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## A carbonaceous chondrite and cometary origin for icy moons of Jupiter and Saturn

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11 **Abstract**

12         The inner structure of icy moons comprises ices, liquid water, a silicate rocky core and  
13 sometimes an inner metallic core depending on thermal evolution and differentiation.  
14 Mineralogy and density models for the silicate part of the icy satellites cores were assessed  
15 assuming a carbonaceous chondritic (CI) bulk composition and using a free-energy  
16 minimization code and experiments. Densities of other components, solid and liquid sulfides,  
17 carbonaceous matter, were evaluated from available equations of state. Model densities for  
18 silicates are larger than assessed from magnesian terrestrial minerals, by 200 to 600 kg.m<sup>-3</sup> for  
19 the hydrated silicates, and 300 to 500 kg.m<sup>-3</sup> for the dry silicates, due to the high iron bulk  
20 concentration in CI. The stability of Na-phlogopite in the silicate fraction up to 1300K favors  
21 the trapping of most <sup>40</sup>K in the rocky/carbonaceous cores with important consequences for  
22 modeling of the thermal evolution of icy satellites. We find that CI density models of icy  
23 satellite cores taking into account only the silicate and metal/sulfide fraction cannot account  
24 for the observed densities and reduced moment of inertia of Titan and Ganymede without  
25 adding a lower density component. We propose that this low-density component is  
26 carbonaceous matter derived from insoluble organic matter, in proportion of ~30-40% in  
27 volume and 15-20% in mass. This proportion is compatible with contributions from CI and  
28 comets, making these primitive bodies including their carbonaceous matter component likely  
29 precursors of icy moons, and potentially of most of the objects formed behind the snow line of  
30 the solar system.

31

32 **Keywords:** satellite, core composition, Titan, Ganymede, comet, carbonaceous chondrites

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35

## 36 1. Introduction

37

38 Icy satellites are poorly known bodies, with Jupiter's Ganymede and Callisto, and  
39 Saturn's Titan the largest ones, similar in size to Mercury (with radii  $R > 2400$  km) but with  
40 much lower mass  $M$  and mean densities of  $1800\text{-}2000 \text{ kg.m}^{-3}$  consistent with assemblages of  
41 ice and rock in various ratios (Anderson et al., 1996; Anderson et al., 1998). Their reduced  
42 moments of Inertia ( $\text{MoI} = C/MR^2$ , equal to  $2/5$  for homogeneous sphere where  $C$  is the  
43 average moment of inertia relative to the spin axis) differ significantly from 0.311 for  
44 differentiated Ganymede to  $\sim 0.355$  for the least differentiated Callisto, respectively, Titan  
45 being at an intermediate stage of differentiation with a value of 0.3414. We focus here on the  
46 largest icy satellites, Callisto, Ganymede and Titan, and will not discuss the case of Io, which  
47 is totally dehydrated due to high temperatures and volcanic activity, or of smaller Europa.

48 Although quite similar in size and density, Callisto, Ganymede and Titan have  
49 different structures. Callisto and Ganymede present salty internal oceans whose presence is  
50 inferred from the magnetic fields induced by the Jovian environmental plasma (Kivelson et al.,  
51 1996; Kivelson et al., 2002; Zimmer et al., 2000), and similar conclusions were reached for  
52 Titan around Saturn from gravity and radar observations (Baland et al., 2011; Iess et al., 2012).  
53 For Ganymede, the presence of an inner metallic core was inferred from detailed analysis of  
54 its magnetic field (Anderson et al., 1996; Kivelson et al., 2002), a sign of a high degree of  
55 differentiation in agreement with the low MoI. The internal structure of these satellites is  
56 usually described as stratified layers composed of liquid iron sulfide alloy (only for  
57 Ganymede), silicates and iron sulfide mixture, and of ice and liquid water from the innermost  
58 to the outermost shells.

59 Titan's internal structure models match observations with four layers, assuming  
60 hydrated silicate core mineralogy (Castillo-Rogez and Lunine, 2010; Fortes, 2012)

61 surrounded by a high-pressure ice layer, an ocean, and an ice crust (Baland et al., 2011; Iess et  
62 al., 2012). Titan's surface is active with ice mountains, hydrocarbon lakes (Lellouch et al.,  
63 2004; Stofan et al., 2007), and methane atmosphere indicative of internal activity (Tobie et al.,  
64 2006). Callisto is considered as not or only partially differentiated (Mueller and McKinnon,  
65 1988), and surface features do not indicate significant internal activity (Schubert et al., 1981).  
66 Its MoI of 0.3549(42) if hydrostaticity is assumed, but if this assumption is not verified, the  
67 range of possibilities is large for its internal rocky core (Anderson et al., 2001). We therefore  
68 considered only the case of Titan where MoI is determined from a larger number of flybys for  
69 the low-temperature satellites.

70 Ganymede's internal structure models assume at least five layers (Mueller and  
71 McKinnon, 1988; Schubert et al., 1996; Vance et al., 2014) with an iron-rich core at the center,  
72 between 1/4 and 1/3 of the radius depending on sulfur content (Sohl et al., 2002). Silicate  
73 layers surrounding this metallic core have a mineralogy that is dominated by dry silicate  
74 mantle, or by mixed dry/hydrous minerals depending on the thermal model (Scott et al., 2002).  
75 A hydrosphere layer including a ocean sandwiched between a high-pressure ice layer and the  
76 outer ice crust surrounds the silicate-metal core (Grasset et al., 2013).

77 Internal structure and density models are usually based on the assumption that  
78 mineralogy includes only silicates and iron sulfides, and neglect carbonaceous matter present  
79 in primitive material like CI chondrites. Densities of Mg-rich terrestrial hydrated minerals are  
80 used for Titan (Fortes, 2012), while realistic compositions for the icy satellites are likely more  
81 iron-rich (Bland et al., 2004). If iron concentration is a free parameter, it leads to non-  
82 chondritic Fe/Si ratios (Sohl et al., 2002) as it may enter silicates in larger amounts than on  
83 Earth. No formation model accounts for large variations of the Fe/Si ratio inferred in Jovian  
84 satellites (Sohl et al., 2002), nor for the hydrous Mg-silicate composition required for  
85 matching the density and MoI of Titan. Therefore, instead of leaving bulk composition as a

86 free parameter with resulting variations not accounted for by known cosmochemical or  
87 mechanical segregation mechanisms, we chose here to assume a common composition for the  
88 precursors of the rocky cores of icy satellites.

89         We assume a CI carbonaceous chondrite composition for the interiors of icy satellites  
90 (Mueller and McKinnon, 1988), *i.e.* essentially a solar composition except for H, He and the  
91 most volatile components (Lodders, 2003; Wasson and Kallemeyn, 1988), as a common  
92 origin for objects formed mainly beyond the snow line in the solar system. With this  
93 hypothesis, mineral compositions, metal-sulfide-silicates cannot be varied independently  
94 other than by assuming different evolution, in particular the segregation of an inner metallic  
95 core. Silicate mineralogy and density profiles in the icy moons are computed using the  
96 thermodynamic software Perple\_X (Connolly, 1990) at relevant pressures and temperatures.  
97 Preliminary high pressure and temperature experiments are used to check the reliability of the  
98 thermodynamic calculations in iron-rich chondritic compositions of hydrous silicates.  
99 Evolution of organic precursors with bulk chondritic CI composition and their reactivity with  
100 silicates and sulfides were evaluated.

101         Two structural models are built for Titan and Ganymede depending on the degree of  
102 differentiation, resulting in different iron contents in the silicate fraction. Density and MoI  
103 models are constructed with available equations of state of the three main core components,  
104 silicates, iron sulfide and its liquid alloys, and carbonaceous matter derived from insoluble  
105 organic matter (IOM) of CI or comets. Carbonaceous matter proves to be an essential  
106 component for matching density and MoI of the satellites, and is likely derived from cometary  
107 nuclei precursors.

108

## 109         **2. Compositional model**

110

111 CI carbonaceous chondrite composition (Wasson and Kallemeyn, 1988), similar to  
112 that of the solar photosphere (except for volatile elements), was assumed to represent that of  
113 the solar nebula and other primitive objects like comets and interplanetary dust particles,  
114 providing the potential primordial material for formation of dwarf planets, satellites, and  
115 trans-Neptunian objects (TNO) beyond the snow line (McKinnon et al., 2017; Mueller and  
116 McKinnon, 1988).

117 Composition was simplified to major elements O, Fe, Mg, Si, S, Al, Ca, Na (98 wt%)  
118 to perform thermodynamic calculations and experiments in the silicate subsystem. Minor  
119 elements manganese, nickel and chromium (summing to ~1.5wt%) easily substitute to iron  
120 into crystalline structures, and were added to the Fe component ("Fe" = Fe + Mn + Ni + Cr).  
121 Sodium (~0.5%) and potassium (~0.05wt%) also substitute each other in minerals, and the  
122 formation of soluble salts (NaCl and KCl) with chlorine is assumed, which were subtracted  
123 from the "Na" component ("Na" = Na + K - Cl). This accounts for 99.5wt% of the silicates,  
124 and trace elements (~0.5% of the composition) were neglected (Table 1). Slightly different  
125 average CI compositions could be used (Lodders, 2003), with differences between CI  
126 compositions being mainly the sulfur content.

127 From this initial composition, two scenarios of evolution are considered that will result  
128 in different compositions of the rocky cores of icy satellites (Table 1). In model 1, the rocky  
129 core is not differentiated, the temperature remained low enough to avoid sulfide-silicate  
130 separation, the composition of the rock is homogeneous, and identical to that of CI chondrites.  
131 In model 2, the rocky core is differentiated and an inner metallic core segregated in the liquid  
132 state assuming a Fe-FeS eutectic composition (Fei et al., 1997), because segregation takes  
133 place as soon as melting occurs. The resulting composition of the silicate fraction is that of CI  
134 chondrites from which more iron is subtracted than in model 1 to form an inner core with 25

135 wt.% S, corresponding to the eutectic composition at average pressure of about 4 GPa in the  
136 metallic core of Ganymede.

137 Carbonaceous and organic compounds containing C, H, N, O are an important  
138 component of CI, representing up to 4 wt% of the rock (Derenne and Robert, 2010). They are  
139 classically divided into soluble and insoluble organic matter (IOM), the latter representing  
140 about 75% of carbonaceous matter.

