregated from the uprising mantle. For a comparison, the seismic and  
 196 electrical-conductivity anomalies observed between 80 and 150 km depth are compatible with more than  
 197 0.2% (Chantel et al., 2016) and between 0.3 to 2.0 wt% (Ni et al.,  
 198 2011) of melt, respectively. At intermediate upper-mantle depths,  
 199 the melt is buoyant and can travel upwards toward the Earth's  
 200 surface. This could make its presence undetectable to  
 201 geophysical measurements.



**Fig. 3: Dehydration melting in a rising upper mantle:** the uplift (i.e. adiabatic decompression) of a water-saturated residue induces continuous dehydration melting, due to a decrease of the water-saturation limit with decreasing mantle depth (Fig. 1). Here, we report the fraction of melt that can be expected at a given depth when a water-saturated mantle residue comes from 10, 30 or 60 km deeper, without loss of water during its ascent. Uplift from the 410 km seismic discontinuity are the colored dots. The curved shape with maxima around 275-300 km depth is due to the complex shape of the maximum water solubility limit in the mantle (Fig. 1). Uncertainties are +/- 15%, same as in Fig. 1.

202

### 203 Dehydration melting in the LVL: melt composition and phase diagram

204 The occurrence of a global low velocity layer (LVL) above the 410 km discontinuity (Revenaugh  
 205 and Sipkin, 1994; Tauzin et al., 2010; Vinnik and Farra, 2007) is unanimously interpreted as being due  
 206 to dehydration melting (Revenaugh and Jordan, 1991; Song et al., 2004; Tauzin et al., 2010). This is a  
 207 strong indication that the transition zone is hydrous. Melting triggered by dehydration of the rising  
 208 mantle occurs if the water content in the MTZ exceeds the water solubility of the olivine-bearing mantle  
 209 (OBM). The LVL displays an average shear-wave velocity drop ( $\delta V_s/V_s$ ) between 4 and 5.6 %, as  
 210 reported in previous works, compared to the overlying OBM. Based on the experimental relationship  
 211 between the  $V_s$  drop in hydrous peridotite and the amount of melt produced, the melt content in the LVL  
 212 was constrained between 0.5 to 1.0 vol. % (Freitas et al., 2017; Xiao et al., 2020). Dihedral angles are  
 213 much lower (less than  $10^\circ$ ) in the hydrous peridotite system at high-pressure (Freitas et al., 2017;  
 214 Yoshino et al., 2007) and the amount of melt necessary to explain  $\delta V_s/V_s$  is predicted to decrease at  
 215 lower wetting angle (Hier-Majumder et al., 2014). Low wetting angles and low surface tension (Hier-  
 216 Majumder et al., 2006; Yoshino et al., 2007) favor the fast mobility of the melt at grain boundaries. This  
 217 is compatible with another major seismic observation in the LVL; no major melt ponds but instead a  
 218 continuous global layer of partial melt that is not limited to regions associated with subduction or mantle  
 219 plumes (Tauzin et al., 2010).

220 The composition of the water-bearing LVL-melt can be studied experimentally by reproducing an  
 221 equilibrium texture between peridotite and 0.5 to 1 wt% melt. Analyses of thin veins of melt  
 222 corresponding to ~2% partial melting indicate ~21.2 wt% FeO (recalculated after (Freitas et al., 2017)).  
 223 However, the composition of liquids originating from low degrees of partial melting can be biased by

224 crystallization upon quenching, inducing an enrichment of incompatible elements in the residual melt.  
 225 **Alternative** methods have been developed in order to determine the composition of incipient melts  
 226 accurately, such as the iterative sandwich method (Dasgupta and Hirschmann, 2007), which yields a  
 227 melt FeO-content of ~13.4 wt% for the LVL melt (Condamine and Frost, 2018). On the other hand,  
 228 density measurements show variations of +21 and -16.5 kg/m<sup>3</sup> for every additional mol% FeO and wt%  
 229 H<sub>2</sub>O, respectively (Matsukage et al., 2005), in agreement with thermodynamical modeling (Freitas et  
 230 al., 2017). As a result, a melt with an FeO-content of 11.6 wt% (used in (Matsukage et al., 2005)) or  
 231 13.4 wt% (ref. (Condamine and Frost, 2018), **Table 1**) would be neutrally buoyant at 350 km, at the top  
 232 of the LVL, for H<sub>2</sub>O contents of ~6 or ~7.1 wt% H<sub>2</sub>O, respectively, in good agreement with previous  
 233 work (Matsukage et al., 2005; Sakamaki et al., 2006). For modelling purposes, we use an average LVL-  
 234 melt with reference FeO and H<sub>2</sub>O contents of 12.5(1.0) and 6.5(1.0) wt %, respectively. We note that  
 235 such FeO-content in the LVL of a peridotite-type mantle implies a solid-melt FeO partition coefficient  
 236  $D_{\text{FeO}} \sim 0.65$ , which is compatible with  $D_{\text{Fe}} \sim 0.56$  proposed previously (Mibe et al., 2006).

Composition of LVL melts		SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	MgO	CaO	Na <sub>2</sub> O	H <sub>2</sub> O	Total
Matsukage et al. (2005)	meas. (wt%)	35.5		3.3	11.6	30.4	14.1		6.0	100.9
	norm. (wt%)	35.2		3.3	11.5	30.1	14.0		5.9	100.0
	mol%	27.8		1.5	7.6	35.5	11.8		15.7	100.0
Condamine and Frost (2018)	meas. (wt%)	34.9	0.6	0.7	12.9	27.0	12.5	0.45	7.1	96.1
	norm. (wt%)	36.3	0.6	0.7	13.4	28.1	13.0	0.47	7.4	100.0
	mol%	28.1	0.4	0.3	8.7	32.4	10.8	0.35	19.0	100.0
Freitas et al. (2017) F=2%	meas. (wt%)	29.7		2.6	21.2	2.6	9.0	0.07	13.6	78.8
	norm. (wt%)	37.8		3.2	27.0	3.3	11.4	0.09	17.3	100.0
	mol%	27.6		1.4	16.4	3.6	8.9	0.06	42.0	100.0
Preferred values	wt%				12.5				6.5	

**Table 1: Composition of the LVL melt:** (Matsukage et al., 2005) reported the neutral buoyancy in the LVL of a melt containing 11.6 wt% FeO and 6.0 wt% H<sub>2</sub>O. The concentration in major elements of the LVL-melt was further refined using the iterative sandwich method (Condamine and Frost, 2018). They refined a melt Fe-content of ~13.4 wt%, which should be neutrally buoyant in the LVL for an H<sub>2</sub>O content of ~7.1 wt% (Freitas et al., 2017; Matsukage et al., 2005). In a later work, incipient melting was induced by dehydration melting while measuring the sound wave velocities under conditions relevant to the LVL (Freitas et al., 2017). In these experiments, however, crystallization of some olivine could have occurred upon quenching, making the primary melt composition difficult to retrieve. We calculate a solid-melt FeO partitioning coefficient  $D_{\text{FeO}} \sim 0.65$  between the LVL melt and the solid mantle residue.

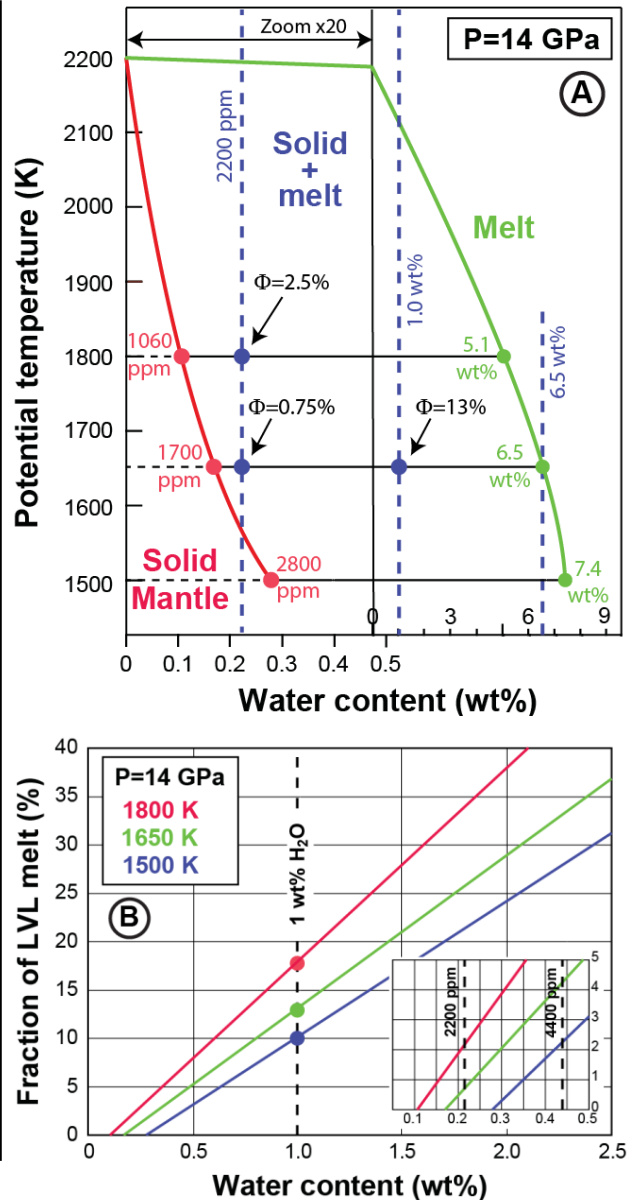
237 We now draw the melting diagram of the hydrous mantle at 14 GPa (**Fig. 4A**). The solidus is the  
 238 water storage capacity based on mineral properties (**Fig. 1**). At temperatures of 1500, 1650 and 1800 K,  
 239 the maximum H<sub>2</sub>O-content in the mantle is 2800(300), 1700(200) and 1060(100) K, respectively. For a  
 240 mantle potential temperature ( $T_p$ ) of 1650 K, the hydrous melt that is in equilibrium with the water-  
 241 saturated mantle should contain ~6.5 wt% H<sub>2</sub>O (Table 1). Using mass conservation we calculate the  
 242 melt fraction in the LVL as a function of the bulk water content (**Fig. 4B**). A bulk-mantle water content  
 243 of 2200 ppm (left vertical dashed line), 0.5 or 1 wt% (middle vertical dashed line) leads to the formation  
 244 of 0.75, 5.2 or 13 wt% of melt in the LVL. The latter values are well above the 0.5-1% of melt required  
 245 to account for low LVL velocities (Freitas et al., 2017), clearly indicating that the bulk water content of  
 246 the LVL, and that of the MTZ as well, must be well below 1 wt%. One may argue that higher MTZ  
 247 water-content could be possible, if the LVL-melt returns quickly to the MTZ (Leahy and Bercovici,  
 248 2007). However, (i) the LVL-melt is lighter than the MTZ (**Fig. 5**), with therefore no reason to sink  
 249 through the 410-km discontinuity, and (ii) return of hydrous LVL-melt into the MTZ would indefinitely  
 250 increase its water content, which does not seem sustainable. A higher water content in the MTZ is only



251 possible if the water-content in the LVL-melt has been underestimated in previous studies (Table 1).  
 252 We will discuss the fate of the LVL-melt later in this article.

