

1 Negligible liquid-vapor separation in the proto-lunar disk: constraints on  
2 evolution

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8 Abstract

9 Despite its importance to lunar origin, the evolution of the proto-lunar  
10 disk eludes precise description. A number of developments nevertheless  
11 make it possible to constrain this evolution. Here, we use a model of the  
12 proto-lunar disk with a silicate vapor atmosphere and a magma layer to  
13 explore the consequences of mixing across the radial extent of the disk.  
14 Four features of such a stratified disk act to radially segregate silicon  
15 isotopes by mass: (i) the magma-vapor interface exhibits a temperature  
16 gradient across the disk radial extent, (ii) silicon experiences distinct  
17 bonding environments in the magma and vapor, producing large equilibrium  
18 isotope effects between the phases, (iii) equilibration between a magma  
19 layer and a atmosphere therefore concentrates heavy isotopes into the  
20 magma, generating an isotopically light atmosphere, with the magnitude of  
21 the isotopic difference dependent on temperature. Temperature gradients  
22 are therefore translated via equilibrium partitioning into compositional  
23 gradients in the magma layer and atmosphere, (iv) radial mixing in the  
24 atmosphere results in net isotopic transport, which redistributes heavy  
25 (light) isotopes to outer (inner) disk regions. That inner disk material  
26 accretes onto Earth whereas the outer disk participates in lunar accretion  
27 permits such a redistribution process to be expressed in the composition  
28 of Earth's mantle and Moon. Existing isotopic data severely constrain the  
29 extent to which the proto-lunar disk existed in such a stratified state.  
30 The silicon isotopic constraint poses a challenge to evolutionary disk  
31 scenarios that efficiently separate proto-lunar liquids from vapors.  
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37 1. INTRODUCTION

38 The Moon is generally thought to have formed from a melt-vapor disk generated  
39 via the off-center impact of a planet-sized body with the proto-Earth towards  
40 the end of Earth accretion (Cameron & Ward 1976; Hartmann & Davis 1975). Such  
41 an origin via giant impact can explain the angular momentum present in the  
42 Earth-Moon (EM) system (Cameron & Ward 1976), the lunar mass and Fe deficit  
43 (Canup & Asphaug 2001), and the small but non-zero inclination of the lunar  
44 orbit (Ćuk et al. 2016; Pahlevan & Morbidelli 2015; Touma & Wisdom 1998; Ward  
45 & Canup 2000). In its simplest formulation, however, the giant impact model  
46 cannot readily explain the precise isotopic similarities between the silicate  
47 Earth and Moon (Kruijer & Kleine 2017; Pahlevan 2014) as most of the material  
48 injected into orbit is sourced from the impacting planet rather than the proto-  
49 Earth, a robust feature of all impact simulations that leave the system with  
50 its current level of angular momentum (Canup 2004; Canup & Asphaug 2001; Canup,  
51 Barr, & Crawford 2013). Here, we refer to the low-velocity, oblique, Mars-mass  
52 impact onto the proto-Earth as the “standard” impact.

53  
54 Motivated by the problem of isotopic homogeneity in the EM system, three new  
55 single-impact scenarios have been proposed. First, it has been suggested that  
56 the Moon-forming impact was a hit-and-run event (Reufer et al. 2012) such that  
57 the impact could be characterized by more angular momentum than currently  
58 present in the EM system, with the excess carried off by the impacting body.  
59 Such an impact can source a somewhat higher percentage of the proto-lunar disk  
60 from Earth’s mantle (~60%) than the standard scenario (~40%) and whereas this  
61 reduces the severity of the isotopic problem, it is unlikely to eliminate it  
62 entirely. Second, it has been suggested that the proto-Earth was rotating near  
63 the threshold for fission before impact – with a  $< 3$  hour rotation period – and  
64 that a high-velocity impactor struck it nearly head-on generating a satellite-  
65 forming disk directly from proto-Earth’s mantle (Ćuk & Stewart 2012). Finally,  
66 it has been proposed that the Moon-forming event may have been a nearly symmetric  
67 impact between two ~half Earth-mass bodies (Canup 2012). In this scenario, both  
68 the emerging planet and disk injected into orbit are sourced – by symmetry –  
69 equally from the impacting bodies. These last two scenarios have the potential  
70 to generate planet-satellite systems with close isotopic similarities, at least  
71 for lithophile elements (Pahlevan 2014). However, both scenarios involve an  
72 overabundance of angular momentum – by a factor of 2 or greater – relative to  
73 the modern EM system. Hence, to be reconciled with the observed dynamical state  
74 of the system, most of the angular momentum in these model systems must be  
75 transferred out. The only mechanisms that might remove this amount of angular

76 momentum are the evection resonance (Ćuk & Stewart 2012; Touma & Wisdom 1998),  
77 a related limit cycle (Rufu & Canup 2020; Tian, Wisdom, & Elkins-Tanton 2017;  
78 Wisdom & Tian 2015), or the Laplace plane instability (Atobe & Ida 2007; Ćuk,  
79 et al. 2016; Tian & Wisdom 2020). The efficacy of all these mechanisms depends  
80 on specific values of the tidal dissipation parameters of the post-impact Earth  
81 and Moon that are likely variable during this epoch and not well-constrained,  
82 making self-consistent thermal-orbital models difficult to construct. Magnetic  
83 winds have also been proposed as a way to carry off angular momentum but the  
84 efficacy of this mechanism for the post-impact Earth-Moon system is also unclear  
85 (Gammie, Liao, & Ricker 2016).

86  
87 A different kind of solution to the problem of EM isotopic homogeneity – called  
88 equilibration – envisions a proto-lunar disk initially isotopically distinct  
89 from Earth, and invokes fluid dynamical mixing of the hot, convective, planet-  
90 disk system into a single isotopic reservoir during the  $\sim 10^3$  years after the  
91 giant impact but before the cessation of lunar accretion (Lock et al. 2018;  
92 Pahlevan & Stevenson 2007). Since the Moon accretes from the outermost disk  
93 material (Salmon & Canup 2012), such a scenario requires that the proto-lunar  
94 disk undergo turbulent mixing across its radial extent. Efficient radial mixing  
95 is possible when the radial mixing timescale is short compared to the disk  
96 evolution timescale. Modeling the effect of turbulence on compositional  
97 transport via a diffusivity  $D$  and the effect of turbulence on angular momentum  
98 transport via a viscosity  $\nu$ , efficient radial mixing occurs when the turbulent  
99 Schmidt number  $Sc(\equiv \nu/D)$  is much less than unity.  $Sc$  depends on the  
100 characteristics of turbulence. Although  $Sc$  has been measured in simulations of  
101 magnetically driven disk turbulence (Carballido, Stone, & Pringle 2005), it has  
102 not yet been measured in simulations of the convectively driven turbulence that  
103 is likely present in the protolunar disk. The effective viscosity associated  
104 with convection is believed to be small (Lesur & Ogilvie 2010; Stone & Balbus  
105 1996).

106  
107 Given the suite of new ideas aimed at understanding EM isotopic homogeneity and  
108 lunar origin, we need new tools to constrain and test the various scenarios.  
109 Here, we describe development of one such tool. We show that turbulent radial  
110 mixing in a stratified proto-lunar disk – with a silicate vapor atmosphere and  
111 a separately convective midplane magma layer co-existing in equilibrium – makes  
112 specific predictions for mass-dependent isotopic offsets in the composition of  
113 lunar and terrestrial rocks. We then use the measured isotopic abundances to  
114 set constraints on how the proto-lunar disk could – and could not – have evolved.