141

### 142 **3. Methods**

143

144 The open-access Perple\_X software (Connolly, 1990), for which updates and  
145 benchmarks are regularly provided online [http://www.perplex.ethz.ch/perplex\\_updates.html](http://www.perplex.ethz.ch/perplex_updates.html),  
146 was used to calculate the equilibrium mineral assemblages and compositions for a given bulk  
147 composition varying pressure and temperature. We used thermodynamic properties and solid  
148 solutions calibrated self-consistently on experiments and natural samples (Holland and Powell,  
149 1998), with recent addition of antigorite serpentine (Padron-Navarta et al., 2013), and revised  
150 antigorite serpentine equation of state providing best agreement with high-pressure phase  
151 equilibrium data (Hilairet et al., 2006). Temperatures and pressures vary in the range 300-  
152 1300 K, 0.8-7 GPa, to cover conditions in icy satellites. Excess water is assumed (water  
153 activity fixed to 1) at all pressure-temperature (P-T) conditions to obtain the maximum  
154 stability of hydrated silicates during the early stages of formation of satellites from a water-  
155 rich ice-rock mixture. Drier conditions would result in lowering the dehydration temperatures.  
156 Built files are given in supplementary material for reproduction of calculations.

157 FeS is assumed to be an independent component whose reaction with silicates is  
158 negligible. This is confirmed by experiments (see below) with only minor destabilization of  
159 troilite to form pyrrhotite, whose composition is close to that of troilite, coexisting with



160 silicates. The silicate fraction composition is obtained by removing the troilite component  
161 (Table 1). Organic or carbonaceous matter is assumed to be a chemically independent system  
162 because it does not react significantly with silicates, at least at low temperatures (Scott et al.,  
163 2002). It is only assumed to maintain reducing conditions, so that ferric iron content is  
164 negligible in silicates. In CI chondrites, ferric iron occurs in the form of magnetite ( $\text{Fe}_3\text{O}_4$ ),  
165 iddingsite and cronstedtite (Hyman and Rowe, 1983; King et al., 2015), as a result of  
166 alteration at low temperatures ( $<100^\circ\text{C}$ ). These near surface conditions on CI parent bodies  
167 are not met in the core of icy satellites, and mineralogy has to be reevaluated with  
168 thermodynamic and experimental modeling. Organic compounds may react with silicates at  
169 temperatures above 1000K, but there are too little experimental data thus far to quantitatively  
170 assess this possibility (Scott et al., 2002).

171 Perple\_X has been calibrated on laboratory experiments that were carried out at  
172 temperatures as low 500-600K in fluid saturated conditions akin to those studied here.  
173 Perple\_X provides accurate phase relations that are compatible with observations in natural  
174 rocks from cold subduction settings, and some of the thermodynamic data (Padron-Navarta et  
175 al., 2013) and phase relations used here have been tested on natural low-temperature  
176 serpentines that were formed over periods of a few to a few tens million years. Kinetic  
177 experiments also point to fast reactions at the geological timescale in the 500-900K  
178 temperature range when hydrated Mg-rich minerals (serpentines) and fluids are involved  
179 (Malvoisin et al., 2012). It is therefore reasonable to assume that equilibrium is reached to  
180 some extent even in the cold rocky core of Titan (Castillo-Rogez and Lunine, 2010) with  
181 reactive Fe-rich composition and longer timescales.

182 Since the database used in Perple\_X is calibrated on terrestrial samples, it is desirable  
183 to check whether the extrapolation to extraterrestrial material compositions is accurate. Three  
184 preliminary experiments were performed to complement earlier work (Scott et al., 2002), in

185 order to validate the consistency of the thermodynamic modeling. A starting material was  
186 synthesized by mixing oxide powders in the proportions given in the undifferentiated model  
187 (Table 1), minus FeO and FeS. The mixture was finely ground in an agate mortar, and held  
188 for one hour at 1650 °C in a platinum crucible, quenched, and the procedure repeated. Raman  
189 spectroscopy, scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy  
190 (EDX) mapping showed a chemically homogeneous, fine-grained mixture of olivine and  
191 pyroxene. Pieces of this material were finely crushed in an agate mortar in ethanol before  
192 each experiment for the high-pressure experiments. Iron was added under the form of troilite  
193 FeS and iron oxides, magnetite (Fe<sub>3</sub>O<sub>4</sub>) or wüstite (FeO), in proportions of the CI composition.  
194 Organic matter was added as an aliphatic chain (solid paraffin wax) and a soluble nitrogenous  
195 base (pyridine). The solid mixture was put into a gold capsule along with pyridine-water  
196 solution in proportions matching the N/C ratio of CI chondrites. Capsules are designed to seal  
197 in the press as pressure is increased. In order to ensure that the experimental load is saturated  
198 in water, diamond powder was added on top of the sample to create a non-reactive porous  
199 reservoir. The material was reacted for ~100 hours at a pressure of 1.5 GPa and temperatures  
200 of 773 and 873K in a belt apparatus. Hydrogen or H<sub>2</sub>O losses are negligible at those  
201 conditions, and temperature is known to ±40K (Pilorgé et al. 2017). In the presence of excess  
202 sulfide or organic matter, it is assumed that the sample was self-buffered regarding the oxygen  
203 fugacity (Scott et al., 2002). At the end of each run, the capsules were opened along a  
204 longitudinal plane, trapped in epoxy resin and polished. They were analyzed using SEM,  
205 EDX, X-Ray Diffraction (XRD) and Raman spectroscopy. XRD showed only pyrrhotite-  
206 troilite solid solution, olivine and pyroxene, as well as gold and diamond from the capsule and  
207 trap. Raman spectroscopy was more efficient to detect minor newly formed phases.

208

#### 209 **4. Mineralogical and density calculations**

210

#### 211 **4.1 Mineralogy of the different models**

212 The difference of iron content has two major consequences on the phase diagram of  
213 the two models (Fig. 1). Dehydration occurs over a broader and lower temperature range in  
214 the iron-rich (model 1) than in the iron-poor silicates (model 2). Each diagram can be divided  
215 in three domains upon increasing temperature, firstly the region where mineralogy is  
216 dominated by hydrous minerals, secondly the dehydration region where hydrous minerals  
217 progressively decompose to form dry minerals, and thirdly the region where mineralogy is  
218 dominated by dry minerals.

219 The evolution of the amount of each phase for three isobars (fixed pressure and  
220 varying temperature) at 1.5, 3.0, and 4.5 GPa shows that the effect of pressure on the stability  
221 fields of minerals is relatively small over the studied range (Fig. 2). In the hydrated domain,  
222 mineralogy is dominated by hydrous phyllosilicates (layered clay-like minerals) chlorite,  
223 antigorite, and Na-rich phlogopite  $(\text{Na,K})\text{Mg}_3\text{AlSi}_3\text{O}_{10}(\text{OH,F,Cl})_2$ . Chlorite and antigorite,  
224 common Mg-rich hydrous minerals in hydrated basic to ultrabasic terrestrial rocks, contain ~  
225 12-13 wt.%  $\text{H}_2\text{O}$ , and Na-phlogopite, less common on Earth, contains ~ 4 wt.% of  $\text{H}_2\text{O}$ .

226 Na-phlogopite is important because potassium may substitute for sodium up to 1wt%  
227 in natural crystals (Schreyer et al., 1980), and it is stable up to 1300K. Thus it may trap all the  
228 available potassium into the rocky core, and control the early thermal history of the body  
229 when heat production was dominated by  $^{40}\text{K}$  decay. Finally up to 20% amphibole with ~2.5  
230 wt.%  $\text{H}_2\text{O}$  is formed at low pressures ( $<2.5$  GPa), and remains stable up to high temperatures.  
231 Amphibole is an additional host for trapping potassium in the rocky core of icy satellites.

232 The difference in iron content between models does not significantly modify the  
233 nature of hydrous mineral phases, only their relative amount and composition. Depending on  
234 the iron content of the bulk rock, antigorite and chlorite behave differently. In model 1, the

235 high amount of iron stabilizes a large amount of chlorite at the expense of antigorite, but their  
236 quantities reverse abruptly at ~ 520 K at nearly constant water content in the solid. In model 2,  
237 antigorite is the dominant phase over the whole hydrated assemblage stability field. This  
238 second model is similar to that of hydrated ultramafic rocks in terrestrial subduction zones  
239 (Padron-Navarta et al., 2013) with only slightly lower Mg number ( $Mg\# = Mg/(Mg+Fe)$ ) of  
240 ~0.87 instead of ~0.90 for common terrestrial peridotites.

241 From about 550 to 900K, the amount of dry minerals (e.g. olivine and pyroxene)  
242 increases at the expense of antigorite. The disappearance of antigorite is the leading reaction  
243 of dehydration: it has the strongest effect on the release of water because of high  
244 concentration (~12 wt.% H<sub>2</sub>O) and high abundance. The effect of the iron content on  
245 dehydration is best seen when plotting the water content of the mineral assemblage in the rock  
246 against temperature (Fig. 3). In model 1, about 60 wt.% of hydrous minerals, chlorite and  
247 antigorite, with an iron-rich composition of the silicate fraction (bulk  $Mg\# \sim 0.69$ ) start to  
248 dehydrate around 550 K. This reaction is progressive up to ~820-870 K where the mineralogy  
249 is almost entirely dry, with only Na-phlogopite as a hydrous phase up to ~1150 K. In iron-  
250 poor model 2, antigorite destabilizes over a narrower and higher temperature range from 800  
251 K to 920 K.

252 At high temperatures, the mineralogy is dominated by dry minerals olivine  
253  $(Mg,Fe)_2SiO_4$  and pyroxenes  $Ca(Mg,Fe)Si_2O_6$  and  $(Mg,Fe)_2Si_2O_6$ . Na-phlogopite and  
254 amphiboles are the only hydrated minerals stable at temperatures higher than 900 K. The  
255 difference in composition between the two models does not change the nature of the dry  
256 mineral phases obtained from the thermodynamic model, only their relative amount and  
257 composition. In model 2, orthopyroxene and olivine are almost equally abundant and  
258 dominate the mineralogy. In the iron-rich composition M1, the proportion of olivine at high

259 temperature is much higher than in model M2. Amphiboles are stable at low pressure and  
260 destabilize into garnet at higher pressure, around 3-3.5 GPa in M2, and 2.5-2.8 GPa in M1.