253 Finally, we draw the liquidus of this phase diagram passing through the melt composition at  
 254  $T_P=1650$  K ( $\sim 6.5$  wt%) and assuming that it has the same, but inversed, curvature than that of the solidus  
 255 (Fig. 5a). The resulting diagram suggests a slight decrease of the water content in the hydrous melt, from  
 256 e.g. 7.4 to 5.1, when the potential temperature increases from 1500 to 1800 K, respectively. This  
 257 indicates that for 2200 ppm of water in the LVL, the melt fraction,  $\Phi$ , increases from 0,  $\sim 0.75$  and  $\sim 2.3$   
 258 %, when temperature is 1500, 1650 and 1800 K, respectively.

**Fig. 4:** (A) Phase diagram relevant to dehydration melting of the mantle: above the 410 km depth discontinuity ( $\sim 14$  GPa), exceeding the maximum water storage capacity of peridotite induces dehydration melting. At a mantle potential temperature ( $T_P$ ) of 1650 K, the solid mantle can store a maximum of  $\sim 1700$  ppm  $H_2O$  (Fig. 1) and the melt contains  $\sim 6.5$  wt%  $H_2O$  (Table 1). It results that a bulk water content of 2200 ppm (left vertical dashed line) leads to the formation of 0.75% of melt, in agreement with seismic observations of the low-velocity layer (Freitas et al., 2017). Thus, the melting diagram is mainly built using constraints from previous reports. Alternatively, a bulk water content of 1 wt% (middle vertical dashed line) would yield 13% melt. At  $T_P=1800$  K, a bulk water content of 2200 ppm would produce 2.5% of melt containing 5.1 wt%  $H_2O$ . This diagram shows that mantle heterogeneities of temperature and water-content affect significantly the degree of partial melting and the melt composition. Uncertainty on water contents is same as in Fig. 1. (B) Fraction of LVL-melt: based on the solidus and liquidus profiles, we calculate, from Fig. 4A, the fraction of melt that must be present in the LVL based on bulk mantle water content and for  $T_P$  of 1500, 1650 and 1800 K. Inset is a zoom of the melt fraction at low water contents. Uncertainty on the melt fraction is estimated to  $\sim 25\%$ .

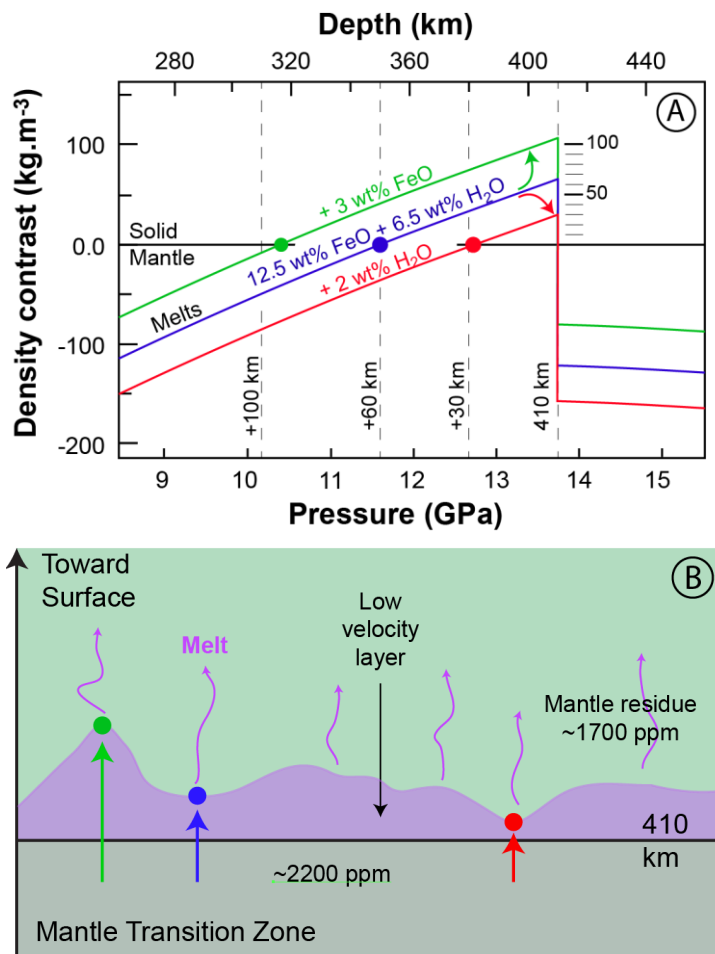


259

## 260 Melt-Mantle density contrast

261 The fate of the LVL-melt is primarily controlled by the melt-mantle density contrast ( $\Delta\rho = \rho_{\text{melt}} -$   
 262  $\rho_{\text{mantle}}$ ). A melt-mantle density crossover (that is where both densities of solid and melt become equal),  
 263 occurs around the 410 km depth corresponding to  $\sim 14$  GPa (Matsukage et al., 2005; Sakamaki et al.,  
 264 2006) (Fig. 5a), due to (i) the higher compressibility of the melt, compared to the solid mantle, (ii) the  
 265 absence of a major phase transition in mantle minerals above the 410 km discontinuity, and (iii) the

266 preference of FeO for the melt. Based on the equations of  
 267 state and the dependence of the melt density on its FeO and  
 268 H<sub>2</sub>O contents (Jing and Karato, 2012; Matsukage et al.,  
 269 2005), we recalculate the density contrast as a function of  
 270 depth for various possible melt compositions (Fig. 5a). As  
 271 expected, there is a maximum in the density contrast just  
 272 above the 410 km discontinuity that then decreases with  
 273 decreasing depth. In addition, we calculate that small  
 274 variations in the FeO (+/- 3 wt%) and H<sub>2</sub>O (+/- 2 wt%)  
 275 contents can significantly affect the mantle depth at which  
 276 neutral melt-mantle buoyancy is achieved.



**Fig. 5: (A) Melt-Mantle density cross-over.** Because melts have a higher compressibility compared to the solid mantle, the density difference between melt and solid increases with increasing depth, and the melt eventually becomes denser than the solid between the neutral buoyancy depth and the 410 km depth, at the olivine-wadsleyite phase transition. This depth interval ( $\Delta Z_{LVL}$ ) defines the thickness of the LVL. Neutral melt-mantle buoyancy is expected at the upper limit of the LVL, which is seismically observed 60 km in average above the 410 km discontinuity (blue dot). For a mantle of peridotite composition, the LVL-melt should contain 12.5 wt% FeO and 6.5 wt% H<sub>2</sub>O (blue line, Table 1). The melt density changes significantly as a function of its FeO (green line) and H<sub>2</sub>O (red line) contents (Matsukage et al., 2005; Sakamaki et al., 2006). Thus, the thickness of the region where the melt is denser (within  $\Delta Z_{LVL}$ ), can vary laterally inside the mantle due to heterogeneities in temperature and composition (see text). Depending on their composition, melts can be buoyant (less dense) at depths shallower than the green, blue or red dot. For comparison, density of the OBM at 410 km depth is ~3500 kg.m<sup>-3</sup>. Uncertainties are detailed in previous works (Matsukage et al., 2005; Sakamaki et al., 2006) **(B) The "mantle-rain" model** proposes that the LVL-melts rise upward through the mantle once they reach the upper limit of the LVL. Melts migration occurs vertically and laterally along grain boundaries. The observed regional variations of the LVL-thickness can be explained by mantle heterogeneities in FeO, H<sub>2</sub>O contents and temperature, which induce LVL-melts that are relatively denser (green dot) or lighter (red dot), compared to the typical LVL-melt (blue dot, Table 1). Still, any melt composition becomes buoyant above a given mantle depths (Fig. 5a).

277 Thus, variations in melt composition can create  
 278 variations of the thickness of the LVL ( $\Delta Z_{LVL}$ ). Indeed, the  
 279 top of the LVL is the point of neutral melt-mantle density  
 280 (red, blue and green dots in Fig. 5a) and further uprising of  
 281 the mantle beyond this limit yields a buoyant melt, which  
 282 upward migration is forced by gravity. A melt composition  
 283 enriched in iron, by e.g. 3 wt%, yields a denser melt that is  
 284 gravitationally stable across a broader region (from 410 to  
 285 ~320 km depth, up to the green dot in Fig. 5a), thus implying  
 286 an LVL which is thicker, by e.g. +40 km, compared to the  
 287 mean mantle, which melt is gravitationally stable up to ~60  
 288 km above the 410 discontinuity (blue dot in Fig. 5a). In

289 contrast, a melt enriched in H<sub>2</sub>O, by e.g. 2 wt%, yields a lighter melt that is gravitationally stable up to  
290 30 km only above the 410 km discontinuity (red dot in Fig. 5a). Chemical variations of the melt can be  
291 induced by mantle heterogeneities in temperature, water content or even fertility. They can induce  
292 variations of the LVL thickness from ~30 to ~100 km that are compatible with seismological  
293 observations (Revenaugh and Sipkin, 1994; Tauzin et al., 2010; Vinnik and Farra, 2007).

294 Knowing the melting diagram of hydrous peridotite (Fig. 4a), the manner in which the variation in  
295 mantle composition and temperature affect the density of the LVL melt can be assessed by constraining  
296 the melt fraction (Fig. 4b). Indeed, given that the 410 km discontinuity is well explained globally by a  
297 peridotite (or pyrolite) mantle, there is no reason at this stage to invoke other components such as  
298 eclogite or sediments.