115 Finally, we discuss implications for equilibration as the origin of the EM  
116 isotopic homogeneity.

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## 118 2. DISK MODEL

119 Currently considered scenarios for a single Moon-forming giant impact (Canup  
120 2004, 2012; Canup & Asphaug 2001; Ćuk & Stewart 2012; Reufer, et al. 2012)  
121 shock-heat and disaggregate the lunar-forming material into a hot, melt-vapor  
122 circumterrestrial disk (Nakajima & Stevenson 2014). At present, no model exists  
123 that begins with such an initial state and follows the evolution, ending with  
124 statements about the stable isotopic composition of major elements constituting  
125 the lunar-forming material. Here, we describe a numerical disk model with the  
126 goal of forging a link between disk processes and stable isotopic data on lunar  
127 and terrestrial rocks. The physical picture envisioned for protolunar disk  
128 evolution is similar to that initially envisioned for the equilibration  
129 hypothesis (Pahlevan & Stevenson 2007). The rationale for this choice is that  
130 developing the consequences of this well-defined scenario yields insights into  
131 how lunar disk evolution more generally could – and could not – have occurred.

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### 133 2.1. Vertical Structure

134 The melt-vapor disk generated by the giant impact settles into a nearly  
135 hydrostatic state – with a vapor cloud expanding to assume a density  
136 distribution according to the local scale height – on dynamical timescales of  
137 hours to days. Radiative cooling subsequently drives the evolution of the proto-  
138 lunar disk. In the standard picture, vigorous turbulent convection is required  
139 to match high radiative losses from a ~2,000 K photosphere (Thompson & Stevenson  
140 1988), above which the thermal structure is determined by radiative transfer.  
141 Calculation of the disk vertical structure is simplified by the fact that a  
142 negligible fraction of atmospheric mass exists above the photosphere at  
143 ~millibar pressures with the convective atmosphere extending to pressures of  
144 hundreds of bars (Thompson & Stevenson 1988). The disk atmosphere is almost  
145 entirely convective and is expected to be characterized by an adiabatic (or  
146 pseudoadiabatic) vertical structure (Holton 1992).

147

148 A major unresolved question – which has consequences for lunar composition – is  
149 whether the liquid in the proto-lunar disk exists as droplets kept in suspension  
150 via vigorous convection (Thompson & Stevenson 1988) or whether it settles into  
151 a midplane layer that dynamically decouples from the overlying atmosphere  
152 (Machida & Abe 2004; Ward 2012). We refer to these two cases as the unstratified  
153 and stratified disks, respectively. An end-member stratified disk scenario

154 (herein called “fully stratified”) is one in which, not only is the midplane  
 155 more liquid-rich than would be expected based on a single vertical disk  
 156 isentrope, but the exchange of matter between the liquid layer and the  
 157 atmosphere is mediated by exchange across a phase boundary, with no advective  
 158 fluid motions carrying atoms directly between the two layers (e.g., no  
 159 convective overshoot). In such a fully stratified scenario, elements that  
 160 readily vaporize (e.g., O) will enter the atmosphere and be available to undergo  
 161 radial mixing and isotopic equilibration (Pahlevan & Stevenson 2007) whereas  
 162 more refractory elements (e.g., Ti) will be concentrated in the midplane liquid  
 163 layer and undergo less Earth-disk equilibration. EM isotopic heterogeneity for  
 164 refractory elements has been sought, but not found (Zhang et al. 2012).

165  
 166 Here, we assume that the disk is fully stratified: that the vertical structure  
 167 consists of a convective liquid-rich midplane layer, a liquid-vapor interface,  
 168 and an overlying convective atmosphere. The separately convective liquid-rich  
 169 and vapor-rich columns are assumed to be in thermodynamic equilibrium at  
 170 precisely defined interface conditions. Vertical mixing *within* each layer is  
 171 assumed to be efficient because convective velocities are high and variations  
 172 in atmospheric composition due to phase separation (“rainout”) are expected to  
 173 be small relative to the compositional contrast between the co-existing liquid  
 174 layer and vapor atmosphere (Pahlevan, Stevenson, & Eiler 2011). Implications of  
 175 relaxing the assumption of efficient vertical mixing are discussed in §4.2.

176  
 177 The pressure at the liquid-vapor interface is determined by hydrostatic balance  
 178 with the self-gravity of the disk and the vertical component of Earth gravity:

$$179 \quad -\frac{1}{\rho} \frac{\partial P}{\partial z} = 2\pi G \sigma(z) + \frac{GM_E}{r^3} z \quad (1)$$

180 where  $\rho$  is the density,  $P$  is the pressure,  $z$  is the height above the midplane,  
 181  $G$  is the gravitational constant,  $\sigma(z)$  is the local disk surface density enclosed  
 182 at height  $z$ ,  $M_E$  is the mass of Earth, and  $r$  is the radial distance in cylindrical  
 183 coordinates. This equation can be integrated to yield:

$$184 \quad P_{int} = \frac{\pi G \sigma_T^2}{2} \left[ 1 - (1 - f_v)^2 \right] + \frac{1}{2} \Omega^2 \sigma_T f_v h_{mass} \quad (2)$$

185 where  $P_{int}$  is the interface pressure,  $\sigma_T$  is the total surface density of the  
 186 column (liquid layer plus atmosphere),  $f_v$  is the mass fraction of the vapor  
 187 atmosphere in the column ( $\equiv \sigma_v / \sigma_T$ ),  $\Omega$  is the Keplerian angular velocity  
 188 [ $\equiv (GM_E / r^3)^{1/2}$ ], and  $h_{mass}$  the mass-weighted scale height of the column which can  
 189 be approximated as the mass-weighted height of the vapor atmosphere:

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$$h_{mass} = 2 \int_{z_{int}=0}^{\infty} z \rho_v(z) dz \Big/ \int_{-\infty}^{\infty} \rho_v(z) dz \quad (3)$$

191 We assume that the second term in Equation (2) corresponding to the vertical  
 192 component of Earth's gravity dominates, such that  $h_{mass} \sim c_s/\Omega$  where  $c_s$  is the  
 193 isothermal sound speed [ $\equiv (kT/\mu)^{1/2}$ ] and  $\mu$  the vapor mean molecular weight. By  
 194 substituting nominal parameter values for the proto-lunar disk ( $\sigma_T = 5 \times 10^6$  g/cm<sup>2</sup>,  
 195  $\Omega = 2 \times 10^{-4}$  s<sup>-1</sup>,  $c_s = 10^5$  cm/s) into both terms in Equation (2), the dominance of  
 196 central gravity in determining disk vertical structure can be confirmed.<sup>4</sup> This  
 197 calculation justifies neglecting disk self-gravity, as also assumed in (Ward  
 198 2012). Previous works have shown that the convective proto-lunar disk is nearly  
 199 isothermal vertically due to large latent heat of condensation of silicates  
 200 (Genda & Abe 2003; Thompson & Stevenson 1988). Accordingly, we adopt an  
 201 isothermal vertical structure for the atmosphere with the well-known Gaussian  
 202 density structure  $\rho(z) = \rho(0) \exp(-z^2/H^2)$  and scale height given by  $H = \sqrt{2} C_s/\Omega$ .  
 203 Substituting this structure into Equation (3) yields a mass-weighted height  
 204  $h_{mass} = \sqrt{2/\pi} C_s/\Omega$ , and substituting this expression into the second term in  
 205 Equation (2) yields an expression for the interface pressure:

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$$P_{int} = \frac{1}{\sqrt{2\pi}} f_v \sigma_T C_s \Omega \quad (4)$$

207 We use this expression to relate pressure at the liquid-vapor interface to other  
 208 variables describing the thermodynamic state of the disk locally.