261

## 262 **4.2 Comparison with experiments**

263 XRD analysis of the experimental charges shows the presence of olivine and pyroxene  
264 in addition to products from the capsule and diamond trap, indicating that reaction with water  
265 was sluggish. This technique is unable to inform on the presence of a phase it is present in  
266 less than 5 % or has a low scattering factor. Raman spectroscopy analysis was conducted to  
267 detect minor phases. It revealed the presence of Na-phlogopite ( $\text{NaMg}_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2$ ) (Fig.  
268 4), one of the phyllosilicates predicted with Perple\_X calculations. The absence of chlorite at  
269 773 K is in agreement with the Perple\_X predictions. The absence of antigorite suggests that  
270 our experiment was performed above the destabilization temperature of antigorite, within the  
271 error bar of the experiment (Fig. 2, M1 model).

272 Olivine and pyroxene are the major phases of the recovered samples, and are always  
273 enriched in Fe with respect to the Fe-free olivine and pyroxene of the starting material. No  
274 residual grains of the starting magnetite ( $\text{Fe}_3\text{O}_4$ ) or wüstite ( $\text{FeO}$ ) are recovered, as they  
275 dissolved into the silicate phases. Olivines adjacent to FeS precursor grains are enriched in  
276 iron from  $\text{Fo}_{81}$  for large patches of crystals to  $\text{Fo}_{32}$  in the most Fe-rich recrystallizations close  
277 to troilite grains partially transformed into pyrrhotite. Similar patterns of Fe-enrichment were  
278 observed on pyroxene grains near sulfide. Troilite reacted to form pyrrhotite ( $\text{Fe}_{1-x}\text{S}$ ,  $x < 1/8$ )  
279 identified by Raman and XRD, accounting for iron reduction in the oxide subsystem with a  
280 global reaction  $\text{FeS} + x \text{Fe}_3\text{O}_4 = \text{Fe}_{1-x}\text{S} + 4x \text{FeO}$ . The replacement of troilite (FeS) by  
281 pyrrhotite contributes to local (typically 10 micron scale) iron enrichment in the olivine and  
282 pyroxene, as a minor addition to iron oxide dissolution. This indicates that global dissolution

283 of iron oxide in silicates took place, and that at least local equilibrium is reached at such low  
284 temperatures over the experiment duration.

285 Former experiments using CI starting composition (Scott et al., 2002) found that  
286 antigorite and chlorite are stable at low temperatures, in consistency with Perple\_X  
287 predictions, but that talc forms at high temperatures, in contradiction with it. Identification of  
288 talc as dominant hydrous phases at high temperatures in their experiments relies partly on  
289 infrared (IR) spectroscopy. IR spectra only identifies vibrational modes associated with  
290 tetrahedral-octahedral-tetrahedral (TOT) layers of phyllosilicates that are common to talc and  
291 Na-phlogopite, with a possibly ambiguous determination. Raman spectroscopy allows  
292 unambiguous determination of the newly formed phyllosilicate as Na-phlogopite with  
293 characteristic Raman peaks including TOT and OH vibrations (Fig. 4), distinct from those of  
294 talc.

295 In spite of the need of further experimental effort to resolve potential issues with  
296 thermodynamic equilibrium and reaction between organic matter and silicates, the  
297 thermodynamic predictions from Perple\_X are consistent with major features of experimental  
298 data indicating dominance of antigorite and chlorite at temperatures in the range 673-873K  
299 (Scott et al., 2002) and formation of Na-phlogopite at  $773\pm 40$  K (this study).

300 Raman spectra of the organic matter show that initial components transformed into  
301 amorphous carbonaceous matter akin to IOM or coals (Quirico et al., 2009), as expected at 1.5  
302 GPa and 773-873K (Chanyshv et al. 2017). This transformation occurs with partial loss of  
303 nitrogen, oxygen and hydrogen from the paraffin and pyridine precursors. The release of  
304 hydrogen may have buffered the oxidation state to ferrous iron, favoring dissolution of iron  
305 oxides in the silicates. Apart from buffering the oxidation state, no reaction between  
306 carbonaceous matter and silicates was observed. Potential reactions between silicates,

307 sulfides and carbonaceous matter at temperature above 1000K (Scott et al., 2002) are not well  
 308 documented and require further investigation.

309

### 310 **4.3 Density profiles and equations of state of silicate-sulfide layers**

311 From the mineral proportions and compositions, density models are computed using  
 312 available equations of state in the Perple\_X database. Silicate and oxide densities are known  
 313 to within 1%. The density of the FeS phase is given by a Murnaghan equation of state:

$$\rho(P, T) = \rho_0 \left[ 1 + P \left( \frac{K'_0}{K_0} \right) \right]^{1/K'_0} (1 - \alpha(T - T_0))$$

314 (1)

315 with:

$$316 \quad K_0 = K_0(T_0) + \left( \frac{\partial K_0}{\partial T} \right)_P (T - T_0) \quad (2)$$

317 where  $K_0$  and  $K'_0$  are the bulk modulus and its pressure derivative, respectively, and  $\alpha$  the  
 318 thermal expansivity. The density of the silicate – sulfide assemblage was calculated from  
 319 volume conservation on mixing:

$$320 \quad \rho_{mix} = \frac{\rho_{silicate} \rho_{FeS}}{\rho_{silicate} \omega_{FeS} + \rho_{FeS} \omega_{silicate}} \quad (3)$$

321 with  $\rho_{mix}$  the density of the rock-metal mixture,  $\rho_{silicate}$  the density of the silicate predicted with  
 322 model 1,  $\rho_{FeS}$  the density of the troilite phase (FeS),  $\omega_{FeS}$  the mass fraction of the troilite in the  
 323 rock-metal mixture,  $\omega_{silicate}$  the mass fraction of the silicate in the rock-metal mixture.

324 Parameters of equation (1) are  $T_0 = 1000\text{K}$ ,  $K_0 = 54.3 \text{ GPa}$ ,  $K'_0 = 4$ ,  $\partial K_0/\partial T = -0.0117 \text{ GPa/K}^{-1}$ ,  
 325  $\alpha = 1.042 \cdot 10^{-4} \text{ K}^{-1}$ , and  $\rho_0 = 4678 \text{ kg/m}^3$  (Urakawa et al., 2004). Pyrrhotite was not considered  
 326 although it forms at the expense of troilite at low temperatures. Troilite was used as a proxy to

327 pyrrhotite, whose equation of state is unknown, because of its similar composition and density,  
328 with an estimated uncertainty of 2%.

329 Assuming that the temperature of the core remained above the eutectic melting around  
330  $1225\pm 10$  K, average densities for the eutectic liquid with 25 wt% (or 37 mol%) S at 4 GPa are  
331 estimated as  $5700\pm 100$  kg.m<sup>-3</sup> (Morard et al., 2018). An inner core mass fraction of 35% is  
332 fixed by the eutectic and chondritic compositions (Table 1), yielding a radius fraction of  
333 61.7% and volume fraction of 23.5 % for the inner liquid core. For the sake of MoI modeling,  
334 liquid densities were obtained by fitting third-order Birch-Murnaghan equation of state  
335 (Morard et al., 2018) for sulfur content  $X$  between 0 and 50 mol% to a simplified Murnaghan  
336 type equation of state similar to equation (1) with  $\rho_{\square}(X) = 6950 - 5176 X - 3108 X^2$  in kg/m<sup>3</sup>,  
337  $T_0 = 1900$ K,  $K_0(X) = 82.6^{(1-X)} \times 1.84^X$  GPa,  $K'_0 = 5.9$ , and  $\alpha = 1.08 \cdot 10^{-4}$  K<sup>-1</sup>. The large  
338 composition range of the experimental data allows interpolation to the eutectic composition.  
339 The Murnaghan equation of state has the advantage of allowing direct calculation of density  
340 as a function of pressure over the Birch-Murnaghan equation of state.

341 Model 1 consists in a partially differentiated body, in which the hydrosphere was  
342 separated from the silicate-troilite assemblage forming a homogeneous rocky core with CI  
343 iron content in the silicate fraction. At temperatures below 900 K, the hydrated silicate  
344 assemblage has a density of  $\sim 3000$  kg.m<sup>-3</sup> (Fig. 5), which is higher than the estimations of  
345  $\sim 2600$ - $2700$  kg.m<sup>-3</sup> in previous studies for Titan (Castillo-Rogez and Lunine, 2010; Fortes,  
346 2012), because of higher iron content in the present model. On increasing temperature, the  
347 density of the solids increases step by step due to the dehydration of various hydrous minerals  
348 until the temperature reaches  $\sim 900$  K where density takes the steady value of the dry  
349 assemblage. When troilite is mechanically mixed with silicates, bulk density increases by  
350 about  $300$  kg.m<sup>3</sup> at 300 K and  $220$  kg.m<sup>3</sup> at 1300 K (green curve, Fig. 5a).



351 Model 2 represents a partially differentiated body, in which an inner core was formed  
352 with the composition of the Fe-FeS eutectic. The lower iron content of the silicate fraction for  
353 this model, Mg#=87 instead of 69 in model 1, results in lower densities (Fig. 5). The narrow  
354 dehydration zone results in a steep density increase in the 800-950 K range. At higher  
355 temperatures, density takes values of ~3150 to 3350 kg.m<sup>-3</sup> depending on pressure from 1 to 5  
356 GPa. Density increases by ~200 kg.m<sup>-3</sup> over the whole pressure range, and a kink is observed  
357 around 3 GPa due to destabilization of Na-phlogopite and amphibole into garnet.

358

#### 359 **4.4 Density of carbonaceous matter**

360 Carbonaceous matter is the third potential major component in CI composition  
361 metamorphosed to the conditions of icy satellite cores. There exist limited data on the density  
362 of such compounds, especially at high pressure, because they are mostly amorphous. Given  
363 the molecular complexity of IOM (Derenne and Robert, 2010), we estimated its density after  
364 thermal metamorphism assuming it is similar to that of carbonized coals (Franklin, 1949).  
365 Starting from values of 1300-1400 kg/m<sup>3</sup>, coal density rises slowly to reach 1400-1500 kg/m<sup>3</sup>  
366 after heating to 873K before increasing abruptly to values of ~1800±100 kg/m<sup>3</sup> in the 1123-  
367 1373K range, finally raising slowly to ~2000-2100 kg/m<sup>3</sup> at 1900K, i.e. close to that of  
368 graphite (~2300 kg/m<sup>3</sup>). Densities may vary with the origin of the coal, and the strong  
369 increase in density around 900 K is associated with loss of volatiles, mostly hydrogen  
370 (Franklin, 1949). Smoother increase above 1000K is attributed to increasing crystallinity.  
371 Decomposition of various complex organic molecules into amorphous carbonaceous matter  
372 and hydrogen occurs at temperatures of 800-900K, similar to hydrogen loss in coals. This  
373 carbonization temperature is insensitive to pressure up to 10 GPa (Chanyshv et al., 2017).  
374 We assume that similar compaction will occur with increasing temperature in the whole depth  
375 range of icy satellite cores. For the sake of simplicity, we assumed a density of 1400 kg/m<sup>3</sup> for

376 temperature up to 900K, i. e. when silicate mineralogy is dominated by hydrous phases, and  
377 of 1800 kg/m<sup>3</sup> above, i. e. when dry silicates are formed, based on carbonized coals  
378 measurements (Franklin, 1949), taking into account an uncertainty of  $\pm 200$  kg/m<sup>3</sup> in inferring  
379 the required fraction of carbonaceous matter.