299 (i) Since water is an incompatible compound, a local increase of the water content in the MTZ  
300 would induce a higher degree of partial melting in the LVL (Fig. 4b). We determine that twice more  
301 water in the MTZ than the value of 2200 ppm would produce ~6 times more LVL-melt (~4.4 wt%, see  
302 inset of Fig. 4b). Such melt would contain the same amount of H<sub>2</sub>O (fixed at the liquidus, Fig. 4a), but  
303 a lower FeO-content (reduced by ~1% compared to 6.5 wt%; Table 1), according to mass conservation  
304  $[\text{FeO}]_{\text{Melt}} = [\text{FeO}]_{\text{Mantle}} / (\Phi + (1 - \Phi)D_{\text{FeO}})$ , where  $\Phi$  is the melt fraction and using  $D_{\text{FeO}} \sim 0.65$ , the solid-melt  
305 FeO partition coefficient. As a result, such melt would be ~0.2% lighter (+21 kg/m<sup>3</sup> for every additional  
306 mol% FeO (Matsukage et al., 2005)), compared to the melt formed from a mantle containing 2200 ppm  
307 H<sub>2</sub>O. Similarly, a bulk 1 wt% H<sub>2</sub>O would produce 13 wt% of LVL-melt (see Fig. 4b) with ~5% less FeO  
308 and a density lowered by ~1%. The mantle residue also loses some Fe with increasing  $\Phi$ , however, to a  
309 much lesser extent. Altogether, bulk water contents of 4400 ppm or 1 wt% imply a decrease of ~6.3 and  
310 ~36 kg.m<sup>-3</sup>, respectively, from a melt-mantle density contrast of ~65 kg.m<sup>-3</sup> at the base of the LVL for a  
311 water content of 2200 ppm H<sub>2</sub>O (Fig 5a). This would yield a LVL up to ~30 km thinner, compared to  
312 the average mantle.

313 (ii) The mantle could also present variations in its fertility, implying significant heterogeneities in  
314 FeO-content, which incompatibility and molecular weight have important consequences on melt density,  
315 as we have seen. A reduction by 10% (from e.g. 8.0 to 7.2 wt% FeO) in the FeO content of the mantle  
316 would result in a stronger decrease of the melt FeO content, compared to that of the residue (due to  
317  $D_{\text{Fe}}^{\text{Solid/Melt}} = 0.65$ ), and a decrease of the melt-mantle density contrast by 0.8% or ~28 kg.m<sup>-3</sup> (Matsukage  
318 et al., 2005). This would yield a LVL ~30 km thicker, compared to the average mantle.

319 (iii) A locally higher mantle temperature would increase  $\Phi$  (Fig. 4b). Such LVL-melt would be  
320 depleted in both H<sub>2</sub>O and FeO, compared to mean mantle conditions as incompatible elements get  
321 diluted with increasing melt fraction. It is difficult to tell which of the two opposite effects (increasing  
322 FeO content or increasing H<sub>2</sub>O content) dominates the melt-mantle density contrast. The imprecise  
323 shape of the liquidus in Fig. 4a yields a large uncertainty on  $\Phi$  as a function of temperature. However,  
324 the effect of bulk water content could be significant, as illustrated by the change from 0.75 to 2.5 wt%  
325 of melt when  $T_P$  increases from 1650 to 1800 K, for a bulk H<sub>2</sub>O-content of 2200 ppm (Fig. 4b).

326 (iv) Finally, the presence of carbonate in the LVL melt cannot be discarded. Carbonates are  
327 subducted to great depths and are known to react to form diamond in the transition zone (Pearson et al.,  
328 2014; Shirey et al., 2021). If present in the melt, C in the form of CO<sub>3</sub><sup>2-</sup> is expected to increase the melt  
329 buoyancy relative to the surrounding mantle. Still, carbonates may more likely be present at depths  
330 shallower than ~150 km where the oxygen fugacity is higher (Stagno et al., 2013).

331 Overall, mantle heterogeneities in FeO and H<sub>2</sub>O contents and possibly also temperature and CO<sub>2</sub>-  
332 content appear to have a significant effect on the melt buoyancy, especially in the shallow part of the  
333 LVL where the melt-mantle density contrast is small (Fig. 5a). We acknowledge important uncertainties  
334 in the amplitude of the melt-density dependence on FeO, H<sub>2</sub>O, CO<sub>2</sub> contents and temperature. However,

335 the trends described above are qualitatively correct and, therefore, mantle heterogeneities could be a  
336 major controlling parameter of the LVL-thickness.

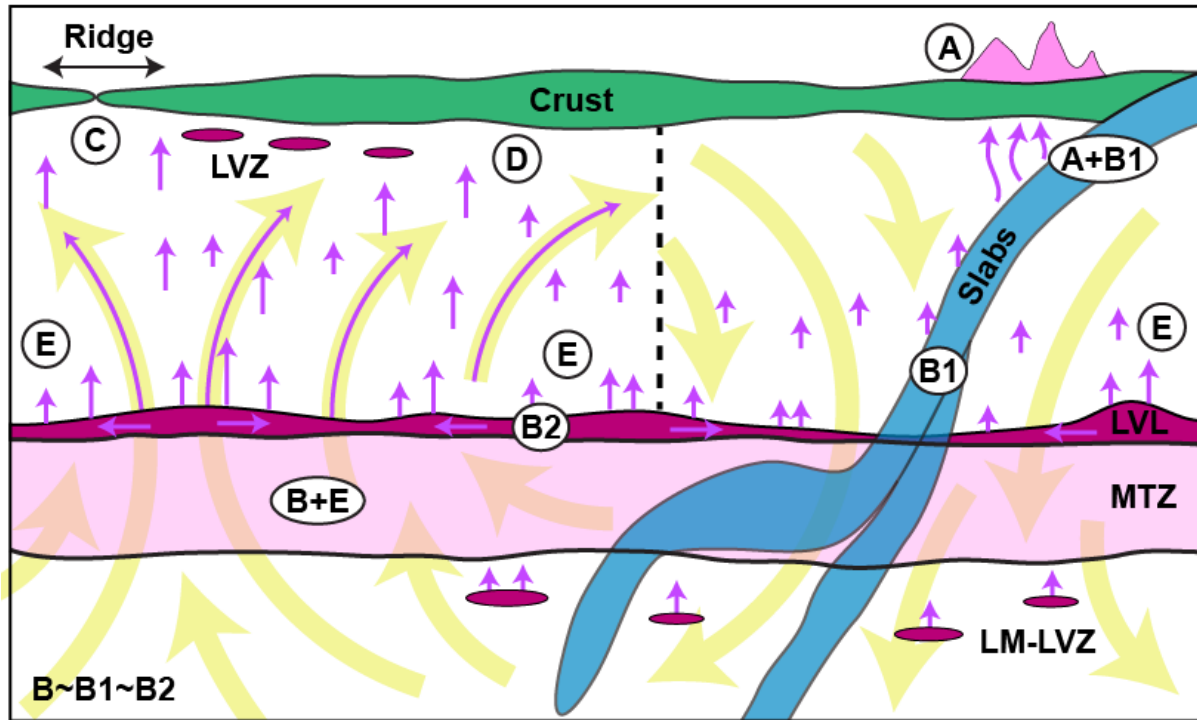
337

### 338 **Fate of hydrous-melt in the upper mantle**

339 Overall, we expect the production of water-rich melts to be widespread in the OBM: not only the  
340 LVL could be a continuous source of buoyant melts, but the uprising residual mantle is expected to  
341 dehydrate progressively on its way toward the surface (Fig. 3). While the vertical mobility of the melt  
342 is certainly triggered by a melt being more buoyant with increasing elevation from the 410 km  
343 discontinuity (Fig. 5a), lateral migration is also facilitated by several parameters: (i) a low dihedral angle  
344 in the hydrous peridotite system at high pressure (Yoshino et al., 2007) (ii) significant grain to grain  
345 surface energy between silicate minerals,  $\sim 4 \text{ J/m}^2$  for olivine (Swain and Atkinson, 1978), though  
346 unfortunately poorly constrained in the presence of majoritic garnet, the second major phase at high  
347 upper-mantle depths and (iii) a major relaxation of the surface energy under hydrous conditions (de  
348 Leeuw et al., 2000). Large-distance melt propagation has already been proposed for the LVL-melt above  
349 the 410 km discontinuity (Leahy and Bercovici, 2007). Widespread circulation of metasomatic agents  
350 is also recognized in the cratonic lithosphere (O'Reilly and Griffin, 2013). We argue that a similar  
351 process also occurs within the convecting mantle and it is possible that chemical reactions favor mantle  
352 refertilization. As observed in nature, the percolation of hydrous melt into a mantle region that is not  
353 saturated in water would induce hydration of NAMs, drying out the melt and inducing its solidification  
354 (Denis et al., 2015; Doucet et al., 2014; Le Roux et al., 2007). Over geological time, the travelling melts  
355 can resolve at least partially the heterogeneity in water content between the different mantle regions. In  
356 this way, all mantle regions (even those that do not undergo dehydration melting upon uprising) could  
357 achieve a water content close to the saturation limit. For further modeling of the total water content in  
358 the upper mantle, we consider that the minerals of the OBM carry an amount of water that is close to  
359 the water saturation limit (Fig. 1).

360 On its way through the OBM, the buoyant hydrous melt eventually meets a critical depth where it  
361 decomposes into a mixture of volatile-bearing fluid and silicate-melt (Kessel et al., 2015; Mibe et al.,  
362 2007). The appearance of a fluid (Gaillard et al., 2008; Hirschmann, 2010) as well as the local  
363 accumulation of a viscous silicate-melt (Chantel et al., 2016; Priestley and McKenzie, 2013) could  
364 produce the low velocity zones (LVZ) detected by seismic and electrical conductivity measurements.  
365 The range of depths between 80 and 150 km where LVZs are detected (e.g. (Debayle and Ricard, 2012;  
366 Kawakatsu et al., 2009)) is compatible with the P-T conditions, around 3.8 GPa and 1273 K, at which  
367 the second critical end point was reported in the peridotite-H<sub>2</sub>O system (Mibe et al., 2007). It is therefore  
368 appealing to make a causal link between the “mantle rain” and the LVZs. A major observation that needs  
369 to be taken into account is that the LVZ seismic and electrical-conductivity anomalies become weaker  
370 and deeper with increasing plate age (e.g. (Debayle et al., 2020)), while we expect ubiquitous upwelling  
371 of the melt from the LVL. A greater impact of the melt in young geological settings could arise from  
372 two major factors: (i) less hydrous melt reaches old compared to young lithospheres. The mantle  
373 underneath mid-ocean ridges is uprising and, therefore, the mantle residue is expected to undergo  
374 continuous dehydration melting on its path (Fig. 3). The hydrous-melt produced this way is added to the  
375 melt coming from the LVL (left side of Fig. 6). Alternatively, the mantle underneath an old lithosphere  
376 should be either stagnant (in depth) or sinking. Thus, no melt is produced by dehydration-melting upon  
377 decompression. Now if we consider that the regions where the mantle is sinking, one can expect that it  
378 would react and stop the LVL-melt at mid upper-mantle depth due to mantle rehydration (right side of  
379 Fig. 6). (ii) Young oceanic crust presents a much steeper temperature gradient in the first 100-150 km  
380 depth, compared to the continental crust. By comparing these temperature profiles and the mantle  
381 solidus, it appears that the formation of a water-saturated melt evolves progressively from  $\sim 150$  ppm at

382 100-150 km depth, to ~700 ppm H<sub>2</sub>O at 250-300 km depth, for young oceanic to old continental settings,  
 383 respectively (Fig. 2). This denotes a larger impact of water in the young lithosphere.