209

## 210 2.2. Radial Structure

211 The disks injected into orbit via giant impacts are generally compact, with  
 212 most of the orbiting mass confined to several planetary radii. For simplicity,  
 213 we assume that the surface density profile characterizing the proto-lunar disk  
 214 scales as  $\sim 1/r$ , as seen in simulations of the standard impact (Canup, et al.  
 215 2013). High angular momentum impacts (Canup 2012; Ćuk & Stewart 2012) tend to  
 216 create even more compact disks, which may cause identifiable differences in the  
 217 satellite that results (Salmon & Canup 2014). For the purpose of this work, we  
 218 neglect differences in radial mass distribution. In order to make a connection  
 219 between disk processes and isotopic data, we assume that the proto-lunar disk  
 220 extends from 1-5 Earth radii ( $R_E$ ) and that the outer disk material (from 3-5  $R_E$ )  
 221 accretes onto the Moon with its isotopic composition expressed in the lunar

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<sup>4</sup> Equivalently, one can check that Toomre's  $Q$  parameter ( $c_{gas}\Omega/\pi G\sigma$ ) is  $\gg 1$ , implying that self-gravity is minor.

222 samples, whereas inner disk material (from 1-3  $R_E$ ) reaccumulates back onto Earth  
223 with its compositional character diluted and essentially erased via mixing into  
224 the massive terrestrial magma ocean. In reality, the processes of disk chemical  
225 evolution studied here and of lunar accretion studied elsewhere (Salmon & Canup  
226 2012) overlap but any coupling between these processes is beyond the scope of  
227 this work and must be subject to future modeling. Here, we restrict the study  
228 to disk processes that can cause the outer disk to acquire compositional  
229 characteristics distinct from the inner disk, a distinctness that can be  
230 expressed in the composition of Earth's mantle and Moon. Simulations find that  
231 the initial thermal structure of the impact-generated disk is radially  
232 isentropic (Nakajima & Stevenson 2014), an initial condition we adopt for  
233 calculations of the subsequent thermochemical evolution.

234

### 235 2.3. Thermodynamic Model

236 We apply a two-phase, two-component thermodynamic model expressly developed to  
237 study liquid-vapor fractionation in the aftermath of the giant impact (Pahlevan,  
238 et al. 2011) to calculate equilibrium thermodynamic conditions (e.g.,  $T$ , degree  
239 of vaporization) in the proto-lunar disk. The model has been previously  
240 described; here we summarize its features and describe its current application.  
241 The silicate liquid is modeled as a binary solution of olivine end-members,  
242  $(\text{Fe,Mg})_2\text{SiO}_4$ , whereas the silicate vapor is described as a mixture of ideal  
243 gases consisting of gaseous species formed upon vaporization of such a liquid:  
244  $\text{SiO}_2$ ,  $\text{SiO}$ ,  $\text{Si}$ ,  $\text{FeO}$ ,  $\text{Fe}$ ,  $\text{MgO}$ ,  $\text{Mg}$ ,  $\text{O}_2$ , and  $\text{O}$ . This choice of compositional model  
245 is motivated by the fact that the bulk Moon – and therefore the proto-lunar  
246 disk – is primarily composed of ferromagnesian silicates (Taylor & Wieczorek  
247 2014). At each time step in the evolution, we divide the proto-lunar disk into  
248 annuli and calculate equilibrium between the liquid layer and vapor atmosphere.  
249 Input parameters include the column entropy ranging from highly vaporized to  
250 condensed states ( $S=0.530\text{--}1.030$  kJ/mol.K where mol refers to moles of olivine  
251 units or silicon atoms) and column composition ( $x_c=\text{Fe}/\text{Fe}+\text{Mg}$ ), initially set  
252 equal to Earth mantle composition ( $\approx 0.1$ ) (Jones & Palme 2000). Using equations  
253 derived from the two-phase equilibrium assumption (Pahlevan, et al. 2011), and  
254 the pressure of equilibration (Equation 4), we solve for the interface  
255 temperature ( $T$ ), the column vapor fraction ( $f_v$ ), the specific entropy ( $S_L$ ,  $S_V$ )  
256 and composition ( $X_L$ ,  $X_V$ ) of the liquid and atmospheric columns, as well as the  
257 partial pressure at the interface of each of nine vapor species ( $P_i$ ) at each  
258 time step and annulus.

259

### 260 2.4. Radial Transport

261 Because of the temperature-dependence of equilibrium partitioning (e.g.,  
 262 Equation 9), radial gradients in interface temperature in the disk are expressed  
 263 as radial gradients in composition of the liquid layer and complementary  
 264 atmosphere. Because the disk atmosphere features turbulent convection (Thompson  
 265 & Stevenson 1988), development of compositional gradients will lead to transport  
 266 of species down gradient. Hence, one consequence of an equilibrium fully-  
 267 stratified (settled liquid) disk model is the development of radial atmospheric  
 268 compositional gradients that – in concert with a convective atmosphere – leads  
 269 to net radial transport of chemical and isotopic species. We assume that  
 270 turbulent convection in the atmosphere can be characterized by an eddy  
 271 diffusivity (Tennekes & Lumley 1972). In the presence of compositional  
 272 gradients, atmospheric motions will then transport species in accordance with  
 273 Fick’s law:

$$274 \quad J_i = -\rho_v D_v \nabla c_{i,v} \quad (5)$$

275 where  $J_i$  [ $\text{kg m}^{-2} \text{s}^{-1}$ ] is the mass flux of an atmospheric constituent  $i$ ,  $\rho_v$  is  
 276 atmospheric mass density,  $D_v$  is turbulent diffusivity, and  $c_{i,v}$  is mass fraction  
 277 of constituent  $i$ . In principle, a similar equation could be written for  
 278 turbulent mixing in the magma layer. However, because of both greater length  
 279 scales and velocity scales associated with atmospheric convection, radial  
 280 transport is dominated by atmospheric motions (Pahlevan & Stevenson 2007).  
 281 Accordingly, we consider a liquid layer that is stationary but rapidly  
 282 equilibrating with the turbulent atmosphere. We take the divergence of Equation  
 283 (5), and combine it with a continuity equation:

$$284 \quad \frac{\partial(\rho_v c_{i,v})}{\partial t} = -\nabla \cdot (\rho_v D_v \nabla c_{i,v}) \quad (6)$$

285 to describe atmospheric transport. Using the continuity equation here without  
 286 sources or sinks is valid for species that are neither created nor destroyed.  
 287 Atomic abundances can be described in this way but molecular abundances cannot  
 288 due to the occurrence of chemical reactions. Such an equation is only used to  
 289 describe compositional changes in the atmosphere and compositional changes to  
 290 the liquid layer arising from liquid-vapor equilibration are treated separately  
 291 in the model. The reason for this separate treatment is that – unlike signatures  
 292 that are identical between a silicate liquid and co-existing vapor (e.g.,  $\Delta^{17}\text{O}$   
 293 studied in (Pahlevan & Stevenson 2007)) – here we are considering chemical and  
 294 isotopic signatures that are distinct between co-existing phases (e.g.,  
 295  $^{30}\text{Si}/^{28}\text{Si}$ , see §2.6). Expecting rapid vertical mixing but rate-limiting radial  
 296 mixing in the disk, we integrate Equation (6) vertically through the atmosphere:  
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$$\sigma_v \frac{\partial c_{i,v}}{\partial t} = - \nabla \cdot (\sigma_v D_v \nabla c_{i,v}) \quad (7)$$