380

## 381 **5. Discussion**

382

### 383 **5.1 Mass-MoI relationship for CI silicate-sulfide models**

384 To our knowledge, models of the density of icy satellites have thus far been built  
385 based on assemblages of ices and water, silicates and iron sulfur alloys. Other components  
386 were not explicitly considered in the calculation even though organic matter is mentioned in  
387 the composition of Titan's core (Fortes, 2012). We first test this hypothesis with the density  
388 profiles for the CI composition model for silicate-iron sulfide cores. This assumption  
389 determines the density profiles of the inner core and silicate layer, and their relative radii in  
390 model 2, and the silicate-sulfide proportion in model 1.

391 The temperature and density profiles in the hydrosphere for Ganymede are taken from  
392 the literature (Vance et al., 2014). The choice of the ice crust thickness is of second order in  
393 terms of the change in mass and MoI compared to the effect of iron on the density of the core.  
394 For Titan, the ice crust thickness is fixed at 76 km (Béghin et al., 2012), the temperature  
395 profile is conductive in the ice crust, adiabatic in the ocean and follows the melting curve in  
396 the high-pressure (HP) ice layer where the boundary with the ocean is a temperate layer  
397 (Kalousová and Sotin, 2018) at the melting temperature of ice. The temperature in the HP ice  
398 layer may be 10 to 20 K lower than the melting temperature, which results in a negligible  
399 density change. Heat transfer by conduction in the silicate shell is calculated using spherical  
400 solution of a volumetrically heated shell., and the density profile in the hydrosphere is  
401 computed using the equation of state for water and ice (Vance et al., 2014). The densities of

402 the crust, ocean, and HP ice vary from 0.832 to 0.928, 1.046 to 1.2038, and 1.3377 to 1.3770,  
403 respectively. The gravity acceleration profile depends on the density profile, which is not  
404 known a priori. Therefore, we use an iterative process to determine the gravity acceleration.  
405 The process converges after the second iteration.

406 The internal structure of Ganymede consists of as much as five layers, an iron-rich  
407 liquid inner core, a silicate layer, a high-pressure ice layer, an ocean, and an ice crust  
408 (Anderson et al., 1996; Schubert et al., 1996). In order to segregate an inner metallic core, the  
409 melting temperature of the Fe-FeS eutectic has been reached through the whole volume of  
410 silicates, i. e. 1200-1300K (Fei et al., 1997). This is well above the dehydration temperature of  
411 major hydrous silicates (Figs. 1 and 2). The mean density of the silicate layer is taken as that  
412 of the nearly entirely dehydrated assemblages for model 2 (3250-3300 kg.m<sup>-3</sup>), potential  
413 hydrous phases being limited to Na-phlogopite and amphibole stable above 900 K (Fig. 2).  
414 Beneath the hydrosphere, there might be a thin hydrated layer due to water rock interactions  
415 near the ice-silicate core interface surrounding the dry silicate layer, and the inner metallic  
416 core with the composition of the Fe-FeS eutectic (Fig. 6). For the sake of simplicity, the small  
417 hydrated silicate layer was not considered in modeling the mass-MoI relationship. As  
418 mentioned above, compositional constrains give a relative radius of 62.5 % for the metallic  
419 inner core. With an estimated ice thickness of 800 km (Vance et al., 2014), this gives a core  
420 radius of 1830 km and an inner metallic core radius of 1140 km, compatible with former  
421 estimates in the 650-1300 km range (Anderson et al., 1996).

422 With its MoI of 0.3414 (Iess et al., 2012), Titan is differentiated, but not as much as  
423 Ganymede. Inner temperatures are low (Fortes, 2012), there is no evidence of a metallic inner  
424 core, and model 1 is applied with a silicate-troilite core composed of hydrous silicates with an  
425 average density of ~3300 kg.m<sup>-3</sup>. This high value is due to the high iron content in chondritic  
426 hydrous and anhydrous silicates with respect to terrestrial value commonly used in modeling

427 of 2520-2670 kg.m<sup>-3</sup> (Castillo-Rogez and Lunine, 2010; Fortes, 2012), and to the addition of  
428 troilite.

429         Integration of density profiles yields the relationship between MoI and mass for model  
430 1 Titan and model 2 Ganymede (Fig. 7) within a fixed radius geometry and CI composition,  
431 and varying the depth of the high-pressure ice-core limit (Table 2). This is equivalent to  
432 varying the bulk water/rock ratio of the satellite. It is clear that none of the CI chondrite  
433 models taking into account only silicates and iron sulfides for the rocky cores can account for  
434 the observed values on Titan and Ganymede (Fig. 7). Considering non-hydrostaticity on Titan  
435 based on the presence of topographic depressions at the poles (Sotin et al., 2018) brings the  
436 MoI value closer to the mass-MoI relationship of the models but is far from solving the  
437 problem. Non-hydrostaticity on Ganymede cannot be determined as only one gravity  
438 coefficient was determined. One way to solve this issue is to reduce the density of the rocky  
439 cores by adding a low-density component to the CI chondrite silicate-sulfide assemblage, with  
440 IOM as a likely low-density candidate.

441

## 442         **5.2 Carbonaceous matter as the required low density component**

443         There exist several solutions to the issue raised in the previous section. A common  
444 one is to infer the density of minerals in the rocky core by assuming values measured on  
445 terrestrial samples. A typical example is serpentine for which zero pressure densities of 2500-  
446 2650 kg.m<sup>-3</sup> are used (Castillo-Rogez and Lunine, 2010; Fortes, 2012). The composition of  
447 terrestrial serpentines is, however, very magnesian (Mg# = 90-93), whereas actual values in  
448 CI are iron-rich with a important cronstedtite component, and with additional magnetite  
449 (Bland et al., 2004). Assuming terrestrial serpentine composition is therefore not justified  
450 because it would require either strong chemical heterogeneities in the region of formation of

451 icy moons, or strong partitioning of iron to a metallic inner core in a CI composition model, at  
452 temperatures incompatible with the stability of hydrous phases (model 2).

453 An alternative approach consists in relaxing the chondritic composition constrain, and  
454 varying major element ratios, in particular the Fe/Si ratio (Sohl et al., 2002). These authors  
455 found that Fe/Si ratios of the Jovian satellites have to be significantly superchondritic for  
456 Callisto and Ganymede, and chondritic to infrachondritic for Europe and Io in order to fit  
457 mass-MoI data with simplified mineralogical models. This hypothesis implies an unknown  
458 mechanism for building chemically distinct regions around Jupiter (Sohl et al., 2002).  
459 Assemblages of rock and ice have also been advocated to explain low densities in satellite  
460 cores, but keeping ice in icy moon cores may prove difficult because of heating and melting  
461 (Fortes, 2012).

462 As an alternative, we test the hypothesis that carbonaceous matter derived from IOM  
463 of CI chondrites provides the missing low-density phase required to match the MoI and mass  
464 observations for Titan and Ganymede. Carbonaceous matter has not been considered  
465 explicitly in former models of inner structure of icy moons, possibly because carbon is  
466 considered a volatile element especially when oxidized.

467 Carbonaceous matter, coals or IOM with relatively low heteroatom (H, O, N) contents  
468 turn out to be refractory phases by losing heteroatoms upon heating with a sharp density  
469 increase near 900K (Franklin, 1949), and resist thermal metamorphism in chondrites (Quirico  
470 et al., 2009). Crystals of large organic molecules transform to carbonaceous matter and  
471 hydrogen over a small temperature range of 800-900K at pressures encountered in icy  
472 satellites (Chanyshv et al., 2017). Also, the formation of carbonaceous matter is reported in  
473 terrestrial subduction metamorphism at temperature around 800-900K and pressure of 2-2.5  
474 GPa (Vitale Brovarone et al., 2017). Thus formation of amorphous then graphitic carbon and  
475 finally graphite upon increasing temperature from organic precursors is the rule rather than

476 the exception in experimental and natural systems, supporting the idea that carbonaceous  
477 matter was preserved during the formation and evolution of bodies as large as  $\sim 2500$  km in  
478 radius. Carbon could also be incorporated in iron alloys to form Ganymede's inner core.  
479 Solubility data in complex Fe-S-Si-C composition suggest little carbon is incorporated in  
480 liquid metal at low pressure and temperature (Deng et al., 2013). It was thus assumed that the  
481 inner core is carbon-free.

482 As discussed earlier, we can distinguish three regions with specific densities of  
483 carbonaceous matter (Franklin, 1949). Firstly a low temperature region corresponding to that  
484 where hydrous silicates are stable, and the density of carbonaceous matter is  $\sim 1400 \text{ kg.m}^{-3}$  for  
485 application to Titan. Secondly a medium temperature region where silicates are mostly dry,  
486 where the density of carbonaceous matter is  $\sim 1800 \text{ kg.m}^{-3}$  for application to Ganymede. Using  
487 these values, we estimated the amount of carbonaceous matter by fixing the mass and radius  
488 to observed values, and using a mean density for the silicate and sulfide fractions of the core.  
489 Carbonaceous matter is added over the whole core in Titan, and only in the silicate portion in  
490 Ganymede (percentage is given below with respect to the whole mass of the rocky core,  
491 including silicate and metal). This second hypothesis has a small effect when compared with  
492 variability of carbonaceous matter densities (Franklin, 1949), estimated here by setting a large  
493 uncertainty of  $\pm 200 \text{ kg.m}^{-3}$ . Taking into account uncertainties on silicate-sulfide assemblages  
494 and carbonaceous matter, and a  $100 \text{ kg.m}^{-3}$  uncertainty on core density, Titan mass and MoI  
495 can be explained with CI chondrite composition by adding  $\sim 23 \pm 10$  wt.% (or  $\sim 45\%$  in volume)  
496 of carbonaceous matter (Fig. 7), and  $17 \pm 10$  wt.% ( $\sim 30\%$  volume) if the MoI is corrected for  
497 non-hydrostaticity estimated from the ellipticity of Titan shape (Sotin et al., 2018). A  
498 remarkably similar amount ( $16 \pm 10$  wt.%) is required to bring the MoI of Ganymede to the  
499 observed value. This yields a molar C/Si ratio  $\sim 4 \pm 2.5$ . These results are much higher than the  
500 organic matter content of 4 wt.% in CI (Derenne and Robert, 2010), or C/Si  $\sim 0.6-1$  (Lodders,

501 2003), but consistent with the high concentration of organics and carbonaceous matter of  
502 comets (Bardyn et al., 2017; Fulle et al., 2016). Comets may represent an additional precursor  
503 to icy satellites, with mineral phases akin to those of chondrites (Brownlee et al., 2006) but  
504 higher organic matter concentrations (Bardyn et al., 2017; Fulle et al., 2016). Degassing of  
505 light hydrocarbons from this primitive material would be consistent with a primordial origin  
506 of Titan atmosphere (Mousis et al., 2009).