**Fig. 6: Internal cycle of water and the mantle rain model.** The mantle convects upward or downward in left and right parts of the diagram, respectively. Upward movement should dominate, though, in order to balance the inflow of slab (blue) into the deep mantle. Upon uprising of the water-rich MTZ material (light pink) and crossing of the 410 km mineralogical discontinuity, the mantle becomes water-saturated and dehydration melting produces the LVL-melt (dark pink layer). In addition, the uprising of the mantle residue continuously produces a water-bearing melt (curved pink arrows, Fig. 3). Above the upper limit of the LVL, the melt is buoyant and rises toward the surface (linear pink arrows, Fig. 5). Migration of the hydrous melt should take place in all directions, due to low surface tension; for clarity, lateral melt movements are not represented in this figure, except in the LVL. In mantle regions that are sinking, chemical reactions with the travelling hydrous melt is expected to hydrate mantle NAMs. Similar reactions occurring in the lithospheric mantle (point D) can produce mantle refertilisation and eventually intraplate volcanism. (A+B1) is the inflow of water at subduction zones;  $\sim 1\text{-}2 \cdot 10^{12}$  kg.yr<sup>-1</sup> but only (B1) penetrates the deep mantle;  $0.3\text{-}0.8 \cdot 10^{12}$  kg.yr<sup>-1</sup>. (B2) is the water contribution to produce the LVL-melt:  $0.5(0.2) \cdot 10^{12}$  kg.yr<sup>-1</sup>. (C) is the outflow of water at the ocean ridges;  $\sim 0.1 \cdot 10^{12}$  kg.yr<sup>-1</sup>. (B2) should roughly equal (C+D), with (D) the water flux into the lithosphere:  $0.2\text{-}0.7 \cdot 10^{12}$  kg.yr<sup>-1</sup>. While (A) to (D) are water fluxes, (E) is the steady water content in the solid upper mantle. Considering that the upper mantle is almost saturated in water yields a total water content of  $8.5(0.8) \cdot 10^{20}$  kg H<sub>2</sub>O or 0.6(0.1) mass of the oceans. Calculations are detailed in the main text and captions of other figures. This cycle assumes a constant amount of water in the lower mantle and in the core. We note that some dehydration melting may occur below the 660 km and induce low seismic-velocity zones (LM-LVZ).

384 The transfer of melts and fluids enriched in incompatible and volatile elements to the lithospheric  
 385 mantle may contribute to its refertilisation. In addition to well identified, hydrous metasomatism in  
 386 mantle xenoliths from the cratonic mantle (Doucet et al., 2014; Le Roux et al., 2007), the presence of a  
 387 few percent of pyroxenite is often invoked to explain the trace element and isotopic composition of mid-  
 388 ocean ridge basalts (MORBs) originating from the oceanic mantle (Lambart et al., 2009). The hydrous  
 389 melts originating from the transition zone could also help explaining the occurrence of less well-known  
 390 magmatism. For example, the presence of residual fluids under-plating the crust could trigger the

391 formation of petit-spot volcanoes (Machida et al., 2017). Hydrous plumes coming from the MTZ have  
392 also recently been invoked to explain continental magmatism such as continental flood basalts beneath  
393 China (Kuritani et al., 2019; Wang et al., 2015), and beneath the Turkish-Iranian plateau  
394 (Soltanmohammadi et al., 2018). It is also notable that only a few identified mantle plumes (Liu et al.,  
395 2018; Vinnik and Farra, 2007; Zhang et al., 2018) come from great depth (Courtillot et al., 2003).

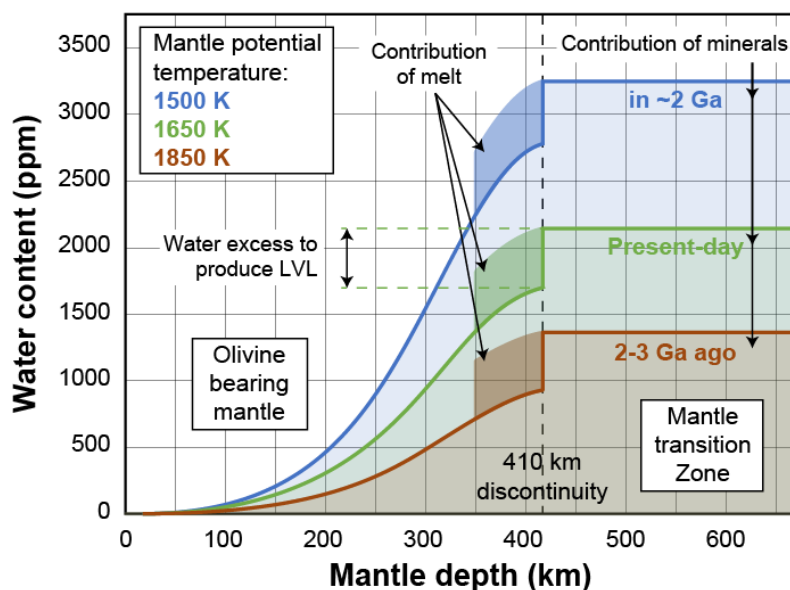
396

### 397 **Water budget in the present-day deep mantle**

398 In mantle regions subjected to continuous upwelling, melt is continuously formed in the LVL, while  
399 a partially depleted, but still water saturated, residue proceeds upwards (Bercovici and Karato, 2003).  
400 On a global scale, an incoming flow of  $\sim 2.5 \cdot 10^{11} \text{ m}^3 \cdot \text{yr}^{-1}$  of subducted slab material (Wen and Anderson,  
401 1995) through the MTZ implies an average uplift of  $\sim 0.6 \text{ mm} \cdot \text{yr}^{-1}$  across the whole 410 km discontinuity.  
402 In order to induce dehydration melting, the water content in the transition zone needs to be higher than  
403 the maximum storage capacity of the OBM, which is 1700(200) ppm  $\text{H}_2\text{O}$  at 410 km for a mantle  
404 potential temperature of 1650 K (Fig. 1, see details in Suppl. Material). In addition, some water is carried  
405 by 0.5 to 1% of LVL melt containing 6.5(1) wt%  $\text{H}_2\text{O}$ . This represents an additional 300 to 700 ppm  
406 (thus, 500(200) ppm)  $\text{H}_2\text{O}$ , yielding a total mantle content of 2200(300) ppm  $\text{H}_2\text{O}$  (Fig. 4) to explain the  
407 seismic velocity drop in the LVL. We now consider that the average water content in the MTZ is  
408 comparable to that in the LVL (2200(300) ppm), which is only  $\sim 30\%$  higher than the maximum  
409 solubility limit of the mantle (1700(200) ppm) in the LVL. This assumption implies that there is no  
410 accumulation of melt in the LVL, but instead continuous melt formation by dehydration melting and  
411 escape by *Mantle Rain* of a same amount of LVL-melt. Here, we evaluate what are the consequences of  
412 this assumption: (i) if the water content of the mantle uprising from the MTZ would be lower, between  
413 1700(200) and 2200(300) ppm water, then there would still be dehydration melting. There would be less  
414 LVL-melt produced for a same amount of uprising MTZ material. This would lower the fluxes of LVL-  
415 melt out of the LVL and loss by uprising. (ii) Alternatively, more than 2200(300) ppm water in the MTZ  
416 would produce a fraction of LVL-melt higher than expected from the comparison between experiments  
417 and seismic data. This could be accommodated by the *Mantle Rain* model with a greater flux of uprising  
418 LVL-melt. Still, it should be compensated by a higher influx of water in the MTZ, which does not seem  
419 to be available (see text above and Fig. 6). It makes it unreasonable a MTZ water content significantly  
420 higher than 2200(300) ppm. In summary, slight variations of the MTZ water content away from the  
421 average value of 2200(300) ppm does not significantly change either the *Mantle Rain* model or the upper  
422 mantle water balance estimate that we develop below. We note that this value is far below the level of  
423 water saturation in MTZ, because wadsleyite can include up to 5 times more hydrogen than olivine at  
424 these P-T conditions (Ferot and Bolfan-Casanova, 2012). Based on the average value of 2200(300) ppm,  
425 the MTZ should contain  $8.5(0.8) \cdot 10^{20} \text{ kg}$  of water, corresponding to 0.6(0.1) OM.

426 The continuous rise of the water-bearing MTZ material by  $\sim 0.6 \text{ mm} \cdot \text{yr}^{-1}$  corresponds to a total of  
427  $\sim 2.0(0.3) \cdot 10^{12} \text{ kg} \cdot \text{yr}^{-1} \text{ H}_2\text{O}$  crossing upwards the 410 km discontinuity. This value is significantly larger  
428 than the 0.3 to  $0.8 \cdot 10^{12} \text{ kg} \cdot \text{yr}^{-1} \text{ H}_2\text{O}$  injected in the deep mantle by subduction ((van Keken et al., 2011);  
429 B1 in Fig. 6). Instead, it is the flux of water producing the LVL-melt, about  $0.5(0.2) \cdot 10^{12} \text{ kg} \cdot \text{yr}^{-1}$  (B2 in  
430 Fig. 6), that is well compatible with the flux of water entering the MTZ by subduction. This indicates  
431 that the production of LVL-melt is primarily triggered by the amount of deeply subducted water. On the  
432 other hand, the origin of the water present in NAMs uprising to the LVL cannot come from the subducted  
433 water. In the framework of global convection, NAMs are expected to cross the 410 km boundary in both  
434 directions and, for this reason, their contribution to the water transfer through the 410 km boundary is  
435 balanced. By analogy, the situation is comparable to the presence of incompatible elements in peridotite;  
436 they do not come from the subduction of MORBs. The water content of NAMs sinking through the 410  
437 km discontinuity is unknown. However, it could be close to the NAMs saturation limit due to reaction  
438 with hydrous-melt present in the upper mantle (see above). In addition, they cross the water-saturated

439 LVL and could react there with the water-bearing melt. For this reason, we consider that the water  
 440 content of NAMs in the upper mantle as a constant (contribution E in Fig. 6).