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where  $\sigma_v (= f_v \sigma_T)$  is the atmospheric surface density [ $\text{kg m}^{-2}$ ]. We assume that this parameter has no time-dependence arising from disk turbulence, because turbulent convection leads to little net mass transfer. Indeed, numerical studies of disks have found that convection does not efficiently transport angular momentum (Lesur & Ogilvie 2010; Stone & Balbus 1996), a requirement for net mass transport. This issue is further discussed in §4.1. Atmospheric surface density does vary in the model due to thermal evolution (i.e., condensation, §2.5). At each time step, we use Equation (7) in cylindrical coordinates to describe changes in atmospheric composition with zero net flux boundary conditions,  $\partial c / \partial r(R_E, t) = \partial c / \partial r(5R_E, t) = 0$ , reflecting the simplification of neglecting moonlet formation and satellite-disk interactions in the outer disk (Salmon & Canup 2012) and neglecting turbulent mixing with the silicate vapor atmosphere of Earth in the inner disk (Lock, et al. 2018; Pahlevan & Stevenson 2007). Different boundary conditions are possible, but here we make this choice in order to study processes within the proto-lunar disk itself, relegating interactions with its environment to future works.

## 2.5. Thermal Evolution

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The viscous and thermal evolution of the proto-lunar disk is a closely coupled problem. The material in the proto-lunar disk is susceptible to gravitational instability (Cameron & Ward 1976) with a viscous evolution timescale for particles of order  $\sim 1$  year (Ward & Cameron 1978). Such a rapid evolutionary timescale liberates enough gravitational energy for the material to self-vaporize, violating the original particle assumption (Thompson & Stevenson 1988). Hence, the proto-lunar disk is thought to be thermally regulated, whereby heating due to viscous gravitational energy release would be controlled by radiative losses. Accordingly, subsequent works explore coupled solutions to the viscous and thermal evolution (Charnoz & Michaut 2015; Gammie, et al. 2016; Ward 2012). Here, we adopt a simplified thermal history in which radiative losses are powered by secular cooling alone; we do not consider viscous heating. The motivation for, and limitations of, this assumption are discussed in §4.1.

Treating radiative cooling as a quasistatic process, we relate the outgoing heat and entropy fluxes using the thermodynamic identity,  $dQ = TdS$ , where  $T$  is the temperature of the radiated photons. Further approximating the radiation as

334 a blackbody, we write an expression for the entropy per unit area being radiated  
335 from both sides of the proto-lunar disk per unit time:

$$336 \quad dS = 2\sigma_{SB} T_{ph}^3 dt \quad (8)$$

337 where  $\sigma_{SB}$  is the Stefan-Boltzmann constant,  $T_{ph}$  is the photospheric temperature.  
338 We can use this expression to calculate radiative entropy losses from disk  
339 annuli. Because of vertical convection, the effects of radiative cooling rapidly  
340 propagate down through the column; equivalently, the entire thermal reservoir  
341 of the disk is tapped by radiative losses. At each time step, new values of  
342 entropy are derived for each annulus after radiative losses, whereas radial  
343 transport (§2.4) and liquid/vapor re-equilibration (§2.3) are calculated using  
344 an operator split scheme, i.e., calculating the effects of each process in  
345 sequence.

346  
347 We follow the thermal evolution of the disk from an initial to a final state.  
348 Motivated by the impact simulations, we adopt initial thermal states – expressed  
349 as vapor fraction – ranging from mostly (~80%) liquid (Canup 2004) to mostly  
350 (50–95%) vapor (Canup 2012; Ćuk & Stewart 2012). For final states, because we  
351 are interested in articulating expected compositions of the outer disk material,  
352 and because atmospheric transport dominates convective transport in the liquid  
353 layer (§2.4), when the vapor in the outer disk (3–5  $R_E$ ) has fully condensed ( $f_v$   
354 < 0.01), the composition of the liquid is considered frozen-in, to be later  
355 expressed in the isotopic composition of the Moon and its rock samples. Hence,  
356 we consider the evolution until condensation of the outer disk. In addition to  
357 neglecting viscous heating, we do not consider turbulent radial transport of  
358 entropy and heat in the atmosphere, or other possible triggers of compositional  
359 freeze-in, e.g., disk fragmentation into moonlets, but discuss possible  
360 variations in disk models in §4.2 and §4.3.

## 361 362 2.6. Silicon Isotopes as Tracers

363 We choose silicon isotopes as tracers for several reasons. First, at high  
364 temperature, equilibrium isotopic differences between co-existing phases  
365 approach zero (Urey 1947) and small but significant isotopic fractionation is  
366 only possible where there are significant differences in bonding environment  
367 between the phases present for the element under consideration. Silicon, for  
368 example, exists as  $\text{SiO}_4^{4-}$  in silicate melts and predominantly as SiO in co-  
369 existing vapors (Visscher & Fegley 2013), permitting large liquid-vapor isotopic  
370 fractionation (~0.5 per mil in  $^{30}\text{Si}/^{28}\text{Si}$ ) in the high-temperature environment  
371 (~3,000–4,000 K) encountered during lunar origin (Pahlevan, et al. 2011).

372 Second, unlike magnesium, iron, and oxygen, which significantly reside in  
 373 multiple gas species in silicate vapors, SiO dominates the vapor speciation of  
 374 silicon (e.g., over SiO<sub>2</sub> and Si) for the full range of conditions in the proto-  
 375 lunar disk (Visscher & Fegley 2013). Because of such a simple monopoly in vapor  
 376 speciation, even a two-component thermodynamic model (§2.3) accurately captures  
 377 the speciation of silicon in the proto-lunar disk, a requirement for accurately  
 378 articulating predictions of isotopic fractionation in evolutionary scenarios.  
 379 Finally, to forge a connection with the observable data, we require an element  
 380 whose isotopic abundances in Earth's mantle and Moon are precisely known.  
 381 Because the bonding environment for silicon in silicate melts and crystals are  
 382 similar (Huang et al. 2014), substantial silicon isotopic variations are not  
 383 generated during ordinary (i.e., crystal-liquid) petrologic processes such as  
 384 mantle melting (Zambardi et al. 2013) or during magma ocean crystallization.  
 385 Hence, the isotopic composition of Earth's mantle and Moon can be derived from  
 386 measurements on samples and are known to high precision, permitting comparison  
 387 of the observational data with predictions of post giant-impact liquid-vapor  
 388 evolution (Armytage et al. 2012; Pahlevan 2014) without any substantial signal  
 389 degradation from subsequent geological processes, to which silicon isotopes are  
 390 essentially impervious.