507

## 508 **6. Conclusions**

509 We have evaluated the consequences of chemical and mineralogical composition  
510 hypotheses on the internal structure of icy satellites. Within the carbonaceous chondrite  
511 composition model, mass and MoI of Ganymede, a hot and differentiated satellite with mostly  
512 dehydrated core, and of Titan, a cold and mildly differentiated satellite with mostly hydrated  
513 core, can be accounted for provided a low density component is added to classical models to  
514 compensate for the high-density of iron-bearing silicates.

515 This low-density component is refractory carbonaceous matter deriving from IOM of  
516 the CI chondrites or comets, in proportions that match observed concentrations of the latter.  
517 High organic contents have already been assumed in modeling of for the Pluto-Charon system  
518 (McKinnon et al., 2017) based on cometary precursor compositions (Bardyn et al., 2017;  
519 Fulle et al., 2016). More recently, it was found that Ceres surface would contain up to 20 wt%  
520 of organic matter (Marchi et al., 2019). Like the large icy moons Ganymede and Titan, a  
521 number of water rich bodies may have formed from a cosmochemical reservoir defined by  
522 high-organic-content comets and carbonaceous chondrites beyond the snow line of the solar  
523 system.

524 A second consequence of the CI composition model is that large amounts of  $^{40}\text{K}$  can  
525 be stored in the rocky cores of icy satellites in Na-phlogopite and amphiboles, with important

526 consequences on their thermal evolution and dynamics. Solid-state convection in the rocky  
527 core may be needed to extract excess heat, and conditions for establishing convection have to  
528 be assessed.

529         The conclusions reached here are based on the assumptions that carbonaceous matter  
530 and silicates/iron sulfide do not react, an hypothesis that seems valid up to moderate  
531 temperatures of ~900 K but may fail at higher temperatures where carbonate formation was  
532 observed (Scott et al., 2002).

533         The ESA JUICE mission to be launched in 2022 will orbit Ganymede to provide the  
534 degree 2 gravity coefficients with orders of magnitude better accuracy, higher degree gravity  
535 coefficients, higher degree magnetic coefficients, and time-dependent (tidal) topography.  
536 With these measurements, the thickness of the different layers, the non-hydrostatic effects and  
537 the values for the density of the ocean if it exists (Grasset et al., 2013) will be precisely  
538 determined. The JUICE mission will also perform a dozen flybys of Callisto, from which  
539 interior models will be constrained although not as well as for Ganymede.

540         Finally, the recently selected NASA New Frontiers 4 Dragonfly mission includes a  
541 seismometer (Lorenz et al., 2019) that will improve our knowledge of Titan's interior  
542 structure. All these data will provide further tests for the presence of a large fraction of  
543 organic matter in the interior of the large icy moons.

544

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556

## 557 **References**

558

- 559 Anderson, J.D. et al., 2001. Shape, Mean Radius, Gravity Field, and Interior Structure of Callisto. *Icarus*, 153:  
560 157-161.
- 561 Anderson, J.D., Lau, E.L., Sjogren, W.L., Schubert, G., Moore, W.B., 1996. Gravitational constraints on the  
562 internal structure of Ganymede. *Nature*, 384: 541-543.
- 563 Anderson, J.D. et al., 1998. Distribution of rock, metals, and ices in Callisto. *Science*, 280: 1573-6.
- 564 Baland, R.M., Van Hoolst, T., Yseboodt, M., Karatekin, O., 2011. Titan's obliquity as evidence of a subsurface  
565 ocean? *Astronomy & Astrophysics*, 530: A141.
- 566 Bardyn, A. et al., 2017. Carbon-rich dust in comet 67P/Churyumov-Gerasimenko measured by  
567 COSIMA/Rosetta. *Monthly Notices of the Royal Astronomical Society*, 469: S712-S722.
- 568 Béghin, C. et al., 2012. Analytic theory of Titan's Schumann resonance: Constraints on ionospheric conductivity  
569 and buried water ocean. *Icarus*, 218: 1028-1042.
- 570 Bland, P.A., Cressy, G., Menzies, O.N., 2004. Modal mineralogy of carbonaceous chondrites by X-ray  
571 diffraction and Mössbauer spectroscopy. *Meteoritics & Planetary Science*, 39: 3-16.
- 572 Brownlee, D. et al., 2006. Comet 81P/Wild 2 under a microscope. *Science*, 314: 1711-1716.
- 573 Castillo-Rogez, J.C., Lunine, J.I., 2010. Evolution of Titan's rocky core constrained by Cassini observations.  
574 *Geophysical Research Letters*, 37: L20205.
- 575 Chanyshv, A.D. et al., 2017. Transition from melting to carbonization of naphthalene, anthracene, pyrene and  
576 coronene at high pressure. *Physics of the Earth and Planetary Interiors*, 270: 29-39.
- 577 Connolly, J.A.D., 1990. Multivariable phase diagrams; an algorithm based on generalized thermodynamics.  
578 *American Journal of Science*, 290: 666-718.
- 579 Deng, L., Fei, Y., Liu, X., Gong, Z., Shahar, A., 2013. Effect of carbon, sulfur and silicon on iron melting at  
580 high pressure: Implications for composition and evolution of the planetary terrestrial cores. *Geochimica  
581 et Cosmochimica Acta*, 114: 220-233.
- 582 Derenne, S., Robert, F., 2010. Model of molecular structure of the insoluble organic matter isolated from  
583 Murchison meteorite. *Meteoritics & Planetary Science*, 45: 1461-1475.
- 584 Fei, Y., Bertka, C.M., Finger, L.W., 1997. High-Pressure Iron-Sulfur Compound, Fe<sub>3</sub>S<sub>2</sub>, and Melting Relations  
585 in the Fe-FeS System. *Science*, 275: 1621-1623.
- 586 Fortes, A.D., 2012. Titan's internal structure and the evolutionary consequences. *Planetary and Space Science*,  
587 60: 10-17.
- 588 Franklin, R.E., 1949. A study of the fine structure of carbonaceous solids by measurements of true and apparent  
589 densities. Part II.—carbonized coals. *Transactions of the Faraday Society*, 45: 668-682.
- 590 Fulle, M. et al., 2016. Comet 67P/Churyumov-Gerasimenko preserved the pebbles that formed planetesimals.  
591 *Monthly Notices of the Royal Astronomical Society*, 462: S132-S137.
- 592 Grasset, O. et al., 2013. JUPITER ICy moons Explorer (JUICE): An ESA mission to orbit Ganymede and to  
593 characterise the Jupiter system. *Planetary and Space Science*, 78: 1-21.
- 594 Hilairet, N., Daniel, I., Reynard, B., 2006. Equation of state of antigorite, stability field of serpentines, and  
595 seismicity in subduction zones. *Geophysical Research Letters*, 33: L02302.
- 596 Holland, T.J.B., Powell, R., 1998. An internally consistent thermodynamic data set for phases of petrological  
597 interest. *Journal of Metamorphic Geology*, 16: 309-343.
- 598 Hyman, M., Rowe, M.W., 1983. Magnetite in CI chondrites. *Journal of Geophysical Research*, 88: A736.
- 599 Iess, L. et al., 2012. The tides of Titan. *Science*, 337: 457-9.
- 600 Kalousová, K., Sotin, C., 2018. Melting in High-Pressure Ice Layers of Large Ocean Worlds—Implications for  
601 Volatiles Transport. *Geophysical Research Letters*, 45: 8096-8103.
- 602 King, A.J., Schofield, P.F., Howard, K.T., Russell, S.S., 2015. Modal mineralogy of CI and CI-like chondrites  
603 by X-ray diffraction. *Geochimica et Cosmochimica Acta*, 165: 148-160.
- 604 Kivelson, M.G. et al., 1996. Discovery of Ganymede's magnetic field by the Galileo spacecraft. *Nature*, 384:  
605 537-541.

606 Kivelson, M.G., Khurana, K.K., Volwerk, M., 2002. The Permanent and Inductive Magnetic Moments of  
607 Ganymede. *Icarus*, 157: 507-522.

608 Lellouch, E., Schmitt, B., Coustenis, A., Cuby, J.G., 2004. Titan's 5-micron lightcurve. *Icarus*, 168: 209-214.

609 Lodders, K., 2003. Solar System Abundances and Condensation Temperatures of the Elements. *The*  
610 *Astrophysical Journal*, 591: 1220-1247.

611 Lorenz, R.D. et al., 2019. Titan Seismology with Dragonfly : Probing the Internal Structure of the Most  
612 Accessible Ocean World, 50th Lunar and Planetary Science Conference, Houston, pp. 2173.

613 Malvoisin, B., Brunet, F., Carlut, J., Roumejon, S., Cannat, M., 2012. Serpentinization of oceanic peridotites: 2.  
614 Kinetics and processes of San Carlos olivine hydrothermal alteration. *Journal of Geophysical Research-*  
615 *Solid Earth*, 117: B04102.

616 Marchi, S. et al., 2019. An aqueously altered carbon-rich Ceres. *Nature Astronomy*, 3: 140-145.

617 McKinnon, W.B. et al., 2017. Origin of the Pluto–Charon system: Constraints from the New Horizons flyby.  
618 *Icarus*, 287: 2-11.

619 Morard, G. et al., 2018. Liquid properties in the Fe-FeS system under moderate pressure: Tool box to model  
620 small planetary cores. *American Mineralogist*, 103: 1770-1779.

621 Mousis, O. et al., 2009. A primordial origin for the atmospheric methane of Saturn’s moon Titan. *Icarus*, 204:  
622 749-751.

623 Mueller, S., McKinnon, W.B., 1988. Three-layered models of Ganymede and Callisto: Compositions, structures,  
624 and aspects of evolution. *Icarus*, 76: 437-464.