**Fig. 7: Water content in the upper mantle as a function of mantle potential temperature.** At a depth shallower than 410 km, the H<sub>2</sub>O content of the mantle follows the water-saturation profiles (Fig. 1). On the other hand, the water content in the mantle transition zone can be estimated as being the sum of the solubility limit in the mantle above 410 km and the water content needed to produce 0.5 to 1 wt% of hydrous-melt in the low-velocity layer, in agreement with seismic data. The mean thickness of the LVL is ~60 km, but depends on mantle heterogeneities in H<sub>2</sub>O and FeO contents, and temperature (see text). The same calculation is performed for mantle potential temperatures of 1850 K (red: 2-3 Ga ago), 1650 K (green: ~present-day) and 1500 K (blue: in the future). Uncertainty on water contents is same as in Fig. 1.

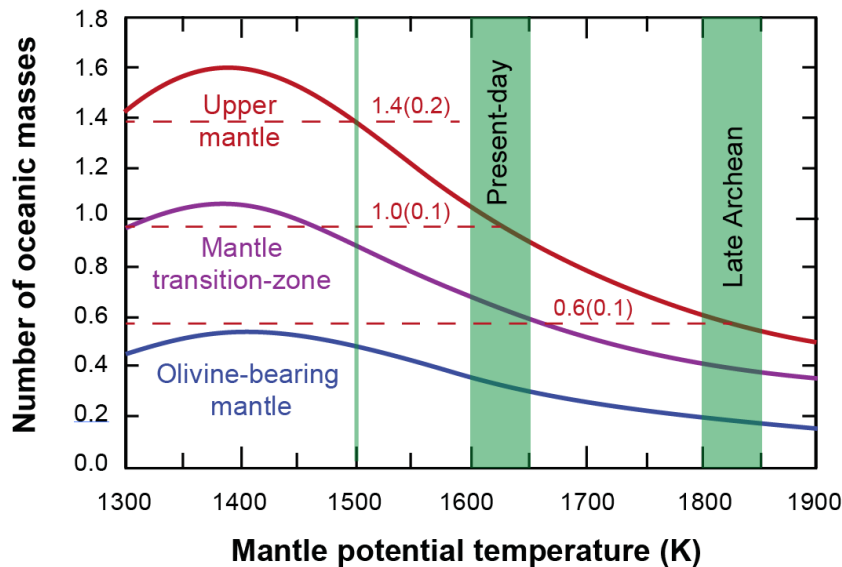
441 Now we calculate the total amount of water present in the  
 442 upper mantle (Fig. 7). An OBM close to its water saturation limit  
 443 yields an integrated value of its water content of  $\sim 4.3(0.4) 10^{20}$  kg  
 444 or  $\sim 0.31(0.03)$  OM. Added to the  $0.6(0.1)$  OM in the MTZ (see  
 445 above), we conclude that a water-equivalent of  $\sim 1.0(0.1)$  OM of  
 446 water currently resides in the mantle above the 660 km  
 447 discontinuity. We acknowledge that this calculation does not  
 448 include possible effects associated with an increase in the water  
 449 solubility limit in the lithospheric mantle and the crust (Green et al.,  
 450 2014; Mierdel et al., 2007; Sarafian et al., 2017). For estimating a  
 451 water budget over the entire mantle, the amount of water residing  
 452 in the lower mantle should also be considered: different  
 453 experimental works based on high-quality single-crystal of  
 454 bridgmanite find a radically different water storage capacity from  $\sim 100$  ppm (Bolfan-Casanova et al.,  
 455 2003) to  $\sim 1000$  ppm (Fu et al., 2019), which translates into maxima of  $\sim 0.3$  to  $\sim 3$  OM in the lower  
 456 mantle. Ferropiclasite and CaSiO<sub>3</sub>-perovskite may also integrate minor amounts of H. In addition, some  
 457 water may be carried below the 660 km discontinuity in dense hydrous phases that remain stable in low-  
 458 temperature slabs (Nishi et al., 2014; Pamato et al., 2015). Still, partition coefficients greater than 10  
 459 between most NAMs and bridgmanite favor the storage of water in the MTZ, compared to the lower  
 460 mantle (Bolfan-Casanova et al., 2003) and thus we consider that the lower mantle should be fairly dry.

461  
 462 **Water budget of the mantle over the Earth's history**

463 We evaluate the amount of water that has been stored in the mantle over the course of the Earth's  
 464 history. Analyses of ancient non-arc basalts and komatiites suggest a rate of mantle cooling of  $\sim 100$   
 465 K/Ga (e.g. (Herzberg et al., 2010)). A higher mantle temperature in the past implies a lower water storage  
 466 capacity for NAMs (Fig. 1) and thus less water stored in the upper mantle (Fig. 7). The amount of water  
 467 stored in the MTZ in the past can be estimated from the maximum water storage capacity of the mantle  
 468 just above the 410 km discontinuity making following assumptions: (i) the LVL has persisted since the  
 469 establishment of slab subduction, due to the parental link between subducted water and formation of the  
 470 LVL (see above). (ii) LVL properties, e.g. its dependence on mantle temperature and water content,

471 have remained the same over time, which enables the use of the melting diagram (Fig. 4). It implies that  
 472 the excess water in the MTZ, compared to the OBM, is regulated by the LVL (Fig. 7). In other words,  
 473 we assume that although the deep water cycle has evolved over the last 2-3 Ga, a stable equilibrium  
 474 between the flow of water into the MTZ via subduction and the flow of water out the MTZ via melt  
 475 formed at the LVL was established early on and maintained over geological times. We thus obtain water  
 476 contents of 930(100) or 2800(300) ppm for the OBM at 410 km depth from Fig. 1, and 1400(200) or  
 477 3300(500) ppm for the MTZ, for  $T_P$  varying from the highest values of 1850 K (in the past) to 1500 K  
 478 (in the future), respectively (Fig. 7).

479 The water storage capacity in the entire upper mantle (above 660 km depth) is found to increase  
 480 from  $\sim 0.6(0.1)$  to  $\sim 1.4(0.2)$  OM from 2-3 Ga ago ( $T_P \sim 1850$  K) to a  
 481 point in the future when  $T_P$  will achieve 1500 K (Fig. 8). This  
 482 corresponds to a net decrease of the water elevation of the oceans by  
 483  $\sim 900$  m in the last 2-3 Ga, for a mean elevation today of 4.5 km. These  
 484 trends correspond to a decrease of the oceanic mass of  $\sim 20\%$  per Ga, in  
 485 agreement with previous findings of a loss of several 10s % of the OM  
 486 over geological time, based on hydrogen isotopes (Lecuyer et al., 1998;  
 487 Pope et al., 2012) and geodynamic modeling (Flament et al., 2013). The  
 488 mantle storage capacity will continue to increase to a maximum value  
 489 of  $\sim 1.6$  OM, when  $T_P$  may eventually decrease to 1400 K in several Ga  
 490 from now.



**Fig. 8: Water content of upper mantle reservoirs throughout Earth's history.** Water contents in the entire upper mantle (red), constituted of the OBM (blue) and of the MTZ (purple), calculated from the integration of water contents as a function of depth (Fig. 7) for various  $T_P$ . With present-day  $T_P \sim 1650$  K, we expect  $\sim 1.0(0.1)$  ocean mass (OM) trapped in the mantle above 660 km (central red dashed line), with  $\sim 0.4$  and  $\sim 0.6$  OM in OBM and MTZ, respectively). The water storage capacity in the upper mantle increases with secular cooling at a rate of  $\sim 20\%$  of the ocean mass per billion years. Uncertainty is  $\pm 15\%$  similar to that of Fig. 1.

491 Finally, we investigate the critical amount of water needed to produce a planet like the Earth in  
 492 terms of the surface ratio between oceans and emerged-continents. For the Earth, the present-day  $\sim 2.0$   
 493 OM (1.0 OM residing at the surface and 1.0 OM in the mantle) stored above the 660 km discontinuity  
 494 ensures the existence of an ocean at the surface of the planet regardless of its age (i.e. its mantle  
 495 temperature). During the Archean, the mantle was hotter and thus its water storage capacity was lower,  
 496  $\sim 0.6$  OM, implying a volume of Archean oceans  $\sim 1.3$  times that of today's. This agrees with inferences,  
 497 based on the  $^{87}\text{Sr}/^{86}\text{Sr}$  isotopic composition of marine carbonates, that there was little (less than 4%)  
 498 emerged land 2.5 Ga ago (Flament et al., 2008). With cooling to the present-day potential temperature  
 499 of 1650 K, the hydrosphere would have been entirely buried into the solid mantle if the total terrestrial  
 500 water content had been less than  $\sim 1.0$  OM (Fig. 8). In the future, the water storage capacity of the upper  
 501 mantle is expected to further increase with secular cooling, possibly reaching a maximum of  $\sim 1.6$  OM  
 502 for a mantle potential temperature of 1400 K. This would leave only 0.4 OM at the Earth's surface.  
 503 Therefore, variations of the total amount of water in the mantle of a planet with the size of the Earth



504 may easily result in no emerged land early in the history, or the absence of a surface ocean a few Ga  
505 after the planet's formation, just because of the interplay with the mantle.

506

## 507 **Conclusion**

508 In this article, we develop the "mantle rain" model and its implications. The model is based on the  
509 mechanism of dehydration melting taking place above the 410 km discontinuity and producing a water-  
510 saturated melt and solid residue, which together form the LVL observed by seismology. Starting from  
511 the well-accepted concept of a neutral melt-mantle buoyancy at the upper limit of the LVL, we develop  
512 the idea that the LVL is a source for upwelling melts. With such mechanism, it becomes possible to  
513 balance the incoming (by subduction) and outgoing (by rise of LVL-melt) water flows for the MTZ.  
514 Within the upper mantle, a water-bearing melt is expected to travel several orders of magnitude faster  
515 than the mantle residue stirred by convection movements. The melt could travel relatively fast laterally  
516 and vertically, favoring homogenization of the water content, mantle refertilisation, seismic anomalies  
517 such as the LVZ, intraplate volcanism, etc.