391  
 392 Silicon isotopes are considered as passive tracers. At each time step, liquid-  
 393 vapor equilibrium at each annulus is characterized by a temperature, T (§2.3).  
 394 Because the isotopic fractionation is only a function of temperature in this  
 395 low-pressure (<1GPa) system, we calculate the partitioning of Si isotopes  
 396 between the silicate liquid and vapor as:

$$397 \quad \Delta_{L-V} = 5 \left( 10^3 / T \right)^2 \text{ per mil} \quad (9)$$

$$398 \quad \delta_C = F_V^{Si} \delta_V + \left( 1 - F_V^{Si} \right) \delta_L \quad (10)$$

399 where  $\Delta_{L-V} = \delta_L - \delta_V$  is the part per thousand difference of the <sup>30</sup>Si/<sup>28</sup>Si between the  
 400 liquid and vapor, as previously calculated using standard procedures (Pahlevan,  
 401 et al. 2011) and  $\delta_L$ ,  $\delta_V$ ,  $\delta_C$  represent the isotopic compositions of the liquid  
 402 layer, the vapor atmosphere, and the entire column. We set the initial disk  
 403 composition  $\delta_C(r,0)$  to zero for reference. In this way, we calculate radial  
 404 isotopic structure of the proto-lunar disk atmosphere and liquid layer, given  
 405 a radial disk compositional  $[\delta_C(r,t)]$  and thermal  $[T(r,t)]$  structure. Two  
 406 features of the isotopic partitioning are notable. First,  $\Delta_{L-V}$  is positive, such  
 407 that the heavier isotopes of silicon concentrate into silicate liquids. Second,  
 408 the temperature dependence of the fractionation guarantees spatial variations  
 409 in the liquid-vapor partitioning in the proto-lunar disk. In particular, the

410 radial gradient in equilibration temperature will be expressed – through  
411 equilibrium partitioning (Equation 9) – as silicon isotopic gradients in the  
412 atmosphere and liquid layer. Once this equilibrium state is calculated, we  
413 propagate the evolution forward by calculating the radial isotopic transport  
414 via Equation (7), where  $c_{i,v}$  now represents  $\delta_v$ . After calculating radial  
415 transport, we permit the atmosphere – now out of equilibrium with the liquid  
416 layer – to re-equilibrate, thereby changing the composition of the lunar-forming  
417 liquid. In this way, turbulent motions in the convective vapor atmosphere can  
418 change the isotopic composition of a quasi-stationary Moon-forming liquid. In  
419 the case of silicon, such isotopic redistribution tends to concentrate the heavy  
420 isotopes in the outer (i.e., Moon-forming) regions of the disk.

421

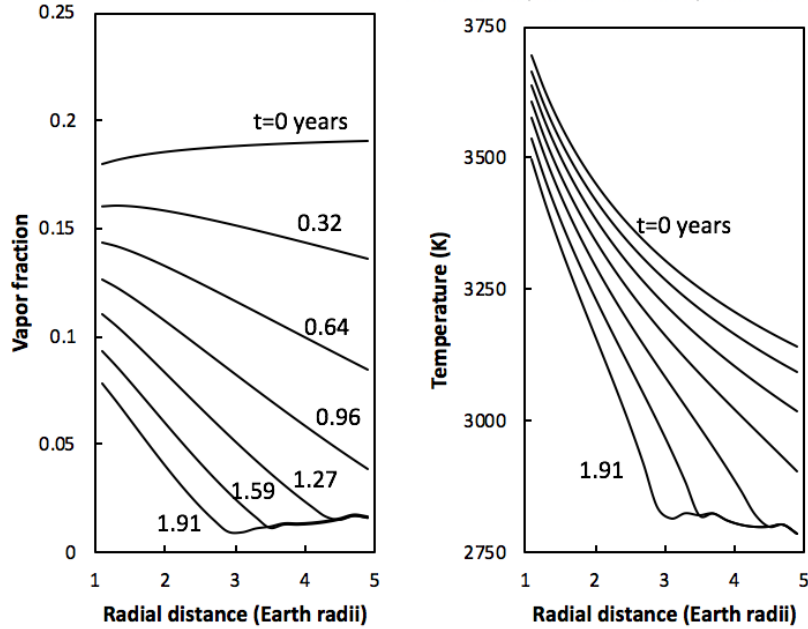
### 422 3. RESULTS

#### 423 3.1. Thermal State and History

424 Because of the weak (logarithmic) dependence of the entropy of ideal gases on  
425 pressure, the initial isentropic state of the proto-lunar disk translates to a  
426 radially ~constant vapor fraction initially (Figure 1a). Because of high latent  
427 heat of condensation and high vapor fractions (~tens of percent), the secular  
428 cooling heat budget of the proto-lunar disk is dominated by latent heat of  
429 condensation, with minor contributions from sensible heat ( $c_p\Delta T$ ). Hence, unlike  
430 the post-impact atmosphere of Earth which is thermally buffered from below,  
431 secular cooling of the proto-lunar disk in the absence of viscous heating is  
432 rapid. Because of an outwardly decreasing surface density profile, the disk  
433 first cools at large distances with a condensation front that propagates inwards  
434 on a timescale of years (Figure 1a).

435

436 The temperature structure of the disk has several notable features. First, even  
437 at the outset of the evolution, an isentropic disk is characterized by a  
438 significant radial temperature gradient, as also observed in (Nakajima &  
439 Stevenson 2014). Second, because of more rapid cooling in the outer disk, the  
440 radial T-gradient of the disk steepens with time, ranging from 125–350 K/ $R_E$   
441 (Figure 1b). The existence of such a radial temperature gradient is expected to  
442 be a universal feature of any proto-lunar disk model and plays a central role  
443 in the evolution of isotopic signatures considered below.

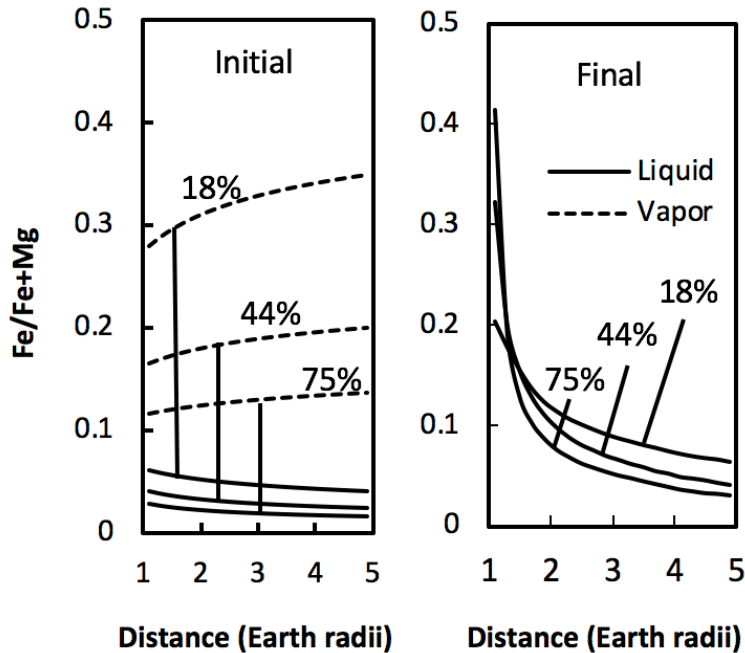


**Figure 1.** Disk thermal state and evolution. (a) Disk vapor fraction (atmospheric mass / total disk column mass) versus radial distance. This cooling history corresponds to an initial condition ( $f_v \sim 0.2$ ) generated via the “standard” impact. The simulations are halted at  $f_v \sim 0.01$  for numerical reasons. (b) Equilibration temperatures at the liquid-vapor interface versus radial distance. Note that, even in an initially isentropic disk, the disk is characterized by a radial temperature gradient due to the radial pressure gradient arising from, e.g., radial surface density gradients in the disk, and cooling of the disk only accentuates a pre-existing T-gradient.