625 Padron-Navarta, J.A. et al., 2013. Tschermak's substitution in antigorite and consequences for phase relations  
626 and water liberation in high-grade serpentinites. *Lithos*, 178: 186-196.

627 Quirico, E. et al., 2009. Precursor and metamorphic condition effects on Raman spectra of poorly ordered  
628 carbonaceous matter in chondrites and coals. *Earth and Planetary Science Letters*, 287: 185-193.

629 Schreyer, W., Abraham, K., Kulke, H., 1980. Natural sodium phlogopite coexisting with potassium phlogopite  
630 and sodian aluminian talc in a metamorphic evaporite sequence from Derrag, Tell Atlas, Algeria.  
631 *Contributions to Mineralogy and Petrology*, 74: 223-233.

632 Schubert, G., Stevenson, D.J., Ellsworth, K., 1981. Internal structures of the Galilean satellites. *Icarus*, 47: 46-  
633 59.

634 Schubert, G., Zhang, K., Kivelson, M.G., Anderson, J.D., 1996. The magnetic field and internal structure of  
635 Ganymede. *Nature*, 384: 544-545.

636 Scott, H.P., Williams, Q., Ryerson, F.J., 2002. Experimental Constraints on the Chemical Evolution of Icy  
637 Satellites. *Earth and Planetary Science Letters*, 203: 399-412.

638 Sohl, F., Spohn, T., Breuer, D., Nagel, K., 2002. Implications from Galileo Observations on the Interior  
639 Structure and Chemistry of the Galilean Satellites. *Icarus*, 157: 104-119.

640 Sotin, C. et al., 2018. Titan's interior structure after Cassini/Huygens, European Planetary Science Congress.  
641 EGU, pp. EPSC2018-1203.

642 Stofan, E.R. et al., 2007. The lakes of Titan. *Nature*, 445: 61-4.

643 Tobie, G., Lunine, J.I., Sotin, C., 2006. Episodic outgassing as the origin of atmospheric methane on Titan.  
644 *Nature*, 440: 61-64.

645 Urakawa, S. et al., 2004. Phase relationships and equations of state for FeS at high pressures temperatures and  
646 implications for the internal structure of Mars. *Physics of the Earth and Planetary Interiors*, 143: 469-  
647 479.

648 Vance, S., Bouffard, M., Choukroun, M., Sotin, C., 2014. Ganymede's internal structure including  
649 thermodynamics of magnesium sulfate oceans in contact with ice. *Planetary and Space Science*, 96: 62-  
650 70.

651 Vitale Brovarone, A. et al., 2017. Massive production of abiotic methane during subduction evidenced in  
652 metamorphosed ophicarbonates from the Italian Alps. *Nature Communications*, 8: 14134.

653 Wasson, J.T., Kallemeyn, G.W., 1988. Composition of Chondrites. *Philosophical Transactions of the Royal*  
654 *Society of London. Series A, Mathematical and Physical Sciences*, 325: 535-544.

655 Zimmer, C., Khurana, K.K., Kivelson, M.G., 2000. Subsurface Oceans on Europa and Callisto: Constraints from  
656 Galileo Magnetometer Observations. *Icarus*, 147: 329-347.

657

658 **Figure captions**

659

660 Fig. 1. Phase diagram for composition models 1 and 2. The grey area shows the major  
661 dehydration interval, lines correspond to major mineral reactions. The gray area is the  
662 dehydration zone that marks the transition between essentially hydrated mineral  
663 assemblage at low temperature and essentially anhydrous mineral assemblage at high  
664 temperature. Model 1 has a broader and lower temperature dehydration zone than model 2.  
665 Amph: amphibole, Atg: antigorite, Br: Brucite, Chl: chlorite, Cpx: clinopyroxene, Gt:  
666 garnet, NaPh: Na-phlogopite, Ol: olivine, Opx: orthopyroxene.

667 Fig. 2. Phase proportions in models 1 and 2 and for three isobars (fixed pressure and  
668 varying temperature). The magnesium-rich model 2 favors the formation of antigorite in  
669 the hydrous part of the diagram at the expense of chlorite, and the formation of  
670 orthopyroxene in the anhydrous part at the expense of olivine, with respect to model 1.  
671 Amph: amphibole, Atg: antigorite, Br: Brucite, Chl: chlorite, Cpx: clinopyroxene, Gt:  
672 garnet, NaPh: Na-phlogopite, Ol: olivine, Opx: orthopyroxene, Tlc: talc.

673 Fig. 3. Water content of the rock as a function of the temperature for model 1 (green curve  
674 for a silicate-FeS core; blue curve for the silicate fraction only) and model 2 (red curve) at  
675 3 GPa.

676

677 Fig. 4. Raman spectra of Na-phlogopite from the experiments (Run 2, blue curve)  
678 compared to a reference spectrum (K-phlogopite R040144 from the RRUFF database  
679 <http://rruff.info>, red curve). The grey vertical lines at frequencies of the main vibration  
680 modes of phlogopite show the match between the reference spectrum and the one obtained  
681 on the present experiment. The two intense peaks in the 1200-1800  $\text{cm}^{-1}$  region correspond

682 to the vibrational modes of amorphous carbon, a result of the transformation of the paraffin  
683 wax and pyridine analogs of organic matter. Aliphatic signature in the 2700-3200  $\text{cm}^{-1}$   
684 region is due to the resin used to consolidate the sample.

685 Fig. 5. Density profiles of the silicate fraction for model 1 (blue curve) and model 2 (red  
686 curve), and density profile of the silicate-sulfide mixture for model 1 (green curve). (a.)  
687 Isotherms 300 K (dashed lines) and 1300 K (solid lines). (b.) Isobar 3 GPa. Dehydration of  
688 hydrous minerals to denser dry minerals results in a progressive increase in density with  
689 increasing temperature.

690 Fig. 6. Models of interior structures for Titan and Ganymede. CM: carbonaceous matter  
691 required for matching the density and MoI with a density of  $1400 \text{ kg/m}^3$  in Titan and  $1800$   
692  $\text{kg/m}^3$  in Ganymede.

693 Fig. 7. Mass vs. MoI relationship for Titan and Ganymede for a range of core radii and  
694 compositions. Empty symbols and dashed curves: CI chondrite composition with ice-  
695 silicate-iron sulfide mixture and fixed radius. Large filled circles: data for Ganymede  
696 (blue) and Titan (orange). Effect of non-hydrostaticity (NH) on Titan MoI determination is  
697 shown. Small circles: effect of different mass fractions (given in wt%) of carbonaceous  
698 matter assuming constant average densities in each layer. Small arrows indicate the effect  
699 of neglecting compressibility in these models.

700

701

702 **Tables**

703 Table 1. Composition of the bulk and silicate fraction of the core in models 1 and 2 assuming  
704 a CI chondrite precursor (Wasson & Kallemeyn 1988).

705

706	Oxide/sulfide (wt.%)	Undifferentiated	Silicate fraction	Silicate fraction
707			Model 1	Model 2
708	MgO	22.85	29.67	36.03
709	SiO <sub>2</sub>	31.91	41.43	50.31
710	CaO	1.83	2.37	2.88
711	Na <sub>2</sub> O	0.92	1.19	1.45
712	Al <sub>2</sub> O <sub>3</sub>	2.31	3.00	3.64
713	FeO	17.21	22.35	5.69
714	FeS	22.98	-	-

715

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717

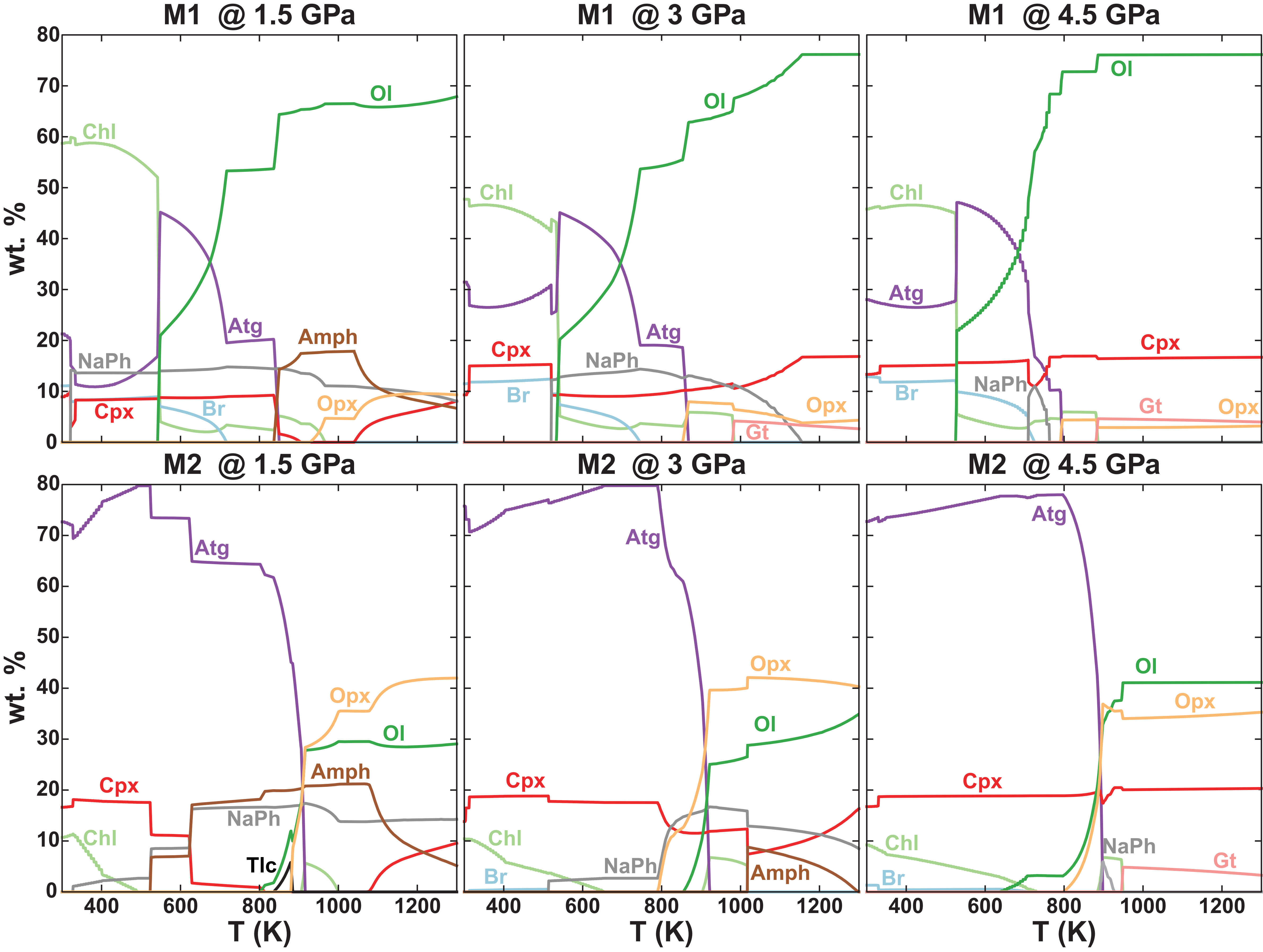
718 Table 2. Starting conditions for MoI calculations displayed in Fig. 7. Radii of the layers and at interfaces are given in km. X is the mass fraction of  
 719 carbonaceous matter added to the core to match the total mass of the moon.