518 The occurrence of dehydration melting in the LVL also offers a clue to estimate the average water  
519 content in the MTZ. It should be higher than the maximum water-storage capacity of the OBM at 410  
520 km depth, 1700(200) ppm, as determined experimentally, and should also account for the continuous  
521 production of the water-bearing LVL-melt: thus, it should be ~2200(300) ppm H<sub>2</sub>O for T<sub>p</sub> of 1650 K.  
522 Based on this value and with the idea that the OBM should be almost saturated in water, we find that ~1  
523 ocean mass of water is stored in the entire upper mantle today. The same calculation can be performed  
524 for mantle potential temperatures higher (in the past) and lower (in the future) than that of 1600-1650 K  
525 prevailing today. With an increasing water storage capacity of the mantle as a function of decreasing  
526 mantle temperature, the mass of the oceans at the Earth's surface is found to decrease by 20% every  
527 billion years, when considering a rate of mantle cooling of ~100 K/Ga.

528

529 **Supplementary Material:**

530

531 **Water fugacity in the upper mantle**

532

533 We calculate the water fugacity ( $f_{H_2O}$ ) of pure water based on the previous equation of state (EoS)  
534 of H<sub>2</sub>O (Pitzer and Sterner, 1994). However, geological fluids/melts are not pure water, at high pressures  
535 above ~3 GPa, as they dissolve a significant amount of silicate at high mantle temperatures (Massuyeau  
536 et al., 2015; Mibe et al., 2007). It makes the  $f_{H_2O}$  significantly lower than that of pure H<sub>2</sub>O. To take this  
537 effect into account, we apply a correction to the  $f_{H_2O}$  of pure water based on a previous experimental  
538 report of the water content of olivine up to 12 GPa and 1673 K (Suppl. Fig. 1) (Bali et al., 2008):  
539

540 
$$f_{H_2O} = f_{H_2O}^{\text{Pure-water}} * [1 - A_1 \exp(-\Delta G_1/RT)]$$
 (Suppl-Eq. 1)

541

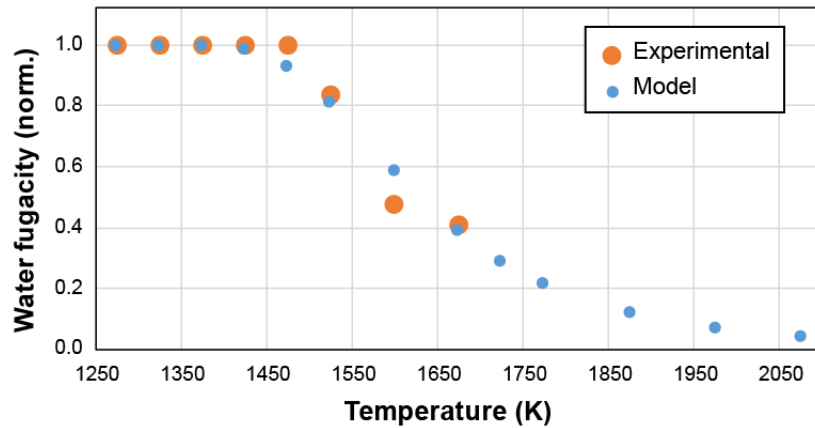
542 with  $\Delta G_1$  related to the energy of dissolution of the silicate component in the fluid.  $\Delta G_1$  can vary  
543 with several parameters, in particular temperature and fluid composition. In order to reproduce the ratio  
544 of real water fugacity over that of the pure fluid reported previously (Suppl. Fig. 1; (Bali et al., 2008)),  
545 we adopted a  $\Delta G_1$  value that decreases exponentially with increasing temperature:  
546

547 
$$\Delta G_1 = A_2 \exp(-\Delta G_2/RT)$$
 (Suppl-Eq. 2)

548

549 with  $A_1=1$ ,  $A_2=2.50 \cdot 10^{-2}$  and  $\Delta G_2=-140$  kJ/mol. For convenience, we use this formalism for further  
550 modeling of  $f_{H_2O}$  in supercritical fluids or hydrous melts at upper mantle pressures and temperatures.  
551

552



**Suppl. Fig. 1: Temperature dependence of water fugacity (i.e. the correction to apply to the  $f_{H_2O}$  of pure water).** Experimental data obtained between 3 and 12 GPa (Bali et al., 2008) are fitted using a double exponential function (Suppl-Eq. 1 and 2). The model reproduces well the amplitude of the  $f_{H_2O}$  decrease induced by a change of the melt composition (from water-rich to silicate-rich) with increasing the temperature above ~1500 K.

553 **Acknowledgements:** We thank E. Debayle and B. Tauzin for major inputs concerning seismic  
554 tomography in the upper mantle, P. Condamine for precisions about his sandwich-derived composition  
555 of LVL-melt, S. Demouchy, D. Freitas, M. Garçon, D. Laporte, G. Manthilake, E. Médard, M.  
556 Monnereau and anonymous reviewers for fruitful discussions. This research was financed by the  
557 French Government Laboratory of Excellence initiative n°ANR-10-LABX-0006, the Région  
558 Auvergne and the European Regional Development Fund. This is Laboratory of Excellence ClerVolc  
559 contribution N°XX.

560

561

562 **References cited:**

563 Andrault, D., Pesce, G., Manthilake, G., Monteux, J., Bolfan-Casanova, N., Chantel, J., Novella, D.,  
564 Guignot, N., King, A., Itie, J.-P., Hennet, L., 2018. Deep and persistent melt layer in the Archaean  
565 mantle. *Nat. Geosci.* 11, 139-+.

566 Ardia, P., Hirschmann, M.M., Withers, A.C., Tenner, T.J., 2012. H<sub>2</sub>O storage capacity of olivine at 5-8  
567 GPa and consequences for dehydration partial melting of the upper mantle. *Earth Planet. Sci. Lett.*  
568 345, 104-116.

569 Aubaud, C., Hauri, E.H., Hirschmann, M.M., 2004. Hydrogen partition coefficients between nominally  
570 anhydrous minerals and basaltic melts. *Geophys. Res. Lett.* 31.

571 Bali, E., Bolfan-Casanova, N., Koga, K.T., 2008. Pressure and temperature dependence of H solubility  
572 in forsterite : an implication to water activity in the Earth interior. *Earth Planet. Sci. Lett.*, 354-363.

573 Bercovici, D., Karato, S., 2003. Whole-mantle convection and the transition-zone water filter. *Nature*  
574 425, 39-44.

575 Bolfan-Casanova, N., Keppler, H., Rubie, D.C., 2000. Water partitioning between nominally anhydrous  
576 minerals in the MgO-SiO<sub>2</sub>-H<sub>2</sub>O system up to 24 GPa: implications for the distribution of water in  
577 the Earth's mantle. *Earth Planet. Sci. Lett.* 182, 209-221.

578 Bolfan-Casanova, N., Keppler, H., Rubie, D.C., 2003. Water partitioning at 660 km depth and evidence  
579 for very low water solubility in magnesium silicate perovskite. *Geophys. Res. Lett.* 30, L017182.

580 Buchen, J., Marquardt, H., Speziale, S., Kawazoe, T., Boffa Ballaran, T., Kurnosov, A., 2018. High-  
581 pressure single-crystal elasticity of wadsleyite and the seismic signature of water in the shallow  
582 transition zone. *Earth Planet. Sci. Lett.* 498, 77-87.

583 Chantel, J., Manthilake, G., Andrault, D., Novella, D., Yu, T., Wang, Y.B., 2016. Experimental evidence  
584 supports mantle partial melting in the asthenosphere. *Sci. Adv.* 2.

585 Cogne, J.P., Humler, E., 2006. Trends and rhythms in global seafloor generation rate. *Geochem.,*  
586 *Geophys., Geosyst.* 7.

587 Condamine, P., Frost, D.J., 2018. Kimberlite from incipient melting of the mantle transition zone?,  
588 Annual Report of BGI. Bayerisches Geoinstitute, Bareuth.

589 Courtillot, V., Davaille, A., Besse, J., Stock, J., 2003. Three distinct types of hotspots in the Earth's  
590 mantle. *Earth Planet. Sci. Lett.* 205, 295-308.

591 Dasgupta, R., Hirschmann, M.M., 2007. A modified iterative sandwich method for determination of  
592 near-solidus partial melt compositions. II. Application to determination of near-solidus melt  
593 compositions of carbonated peridotite. *Contrib. Mineral. Petr.* 154, 647-661.

594 de Leeuw, N.H., Parker, S.C., Catlow, C.R.A., Price, G.D., 2000. Modelling the effect of water on the  
595 surface structure and stability of forsterite. *Phys. Chem. Mineral.* 27, 332-341.

596 Debayle, E., Bodin, T., Durand, S., Ricard, Y., 2020. Seismic evidence for partial melt below tectonic  
597 plates. *Nature* 586, 555-559.

598 Debayle, E., Ricard, Y., 2012. A global shear velocity model of the upper mantle from fundamental and  
599 higher Rayleigh mode measurements. *J. Geophys. Res.* 117.

600 Demouchy, S., 2010. Hydrogen diffusion in spinel grain boundaries and consequences for chemical  
601 homogenization in hydrous peridotite. *Contrib. Mineral. Petr.* 160, 887-898.

602 Demouchy, S., Shcheka, S., Denis, C.M.M., Thoraval, C., 2017. Subsolidus hydrogen partitioning  
603 between nominally anhydrous minerals in garnet-bearing peridotite. *Am. Miner.* 102, 1822-1831.

604 Denis, C.M.M., Alard, O., Demouchy, S., 2015. Water content and hydrogen behaviour during  
605 metasomatism in the uppermost mantle beneath Ray Pic volcano (Massif Central, France). *Lithos*  
606 236-237, 256-274.

607 Dong, J., Fischer, R.A., Stixrude, L.P., Lithgow-Bertelloni, C.R., 2021. Constraining the Volume of  
608 Earth's Early Oceans With a Temperature-Dependent Mantle Water Storage Capacity Model. *AGU*  
609 *Advances* 2, e2020AV000323.

610 Doucet, L.S., Peslier, A.H., Ionov, D.A., Brandon, A.D., Golovin, A.V., Goncharov, A.G., Ashchepkov,  
611 I.V., 2014. High water contents in the Siberian cratonic mantle linked to metasomatism: An FTIR  
612 study of Udachnaya peridotite xenoliths. *Geochim. Cosmochim. Acta* 137, 159-187.