### 444 3.2. Major Element Chemical Fractionation

445 The two-component [(Fe,Mg)<sub>2</sub>SiO<sub>4</sub>] thermodynamic model adopted has one variable  
 446 for chemical composition,  $x_{Fe} (\equiv Fe/Fe+Mg)$ , making possible studies of chemical  
 447 fractionation. Accordingly, we track the  $x_{Fe}$  of the liquid, vapor, and total  
 448 column in the proto-lunar disk during the evolution. As has been experimentally  
 449 determined, fayalite (Fe<sub>2</sub>SiO<sub>4</sub>) has a higher vapor pressure than forsterite  
 450 (Mg<sub>2</sub>SiO<sub>4</sub>) at identical temperature (Nagahara, Kushiro, & Mysen 1994), a behavior  
 451 clearly expressed in the proto-lunar disk: the vapor atmosphere is enriched in  
 452 FeO ( $x_{Fe} = 0.12-0.35$ ), while the liquid layer is FeO-depleted ( $x_{Fe} = 0.02-0.06$ ),  
 453 relative to the composition of the column as a whole ( $x_{Fe} = 0.1$ , Figure 2a).  
 454 Moreover, due to the temperature and pressure gradients in the disk, even an

455 initial chemical homogeneity will – through equilibrium partitioning – result  
 456 in radial chemical gradients in liquid and atmospheric reservoirs. An increasing  
 457 iron oxide content of the vapor atmosphere with radius (Figure 2a) implies an  
 458 inward turbulent transport of iron in the atmosphere arising from convective  
 459 mixing. Through liquid-vapor re-equilibration, atmospheric transport of species  
 460 also leads to the development of chemical gradients in the lunar-forming liquid.  
 461 In this way, turbulent transport can be expected to redistribute chemical  
 462 species in the proto-lunar disk.



**Figure 2.** Fe/Mg redistribution in the proto-lunar disk. (a) Initial liquid-vapor partitioning in a disk with bulk  $x_{\text{Fe}}(\text{Fe}/\text{Fe}+\text{Mg})=0.1$ . The percentage labels on curves correspond to initial disk vapor fraction. (b) Final compositional structure of the disk when the vapor has condensed and the liquid composition has frozen in. The vigor of turbulence in these simulations is  $\alpha=3\times 10^{-3}$ .

463 Two features of turbulent chemical transport are notable: (i) the outer disk  
 464 material from which the Moon forms is modestly depleted in iron-rich silicates  
 465 relative to the inner disk, which falls back onto Earth (Figure 2b). The sign  
 466 of this fractionation can be understood as follows: as the outermost disk  
 467 condenses, it becomes increasingly iron-rich (Figure 2a), such that turbulent

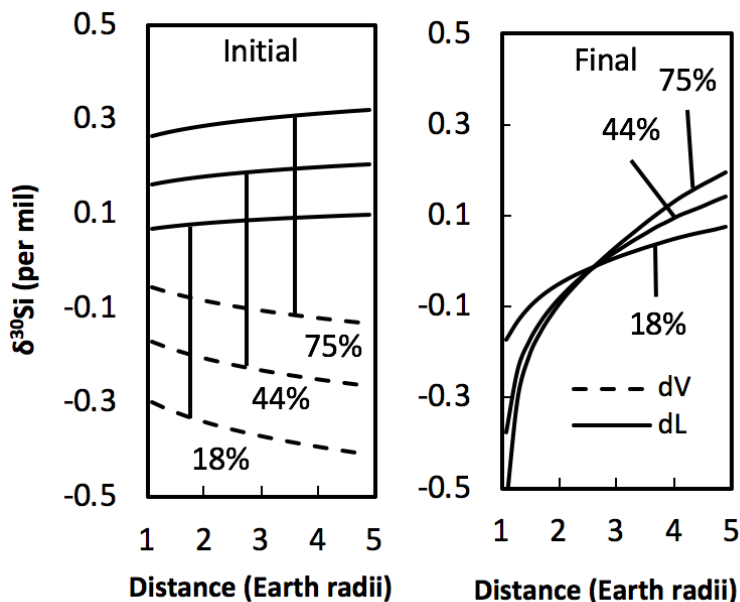
468 mixing with the hotter, more highly vaporized inner disk depletes FeO from the  
469 lunar-forming material, (ii) the magnitude of chemical fractionation is modestly  
470 dependent on initial disk vapor fraction (Figure 2b). This result can be  
471 understood because higher vapor fractions are associated with longer secular  
472 cooling timescales (§3.1), permitting the chemical redistribution to proceed  
473 longer, and because it implies a greater atmospheric mass relative to the mass  
474 of the quasi-stationary melt layer, permitting more efficient turbulent  
475 transport and steeper chemical gradients to develop per unit time. For the major  
476 elements here considered, the redistribution is small but non-negligible: for  
477 the range of initial vapor fractions considered for the proto-lunar disk, the  
478 resulting depletion in outer disk Fe/Fe+Mg is < 2x. Hence, impact models that  
479 begin with an Earth-mantle-like disk (Canup 2012; Čuk & Stewart 2012) cannot  
480 generate an FeO-enriched silicate Moon (Jones & Palme 2000) via turbulent  
481 redistribution: some other process is required (Pahlevan, et al. 2011).  
482 Nevertheless, due to uncertainties in both the initial Fe/Fe+Mg in the proto-  
483 lunar disk as well as in the lunar bulk chemical composition, modest chemical  
484 fractionation in the proto-lunar disk is difficult to rule out using major  
485 elements. We conclude that the major element composition of the Moon places  
486 only modest constraints on chemical fractionation processes in the proto-lunar  
487 disk.

488

### 489 3.3. Silicon Isotope Fractionation

490 Equilibrium silicon isotopic fractionation is significant, resulting in a ~0.4-  
491 0.5 per mil offset between the  $^{30}\text{Si}/^{28}\text{Si}$  of silicate liquids and vapors at the  
492 temperatures prevailing in the proto-lunar disk (Figure 3a). As discussed in  
493 §2.6, the proto-lunar liquid concentrates the heavy isotopes of silicon whereas  
494 the complementary vapor atmosphere is isotopically light, with the difference  
495 between the two reservoirs increasing with increasing radius, due to the  
496 negative radial temperature gradient (Figure 1b) and equilibrium partitioning  
497 law (Equation 9), which dictates enhanced fractionation at lower temperatures.  
498 The resulting composition of the atmosphere exhibits greater isotopic lightness  
499 with increasing radial distance and implies an inward transport of light silicon  
500 isotopes accompanying turbulent convection in the atmosphere. Liquid-vapor  
501 equilibration transmits this signal to the proto-lunar liquid and the process  
502 continues until the proto-lunar disk has condensed and the isotopic structure  
503 has frozen in. As with chemical fractionation (§3.2), steeper gradients are  
504 generated from hotter initial states (Figure 3b), the reasons being two-fold:  
505 longer cooling timescales and greater atmospheric masses causing greater

506 transport and redistribution during the cooling history as a by-product of  
 507 turbulent convection.