720

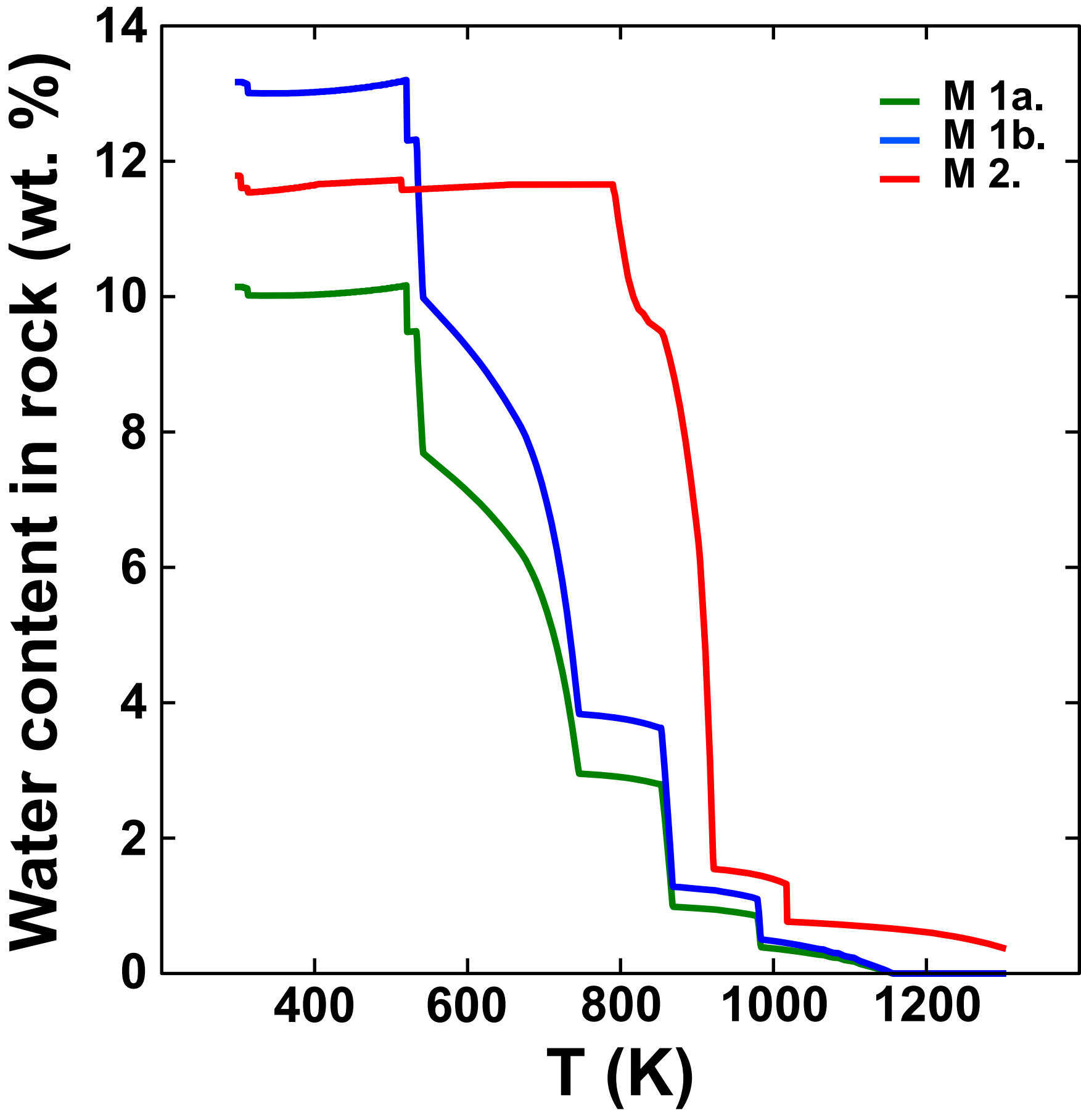
Titan				with carbonaceous matter density of 1400 kg/m <sup>3</sup>			
R surface	R crust/ocean	R ocean/HP ice	Rcore	Rcore	X		
2575	2499	2105	1800	1800	0.06		
2575	2499	2105	1900	1900	0.13		
2575	2499	2105	2000	2000	0.20		
2575	2499	2105	2100	2100	0.26		
2575	2499		2200	2200	0.32		
Ganymede				with carbonaceous matter density of 1800 kg/m <sup>3</sup>			
R surface	R crust/ocean	R ocean/HP ice	R ice/silicate	R Fe-FeS core	R silicate	R Fe-FeS core	X
2631	2539	2193	1800	1080	1800	1055	0.02
2631	2539	2193	1900	1140	1900	1035	0.09
2631	2539	2193	2000	1200	2000	1012	0.16
2631	2539	2193	2100	1260	2100	984	0.23
2631	2539		2200	1320	2200	952	0.29