613 Druzhbin, D., Fei, H., Katsura, T., 2021. Independent hydrogen incorporation in wadsleyite from oxygen  
614 fugacity and non-dissociation of H<sub>2</sub>O in the reducing mantle transition zone. *Earth Planet. Sci.*  
615 *Lett.* 557, 116755.

616 Durand, S., Debayle, E., Ricard, Y., Zaroли, C., Lambotte, S., 2017. Confirmation of a change in the  
617 global shear velocity pattern at around 1000km depth. *Geophys. J. Int.* 211, 1628-1639.

618 Fei, H., Yamazaki, D., Sakurai, M., Miyajima, N., Ohfuji, H., Katsura, T., Yamamoto, T., 2017. A nearly  
619 water-saturated mantle transition zone inferred from mineral viscosity. *Sci. Adv.* 3.

620 Ferot, A., Bolfan-Casanova, N., 2012. Water storage capacity in olivine and pyroxene to 14 GPa:  
621 Implications for the water content of the Earth's upper mantle and nature of seismic discontinuities.  
622 *Earth Planet. Sci. Lett.* 349, 218-230.

623 Flament, N., Coltice, N., Rey, P.F., 2008. A case for late-Archaean continental emergence from thermal  
624 evolution models and hypsometry. *Earth Planet. Sci. Lett.* 275, 326-336.

625 Flament, N., Coltice, N., Rey, P.F., 2013. The evolution of the Sr-87/Sr-86 of marine carbonates does  
626 not constrain continental growth. *Precambrian Res.* 229, 177-188.

627 Freitas, D., Manthilake, G., Chantel, J., Bouhifd, M.A., Andrault, D., 2019. Simultaneous measurements  
628 of electrical conductivity and seismicwave velocity of partially molten geological materials: effect  
629 of evolving melt texture. *Phys. Chem. Mineral.* 46, 535-551.

630 Freitas, D., Manthilake, G., Schiavi, F., Chantel, J., Bolfan-Casanova, N., Bouhifd, M.A., Andrault, D.,  
631 2017. Experimental evidence supporting a global melt layer at the base of the Earth's upper mantle.  
632 *Nat. Commun.* 8.

633 Frost, D.J., Dolejs, D., 2007. Experimental determination of the effect of H<sub>2</sub>O on the 410-km seismic  
634 discontinuity. *Earth Planet. Sci. Lett.* 256, 182-195.

635 Fu, S., Yang, J., Karato, S.-i., Vasiliev, A., Presniakov, M.Y., Gavriliuk, A.G., Ivanova, A.G., Hauri,  
636 E.H., Okuchi, T., Purejav, N., Lin, J.-F., 2019. Water Concentration in Single-Crystal (Al,Fe)-  
637 Bearing Bridgmanite Grown From the Hydrous Melt: Implications for Dehydration Melting at the  
638 Topmost Lower Mantle. *Geophys. Res. Lett.* 46, 10346-10357.

639 Fukao, Y., Obayashi, M., 2013. Subducted slabs stagnant above, penetrating through, and trapped below  
640 the 660 km discontinuity. *J. Geophys. Res.* 118, 5920-5938.

641 Gaillard, F., Malki, M., Iacono-Marziano, G., Pichavant, M., Scaillet, B., 2008. Carbonatite Melts and  
642 Electrical Conductivity in the Asthenosphere. *Science* 322, 1363-1365.

643 Green, D.H., Hibberson, W.O., Rosenthal, A., Kovacs, I., Yaxley, G.M., Falloon, T.J., Brink, F., 2014.  
644 Experimental Study of the Influence of Water on Melting and Phase Assemblages in the Upper  
645 Mantle. *Journal of Petrology* 55, 2067-2096.

646 Greenwood, R.C., Barrat, J.A., Miller, M.F., Anand, M., Dauphas, N., Franchi, I.A., Sillard, P., Starkey,  
647 N.A., 2018. Oxygen isotopic evidence for accretion of Earth's water before a high-energy Moon-  
648 forming giant impact. *Sci. Adv.* 4.

649 Hae, R., Ohtani, E., Kubo, T., Koyama, T., Utada, H., 2006. Hydrogen diffusivity in wadsleyite and  
650 water distribution in the mantle transition zone. *Earth Planet. Sci. Lett.* 243, 141-148.

651 Herzberg, C., Condie, K., Korenaga, J., 2010. Thermal history of the Earth and its petrological  
652 expression. *Earth Planet. Sci. Lett.* 292, 79-88.

653 Hier-Majumder, S., Keel, E.B., Courtier, A.M., 2014. The influence of temperature, bulk composition,  
654 and melting on the seismic signature of the low-velocity layer above the transition zone. *J. Geophys.*  
655 *Res.* 119, 971-983.

656 Hier-Majumder, S., Ricard, Y., Bercovici, D., 2006. Role of grain boundaries in magma migration and  
657 storage. *Earth Planet. Sci. Lett.* 248, 735-749.

658 Hier-Majumder, S., Tauzin, B., 2017. Pervasive upper mantle melting beneath the western US. *Earth*  
659 *Planet. Sci. Lett.* 463, 25-35.

660 Hirschmann, M.M., 2010. Partial melt in the oceanic low velocity zone. *Phys. Earth Planet. Inter.* 179,  
661 60-71.

662 Hirschmann, M.M., Aubaud, C., Withers, A.C., 2005. Storage capacity of H<sub>2</sub>O in nominally anhydrous  
663 minerals in the upper mantle. *Earth Planet. Sci. Lett.* 236, 167-181.

664 Huang, X., Xu, Y., Karato, S., 2005. Water content in the transition zone from electrical conductivity of  
665 wadsleyite and ringwoodite. *Nature* 434, 746-749.

666 Inoue, T., 1994. Effect of water on melting phase relations and melt composition in the system  
667 Mg<sub>2</sub>SiO<sub>4</sub>-MgSiO<sub>3</sub>-H<sub>2</sub>O up to 15 GPa. *Phys. Earth Planet. Inter.* 85, 237-263.

668 Irifune, T., Isshiki, M., 1998. Iron partitioning in a pyrolite mantle and the nature of the 410-km seismic  
669 discontinuity. *Nature* 392, 702-705.

670 Jing, Z., Karato, S.-i., 2012. Effect of H<sub>2</sub>O on the density of silicate melts at high pressures: Static  
671 experiments and the application of a modified hard-sphere model of equation of state. *Geochim.*  
672 *Cosmochim. Acta* 85, 357-372.

673 Katsura, T., Yoneda, A., Yamazaki, D., Yoshino, T., Ito, E., 2010. Adiabatic temperature profile in the  
674 mantle. *Phys. Earth Planet. Inter.* 183, 212-218.

675 Kawakatsu, H., Kumar, P., Takei, Y., Shinohara, M., Kanazawa, T., Araki, E., Suyehiro, K., 2009.  
676 Seismic Evidence for Sharp Lithosphere-Asthenosphere Boundaries of Oceanic Plates. *Science*  
677 324, 499-502.

678 Kessel, R., Pettko, T., Fumagalli, P., 2015. Melting of metasomatized peridotite at 4-6 GPa and up to  
679 1200 degrees C: an experimental approach. *Contrib. Mineral. Petr.* 169.

680 Kohlstedt, D.L., Keppler, H., Rubie, D.C., 1996. Solubility of water in the alpha, beta and gamma phases  
681 of (Mg,Fe)<sub>2</sub>SiO<sub>4</sub>. *Contrib. Mineral. Petr.* 123, 345-357.

682 Kuritani, T., Xia, Q.K., Kimura, J., Liu, J., Shimizu, K., Ushikubo, T., Zhao, D.P., Nakagawa, M.,  
683 Yoshimura, S., 2019. Buoyant hydrous mantle plume from the mantle transition zone. *Scientific*  
684 *Reports* 9.

685 Lambart, S., Laporte, D., Schiano, P., 2009. An experimental study of pyroxenite partial melts at 1 and  
686 1.5 GPa: Implications for the major-element composition of Mid-Ocean Ridge Basalts. *Earth*  
687 *Planet. Sci. Lett.* 288, 335-347.

688 Le Roux, V., Bodinier, J.L., Tommasi, A., Alard, O., Dautria, J.M., Vauchez, A., Riches, A.J.V., 2007.  
689 The Lherz spinel lherzolite: Refertilized rather than pristine mantle. *Earth Planet. Sci. Lett.* 259,  
690 599-612.

691 Leahy, G.M., Bercovici, D., 2007. On the dynamics of a hydrous melt layer above the transition zone.  
692 *J. Geophys. Res.* 112.

693 Lecuyer, C., Gillet, P., Robert, F., 1998. The hydrogen isotope composition of seawater and the global  
694 water cycle. *Chemical Geology* 145, 249-261.

695 Liu, Z., Fei, H., Chen, L., McCammon, C., Wang, L., Liu, R., Wang, F., Liu, B., Katsura, T., 2021.  
696 Bridgmanite is nearly dry at the top of the lower mantle. *Earth Planet. Sci. Lett.* 570, 117088.

697 Liu, Z., Park, J., Karato, S.-i., 2016. Seismological detection of low-velocity anomalies surrounding the  
698 mantle transition zone in Japan subduction zone. *Geophys. Res. Lett.* 43, 2480-2487.

699 Liu, Z., Park, J., Karato, S.-i., 2018. Seismic evidence for water transport out of the mantle transition  
700 zone beneath the European Alps. *Earth Planet. Sci. Lett.* 482, 93-104.

701 Machida, S., Kogiso, T., Hirano, N., 2017. Petit-spot as definitive evidence for partial melting in the  
702 asthenosphere caused by CO<sub>2</sub>. *Nat. Commun.* 8.

703 Mao, Z., Jacobsen, S.D., Jiang, F., Smyth, J.R., Holl, C.M., Duffy, T.S., 2008. Elasticity of hydrous  
704 wadsleyite to 12 GPa: Implications for Earth's transition zone. *Geophys. Res. Lett.* 35.

705 Marty, B., 2012. The origins and concentrations of water, carbon, nitrogen and noble gases on Earth.  
706 *Earth Planet. Sci. Lett.* 313, 56-66.

707 Massuyeau, M., Gardés, E., Morizet, Y., Gaillard, F., 2015. A model for the activity of silica along the  
708 carbonatite-kimberlite-mellilitite-basanite melt compositional joint. *Chemical Geology* 418, 206-  
709 216.

710 Matsukage, K.N., Jing, Z.C., Karato, S., 2005. Density of hydrous silicate melt at the conditions of  
711 Earth's deep upper mantle. *Nature* 438, 488-491.