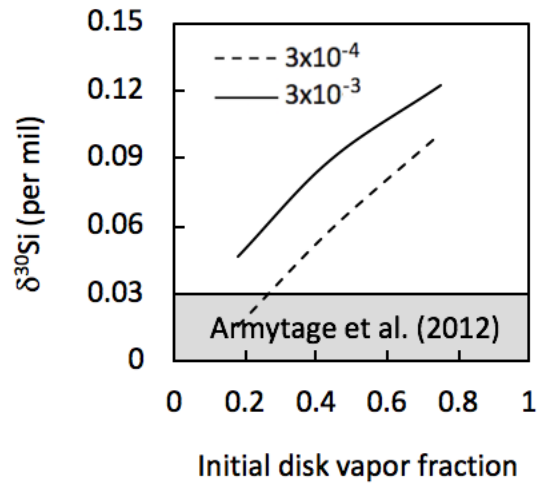


**Figure 3.** Silicon isotopic fractionation in the proto-lunar disk. (a) The initial radial isotopic structure of the proto-lunar disk atmosphere and liquid layer, with disks assumed to have a radially uniform initial composition set to zero for reference ( $\delta^{30}\text{Si}=0$ ). The number labels on the curves correspond to the initial disk vapor fraction. Equilibrium partitioning leads to a liquid layer that is isotopically heavy with a complementary isotopically light vapor atmosphere. (b) Final isotopic structure of the disk at the end of the thermal history when the vapor has condensed and the liquid composition has frozen in. The increasing isotopic lightness of the vapor atmosphere with radius implies an inward transport of light isotopes whose magnitude depends on the disk initial vapor fraction.

508 The magnitude of the silicon isotope fractionation is large relative to the  
 509 precision with which the composition of terrestrial and lunar reservoirs is  
 510 measured, permitting the development of a new stable isotopic constraint on the  
 511 evolution of the proto-lunar disk. As is also true for chemical fractionation  
 512 (§3.2), both more vigorous turbulence (higher  $\alpha$ ) and hotter initial thermal  
 513 states result in greater isotopic fractionation between the outer (i.e., Moon-  
 514 forming) and inner proto-lunar disk (Figure 4). For nominal values of turbulent



515 diffusivity ( $\alpha=3\times 10^{-3}$ ), even modestly vaporized proto-lunar disks ( $f_v\sim 0.2-0.4$ )  
 516 can imprint measurable mass-dependent signatures onto the lunar forming material  
 517 ( $\delta^{30}\text{Si}_M=0.05-0.1\%$  – Figure 4). An Earth-Moon isotopic offset of this magnitude  
 518 can be excluded with existing data (Armytage, et al. 2012; Fitoussi & Bourdon  
 519 2012). Possible interpretations of the data in light of the model results are  
 520 discussed in §4.4.



**Figure 4.** Summary of silicon isotopic redistribution in the proto-lunar disk. The average isotopic composition of the outer Moon-forming disk ( $3-5R_E$ ) is plotted as a function of the initial vapor fraction for two different values of the turbulent  $\alpha$  parameter. The initial composition of the disk ( $\delta^{30}\text{Si}$ ) is set to zero in these calculations for reference. In the absence of radial diffusivity, the final and initial composition of the outer disk would be identical, but for even modest levels of turbulence ( $\alpha=3\times 10^{-4}-3\times 10^{-3}$ ), measurable isotopic differences between the inner and outer disk can evolve. Measurements on terrestrial and lunar samples observe no  $\delta^{30}\text{Si}$  difference between the Earth and Moon ( $\Delta^{30}\text{Si-EM}=\delta^{30}\text{Si}_E-\delta^{30}\text{Si}_M$ ) at a precision of  $< 0.03$  per mil (Armytage, et al. 2012; Fitoussi & Bourdon 2012). Possible interpretations are discussed in §4.

521 4. DISCUSSION

522 We have described a coupled thermochemical model for the evolution of the proto-  
 523 lunar disk with the goal of forging a link between the stable isotopic  
 524 composition of the lunar-forming material and the parameters governing lunar

525 disk evolution. Here, we discuss several simplifying features of the adopted  
526 disk model, compare with other disk models discussed in the literature (§4.1-  
527 4.3), and then consider the general conclusions that can be drawn about the  
528 evolution of the proto-lunar disk using the constraints provided by the stable  
529 isotopic data (§4.4).

530

#### 531 4.1. Viscous Evolution

532 The model we have described neglects the viscous redistribution of mass and  
533 angular momentum and the attendant gravitational energy release. Including  
534 viscous evolution into the disk model would have several effects: (1) thermal:  
535 due to the depth of the terrestrial gravity well, viscous spreading has  
536 significant thermal consequences; where viscous heating takes place, it can  
537 easily dominate the heat budget of the melt-vapor column. For example, (Ward  
538 2012) described a disk model characterized by a stratified melt-vapor structure  
539 with a viscous melt layer and an inviscid atmosphere, and derived a cooling  
540 timescale for the proto-lunar disk of ~250 years, to be compared with a cooling  
541 timescale of ~2 years for a  $f_v \sim 0.2$  disk powered by secular cooling alone (§3.1).  
542 Hence, one consequence for the purely secular cooling powered disk model here  
543 described is that the lifetime – and the magnitude of chemical and isotopic  
544 redistribution calculated for the proto-lunar disk – are lower limits. The  
545 lifetime and degree of chemical fractionation is expected to be greater –  
546 perhaps much greater – in a model such as that described in (Ward 2012), (2)  
547 transport/redistribution: viscous dissipation causes net outward transport of  
548 angular momentum and net inward transport of mass and causes material to spread  
549 radially, a flow that competes with turbulent mixing (Stevenson 1990). Hence,  
550 by neglecting the viscous evolution, we implicitly only consider diffusive ( $D/v$   
551  $\geq 1$ ) transport regimes. We have focused on this regime because it is in this  
552 regime in which extensive turbulent mixing can take place (Pahlevan & Stevenson  
553 2007). However, even if a partially vaporized disk is initially in the diffusive  
554 regime, it is possible that – as the disk condenses – it transitions to the  
555 viscous regime ( $D/v \leq 1$ ) and radial mixing becomes inefficient. The calculations  
556 here presented only apply to redistribution in the diffusive regime.

557

#### 558 4.2. Heat and Entropy Transport

559 We have considered turbulent transport in the presence of compositional  
560 gradients, but neglected the equivalent transport of heat in the presence of  
561 entropy gradients. It is known that – in the presence of radial entropy gradients  
562 – a disk instability may arise at the expense of the entropy gradient (“radial  
563 convection”) and thereby radially transport heat and entropy (Klahr &

564 Bodenheimer 2003; Lesur & Papaloizou 2010). In the proto-lunar disk, such a  
565 radial entropy gradient may arise via preferential cooling or vertical phase  
566 separation of droplets ("rainout") in adjacent annuli of the proto-lunar disk.  
567 This process would have consequences for the calculations here presented: (1)  
568 thermal: since heat transport is outward, to the extent that this process takes  
569 place, it prolongs the thermal history and accentuates the chemical/isotopic  
570 signals that arise, and (2) compositional: since the effective gravity in the  
571 near-Keplerian disk is inwards, this process would also buoyantly exchange  
572 vapor-rich (isotopically light) atmospheric parcels with more droplet-laden  
573 (isotopically heavy) parcels radially and may therefore moderate the magnitude  
574 of radial transport here calculated and soften the constraint here described.  
575 However, we see no reason to suppose that this effect would precisely cancel  
576 the isotopic fractionation based on the radial temperature gradient calculated  
577 in this work. In the absence of such a reason, the possibility of a fortuitous  
578 cancellation of two competing effects is considered unlikely. The constraints  
579 on disk evolution are therefore considered robust to inclusion of this process.