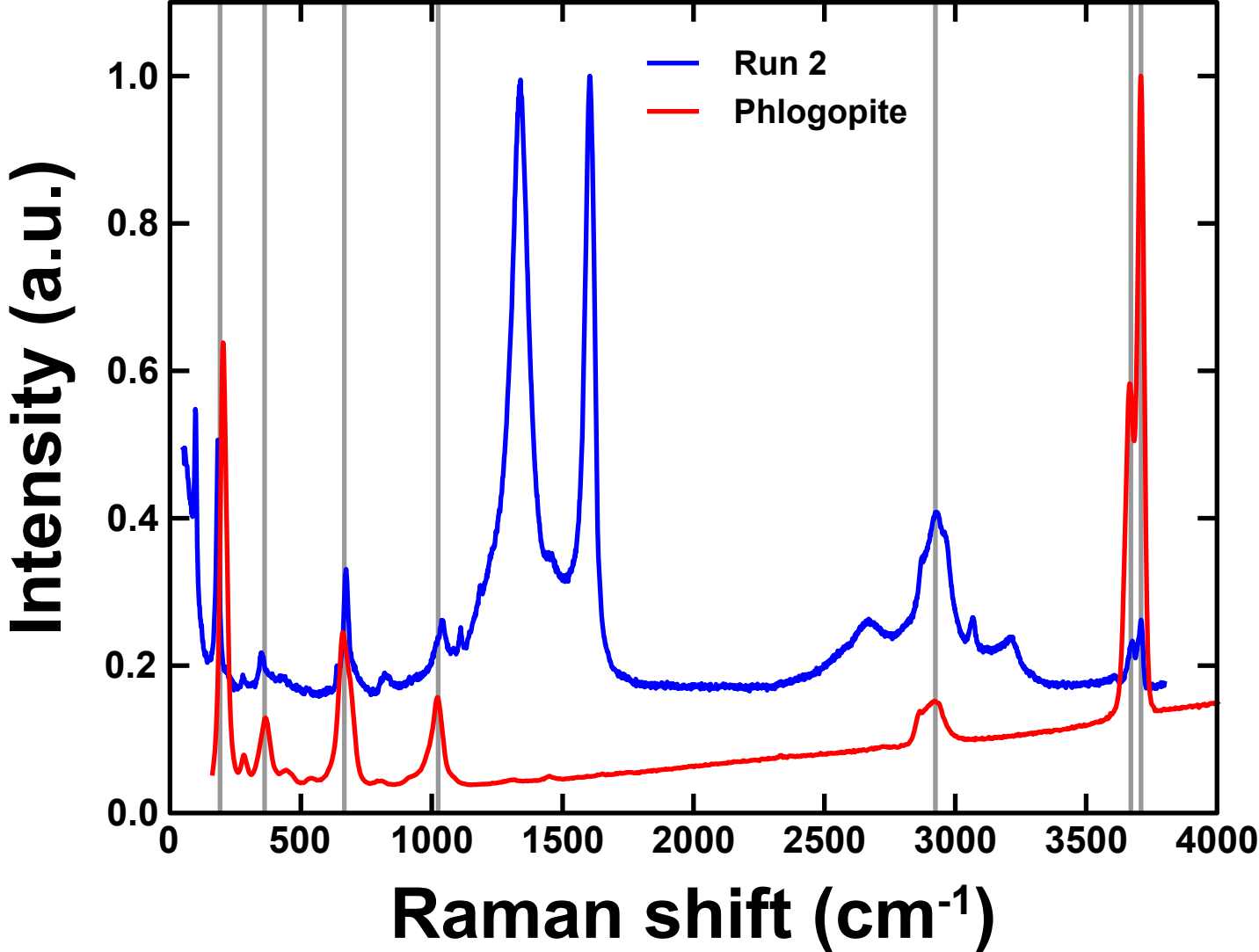
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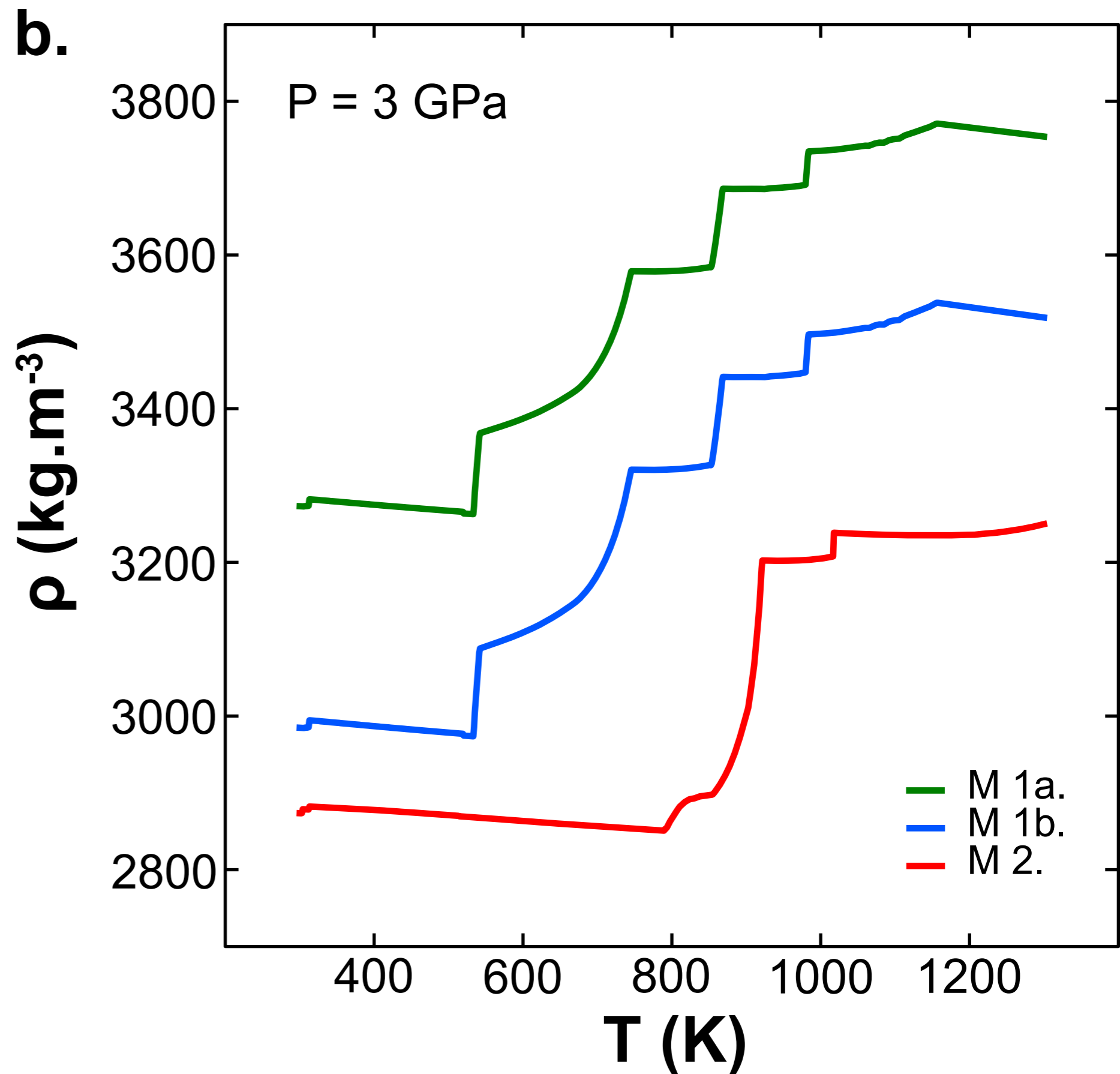
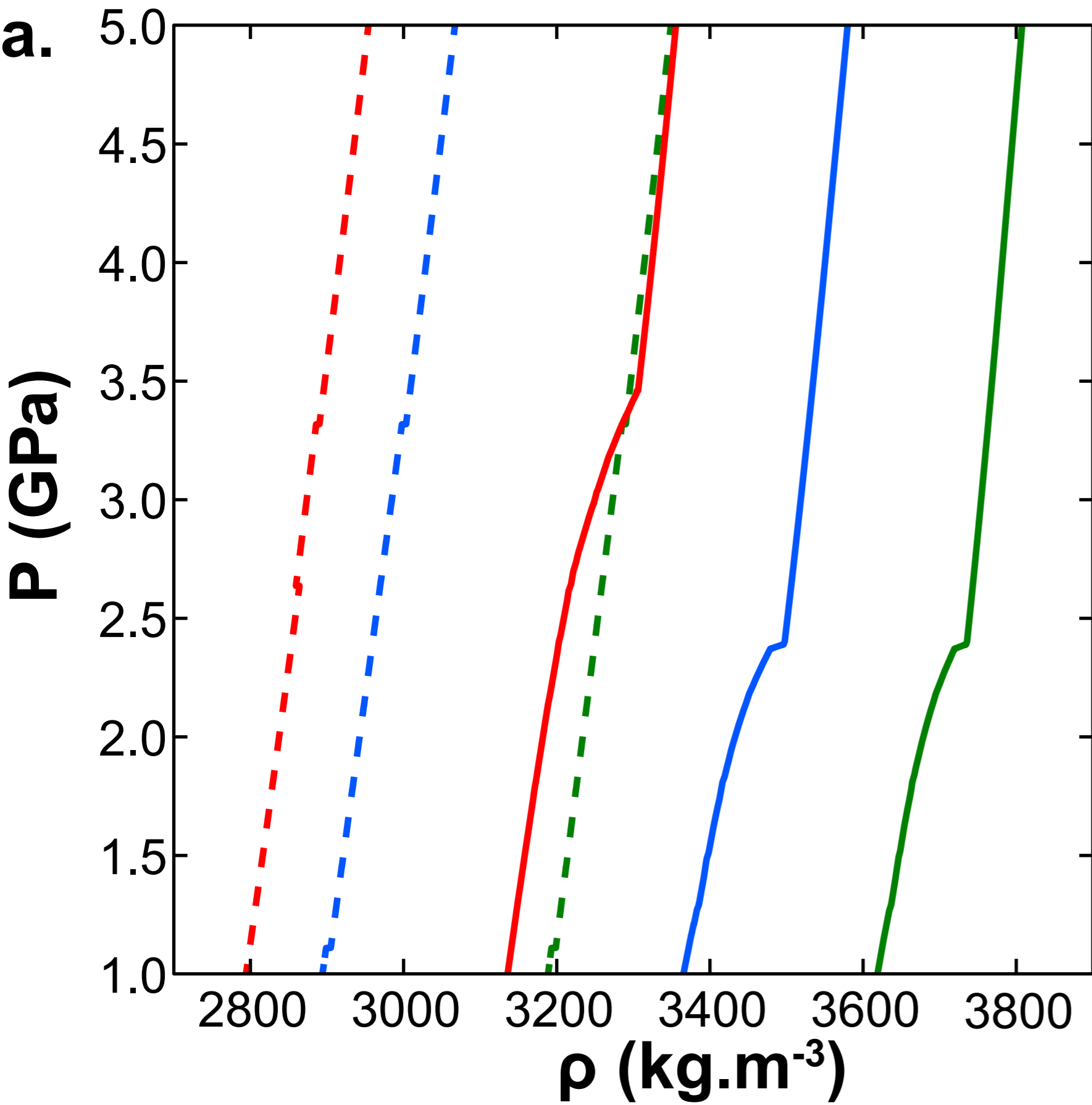


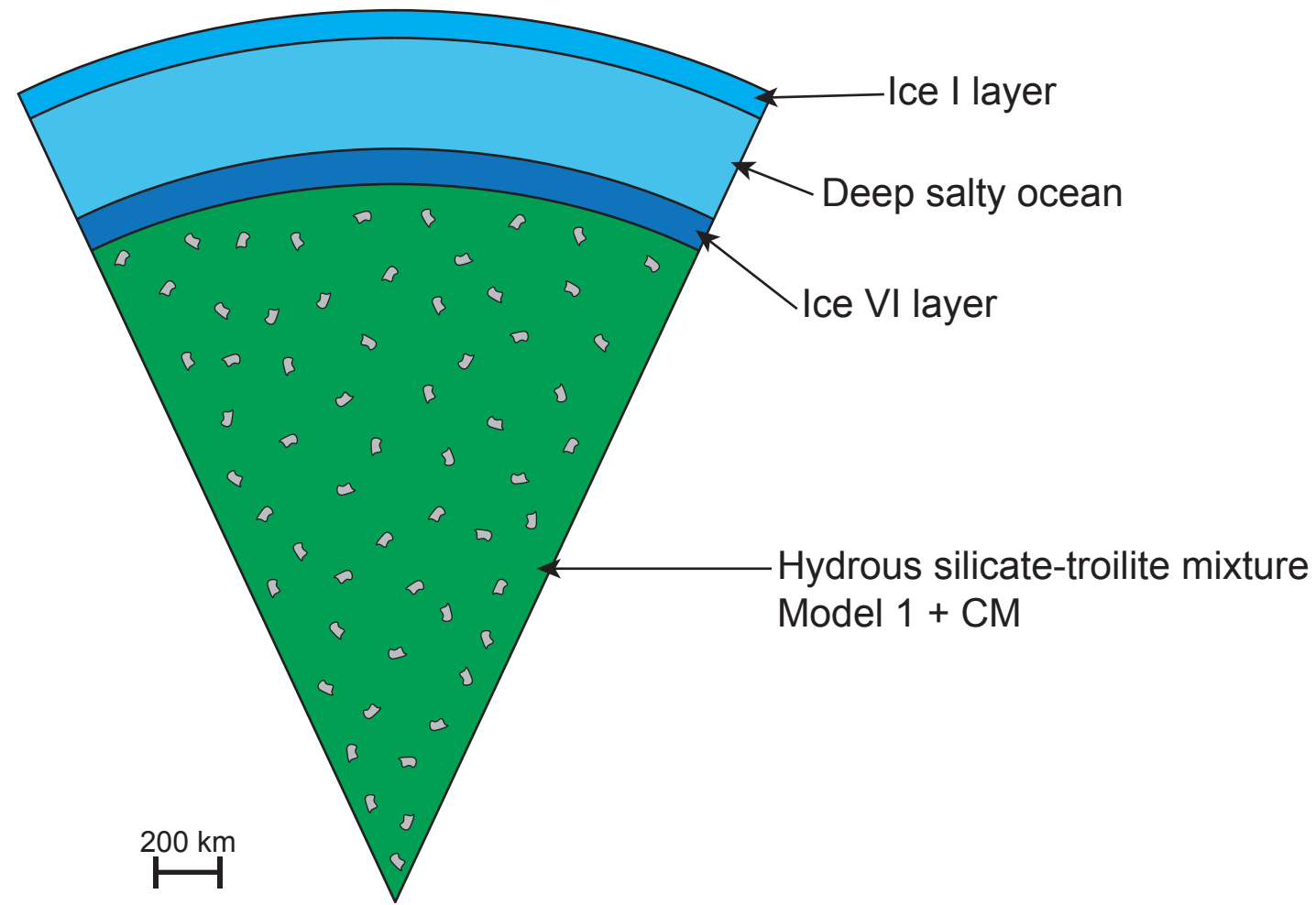










**a.****Model 1****b.****Model 2**