712 Mibe, K., Kanzaki, M., Kawamoto, T., Matsukage, K.N., Fei, Y.W., Ono, S., 2007. Second critical  
713 endpoint in the peridotite-H<sub>2</sub>O system. *J. Geophys. Res.* 112.

714 Mibe, K., Orihashi, Y., Nakai, S., Fujii, T., 2006. Element partitioning between transition-zone minerals  
715 and ultramafic melt under hydrous conditions. *Geophys. Res. Lett.* 33.

716 Michael, P.J., 1988. The concentration, behavior and storage of H<sub>2</sub>O in the suboceanic upper mantle:  
717 Implications for mantle metasomatism. *Geochim. Cosmochim. Acta* 52, 555-566.

718 Mierdel, K., Keppler, H., Smyth, J.R., Langenhorst, F., 2007. Water solubility in aluminous  
719 orthopyroxene and the origin of Earth's asthenosphere. *Science* 315, 364-368.

720 Mosenfelder, J.L., Deligne, N.I., Asimow, P.D., Rossman, G.R., 2006. Hydrogen incorporation in  
721 olivine from 2-12 GPa. *Am. Miner.* 91, 285-294.

722 Ni, H., Keppler, H., Behrens, H., 2011. Electrical conductivity of hydrous basaltic melts: implications  
723 for partial melting in the upper mantle. *Contrib. Mineral. Petr.* 162, 637-650.

724 Nishi, M., Irifune, T., Tsuchiya, J., Tange, Y., Nishihara, Y., Fujino, K., Higo, Y., 2014. Stability of  
725 hydrous silicate at high pressures and water transport to the deep lower mantle. *Nat. Geosci.* 7, 224-  
726 227.

727 Novella, D., Frost, D.J., Hauri, E.H., Bureau, H., Raepsaet, C., Roberge, M., 2014. The distribution of  
728 H<sub>2</sub>O between silicate melt and nominally anhydrous peridotite and the onset of hydrous melting in  
729 the deep upper mantle. *Earth Planet. Sci. Lett.* 400, 1-13.

730 O'Reilly, S.Y., Griffin, W.L., 2013. Mantle metasomatism. *Metasomatism and the Chemical  
731 Transformation of Rock*, Lecture Notes in Earth System Sciences. Springer Berlin Heidelberg, pp.  
732 471-533.

733 Ohtani, E., Mizobata, H., Yurimoto, H., 2000. Stability of dense hydrous magnesium silicate phases in  
734 the systems Mg<sub>2</sub>SiO<sub>4</sub>-H<sub>2</sub>O and MgSiO<sub>3</sub>-  
735 H<sub>2</sub>O at pressures up to 27 GPa. *Phys. Chem. Mineral.* 27, 533-544.

736 Ohtani, E., Yuan, L., Ohira, I., Shatskiy, A., Litasov, K., 2018. Fate of water transported into the deep  
737 mantle by slab subduction. *Journal of Asian Earth Sciences* 167, 2-10.

738 Padrón-Navarta, J.A., Hermann, J., 2017. A Subsolidus Olivine Water Solubility Equation for the Earth's  
739 Upper Mantle. *J. Geophys. Res.* 122, 9862-9880.

740 Pamato, M.G., Myhill, R., Ballaran, T.B., Frost, D.J., Heidelbach, F., Miyajima, N., 2015. Lower-mantle  
741 water reservoir implied by the extreme stability of a hydrous aluminosilicate. *Nat. Geosci.* 8, 75-  
742 79.

743 Pearson, D.G., Brenker, F.E., Nestola, F., McNeill, J., Nasdala, L., Hutchison, M.T., Matveev, S.,  
744 Mather, K., Silversmit, G., Schmitz, S., Vekemans, B., Vincze, L., 2014. Hydrous mantle transition  
745 zone indicated by ringwoodite included within diamond. *Nature* 507, 221-+.

746 Piani, L., Marrocchi, Y., Rigaudier, T., Vacher, L.G., Thomassin, D., Marty, B., 2020. Earth's water  
747 may have been inherited from material similar to enstatite chondrite meteorites. *Science* 369, 1110-  
748 +.

749 Pitzer, K.S., Sterner, S.M., 1994. Equation of state valid continuously from zero to extreme pressures  
750 for H<sub>2</sub>O and CO<sub>2</sub>. *J. Chem. Phys.* 101, 3111-3116.

751 Pope, E.C., Bird, D.K., Rosing, M.T., 2012. Isotope composition and volume of Earth's early oceans.  
752 *Proc. Natl. Acad. Sci. U. S. A.* 109, 4371-4376.

753 Priestley, K., McKenzie, D., 2013. The relationship between shear wave velocity, temperature,  
754 attenuation and viscosity in the shallow part of the mantle. *Earth Planet. Sci. Lett.* 381, 78-91.

755 Revenaugh, J., Jordan, T.H., 1991. Mantle layering from SCS reverberations 3. The upper mantle. *J.  
756 Geophys. Res.* 96, 19781-19810.

757 Revenaugh, J., Sipkin, S.A., 1994. Seismic evidence for silicate melt atop the 410 km mantle  
758 discontinuity. *Nature* 369, 474-476.

759 Rupke, L.H., Morgan, J.P., Hort, M., Connolly, J.A.D., 2004. Serpentine and the subduction zone water  
760 cycle. *Earth Planet. Sci. Lett.* 223, 17-34.

761 Sakamaki, T., Suzuki, A., Ohtani, E., 2006. Stability of hydrous melt at the base of the Earth's upper  
762 mantle. *Nature* 439, 192-194.

763 Sarafian, E., Gaetani, G.A., Hauri, E.H., Sarafian, A.R., 2017. Experimental constraints on the damp  
764 peridotite solidus and oceanic mantle potential temperature. *Science* 355, 942-944.

765 Schmandt, B., Jacobsen, S.D., Becker, T.W., Liu, Z.X., Dueker, K.G., 2014. Dehydration melting at the  
766 top of the lower mantle. *Science* 344, 1265-1268.

767 Shirey, S.B., Wagner, L.S., Walter, M.J., Pearson, D.G., van Keken, P.E., 2021. Slab Transport of Fluids  
768 to Deep Focus Earthquake Depths—Thermal Modeling Constraints and Evidence From Diamonds.  
769 *AGU Advances* 2, e2020AV000304.

770 Smyth, J.R., Frost, D.J., Nestola, F., Holl, C.M., Bromiley, G., 2006. Olivine hydration in the deep upper  
771 mantle: Effects of temperature and silica activity. *Geophys. Res. Lett.* 33.

772 Soltanmohammadi, A., Gregoire, M., Rabinowicz, M., Gerbault, M., Ceuleneer, G., Rahgoshay, M.,  
773 Bystricky, M., Benoit, M., 2018. Transport of Volatile-rich Melt from the Mantle Transition Zone  
774 via Compaction Pockets: Implications for Mantle Metasomatism and the Origin of Alkaline Lavas  
775 in the Turkish-Iranian Plateau. *Journal of Petrology* 59, 2273-2309.

776 Song, T.R.A., Helmberger, D.V., Grand, S.P., 2004. Low-velocity zone atop the 410-km seismic  
777 discontinuity in the northwestern United States. *Nature* 427, 530-533.

778 Stagno, V., Ojwang, D.O., McCammon, C.A., Frost, D.J., 2013. The oxidation state of the mantle and  
779 the extraction of carbon from Earth's interior. *Nature* 493, 84-88.

780 Swain, M.V., Atkinson, B.K., 1978. Fracture surface-energy of olivine. *Pure and Applied Geophysics*  
781 116, 866-872.

782 Tauzin, B., Debayle, E., Wittlinger, G., 2010. Seismic evidence for a global low-velocity layer within  
783 the Earth's upper mantle. *Nat. Geosci.* 3, 718-721.

784 Toffelmier, D.A., Tyburczy, J.A., 2007. Electromagnetic detection of a 410-km-deep melt layer in the  
785 southwestern United States. *Nature* 447, 991-994.

786 Toplis, M.J., 2005. The thermodynamics of iron and magnesium partitioning between olivine and liquid:  
787 criteria for assessing and predicting equilibrium in natural and experimental systems. *Contrib.*  
788 *Mineral. Petr.* 149, 22-39.

789 van Keken, P.E., Hacker, B.R., Syracuse, E.M., Abers, G.A., 2011. Subduction factory: 4. Depth-  
790 dependent flux of H<sub>2</sub>O from subducting slabs worldwide. *J. Geophys. Res.* 116.

791 Vinnik, L., Farra, V., 2007. Low S velocity atop the 410-km discontinuity and mantle plumes. *Earth*  
792 *Planet. Sci. Lett.* 262, 398-412.

793 Wang, X.-C., Wilde, S.A., Li, Q.-L., Yang, Y.-N., 2015. Continental flood basalts derived from the  
794 hydrous mantle transition zone. *Nat. Commun.* 6, 7700.

795 Wen, L.X., Anderson, D.L., 1995. The fate of slabs inferred from seismic tomography and 130 million  
796 years of subduction. *Earth Planet. Sci. Lett.* 133, 185-198.

797 Xiao, J., Hier-Majumder, S., Tauzin, B., Waltham, D., 2020. An inversion approach for analysing the  
798 physical properties of a seismic low-velocity layer in the upper mantle. *Phys. Earth Planet. Inter.*  
799 304.

800 Yang, X., 2016. Effect of oxygen fugacity on OH dissolution in olivine under peridotite-saturated  
801 conditions: An experimental study at 1.5–7GPa and 1100–1300°C. *Geochim. Cosmochim. Acta*  
802 173, 319-336.

803 Yoshino, T., Manthilake, G., Matsuzaki, T., Katsura, T., 2008. Dry mantle transition zone inferred from  
804 the conductivity of wadsleyite and ringwoodite. *Nature* 451, 326-329.

805 Yoshino, T., Nishihara, Y., Karato, S., 2007. Complete wetting of olivine grain boundaries by a hydrous  
806 melt near the mantle transition zone. *Earth Planet. Sci. Lett.* 256, 466-472.

807 Zhang, Z., Dueker, K.G., Huang, H.-H., 2018. Ps mantle transition zone imaging beneath the Colorado  
808 Rocky Mountains: Evidence for an upwelling hydrous mantle. *Earth Planet. Sci. Lett.* 492, 197-  
809 205.

810 Zhao, Y.H., Ginsberg, S.B., Kohstedt, D.L., 2004. Solubility of hydrogen in olivine: dependence on  
811 temperature and iron content. *Contrib. Mineral. Petr.* 147, 155-161.

812