580

#### 581 4.3. Criterion for Compositional Freeze-in

582 Whereas adopting initial conditions for disk evolution from the output of giant  
583 impact simulations is straightforward, selecting a criterion for the cessation  
584 of disk evolution is less certain. Here, we have considered a disk with chemical  
585 and isotopic redistribution but no net mass transport and no fragmentation. In  
586 such a model, the condensation of the vapor atmosphere can be expected to halt  
587 radial redistribution because the turbulent atmosphere is the agent that causes  
588 transport. However, other criteria for compositional freezing of the disk are  
589 possible. (Canup et al. 2015) considered a settled disk with a viscous liquid-  
590 rich midplane layer. In analogy with particle disks, the midplane liquid layer  
591 was assumed to undergo gravitational instability and fragmentation to form  
592 moonlets beyond the classical Roche radius at  $2.9 R_E$  defined by lunar-density  
593 condensates. (Thompson & Stevenson 1988) instead considered a disk model in  
594 which a single vertical isentrope characterized convective columns from the  
595 midplane up to the rarified regions where the disk atmosphere is optically thin  
596 and radiative. In such a picture, radiative cooling in disk patches is balanced  
597 by gravitational energy release until the material spreads to radii where this  
598 equality can no longer be satisfied, at which point the patch instabilities  
599 lead to collapse and moonlet formation and the composition of the moonlets is  
600 the composition of the disk fragment with its constituent droplets and vapor.  
601 Regardless of the precise criterion for compositional freeze-in for disk  
602 patches, it is clear that the liquid composition is effectively frozen-in once

603 the atmosphere condenses. Moreover, satellite-disk interaction may be important  
604 (Salmon & Canup 2012) but its coupling to disk evolution is relegated to future  
605 works.

606

#### 607 4.4. Consequences for Disk Evolution

608 We do not know – a priori – how the proto-lunar disk evolved. Here, we have  
609 considered a disk model in which the liquid settles out into a midplane layer  
610 that rapidly equilibrates with the overlying atmosphere, with the turbulent  
611 diffusivity in the vapor atmosphere operating on a shorter timescale than  
612 viscous dissipation in either the atmosphere (Carballido, Desch, & Taylor 2016)  
613 or the melt-rich layer (Ward 2012). This is the same settling and transport  
614 regime originally envisaged in the equilibration hypothesis (Pahlevan &  
615 Stevenson 2007) and therefore provides constraints on any evolutionary scenario  
616 of this kind. We have shown that in such a picture the proto-lunar disk is  
617 expected to generate isotopic gradients that would be expressed as differences  
618 in the composition of the Earth's mantle and Moon. Such an Earth-Moon isotopic  
619 offset is not observed to a precision of 30 ppm in the case of silicon ( $^{30}\text{Si}/^{28}\text{Si}$ )  
620 (Armytage, et al. 2012; Fitoussi & Bourdon 2012). In light of the isotopic data,  
621 the model results can be interpreted in four ways: (1) imperfect settling: the  
622 proto-lunar liquid may fail to settle into a liquid-rich layer with a separately  
623 convective atmosphere and instead, exchange between the midplane and vapor  
624 atmosphere may be advective (e.g., convective overshoot), as also described in  
625 (Thompson & Stevenson 1988). In such a scenario, equilibration and mixing with  
626 the Earth would involve both a vapor atmosphere as well as the suspended liquid  
627 droplets, and one would expect isotopic homogeneity in the Earth-Moon system  
628 for all elements irrespective of volatility (Zhang, et al. 2012), (2) liquid-  
629 vapor equilibration: we have assumed that liquid-vapor equilibrium in the system  
630 is rapid relative to transport timescales. Although the vigor of convective  
631 motion makes this possible, the timescale for liquid-vapor equilibration may  
632 depend on foam-physics at the liquid-vapor interface and is therefore difficult  
633 to quantify. Hence, one interpretation of the silicon isotopic data is that the  
634 proto-lunar disk liquid and vapor underwent phase separation into two distinct  
635 layers but did not experience continuous and rapid chemical re-equilibration.  
636 Although large depletions of volatile elements in the lunar material suggest  
637 that nearly all proto-lunar liquid equilibrated with some vapor, a timescale  
638 for liquid-vapor equilibration comparable to evolutionary timescales cannot be  
639 ruled out, (3) other transport regimes: we have restricted the discussion here  
640 to transport regimes in which the total diffusivity is greater than the total  
641 viscosity ("the diffusive regime") such that mixing can outcompete disk

642 spreading and homogenize the vapor atmosphere on a rapid timescale. This regime  
643 may be appropriate for a disk whose dominant instability is the convective  
644 instability, because convection in disks is known to produce turbulent mixing  
645 but weak angular momentum transport (Lesur & Ogilvie 2010; Stone & Balbus 1996).  
646 However, there exist other instabilities, e.g., the gravitational instability,  
647 that may entail different mixing and transport properties. Hence, it is also  
648 possible that the disk spent part of its evolution in the viscous regime not  
649 subject to the diffusive redistribution process here calculated, (4) high-  
650 temperature silicate thermodynamics: it is possible that a binary olivine  
651 thermodynamics does not accurately capture the vaporization behavior of silicon  
652 for the full range of thermal states experienced by the proto-lunar disk. In  
653 particular, olivine is known to vaporize congruently (Nagahara, et al. 1994)  
654 such that the degree of bulk vaporization and the degree of silicon vaporization  
655 are equivalent in an olivine thermodynamics. A more complex multi-component  
656 model may not display this behavior, instead revealing silicon isotopes to be  
657 highly sensitive to the evolution of the proto-lunar disk for only a range of  
658 vapor fractions. The degree to which olivine thermodynamics accurately captures  
659 the vaporization behavior of any given element in the proto-lunar disk can be  
660 determined by development of more complex models of multi-component silicate  
661 melts at the temperatures ( $T=3,000-4,000$  K) of relevance.

662

## 663 5. CONCLUSIONS

664 Recent isotopic observations on lunar and terrestrial rocks indicate that the  
665 lunar material is derived from the silicate Earth. The disk stage of lunar  
666 origin has been invoked to explain several observed features of the Earth-Moon  
667 system, including the precise isotopic homogeneity (Lock, et al. 2018; Pahlevan  
668 & Stevenson 2007) and the lunar volatile depletion (Canup, et al. 2015; Nie &  
669 Dauphas 2019). Nevertheless, a quantitatively constrained description of proto-  
670 lunar disk evolution is lacking. Despite uncertainty in the initial thermal  
671 state, the settling and transport regimes, and the degree of thermal and  
672 chemical coupling to the post-impact Earth, it is well established that the  
673 proto-lunar disk experiences a two-phase (liquid-vapor) evolution. Here, we  
674 have developed a thermochemical scenario of disk evolution incorporating silicon  
675 isotopes as passive tracers. Our results demonstrate that silicon isotopic  
676 fractionation in the proto-lunar disk is significant relative to the precision  
677 of existing measurements, yielding a new constraint on disk evolution. The  
678 principal conclusion of the model is that a “settled” proto-lunar disk in the  
679 diffusive regime would have developed isotopic offsets in the silicate EM system  
680 that are not observed. Hence, it is possible to use silicon isotopes to set

681 constraints on Earth-disk equilibration: if system-wide turbulent mixing of the  
682 Earth-Moon system took place, it must have involved advective mixing of the  
683 proto-lunar liquid and the vapor atmosphere (e.g., with suspended droplets).  
684 More generally, the silicon isotopic identity of Earth's mantle and Moon poses  
685 a challenge to any model of the proto-lunar disk in which the liquid and vapor  
686 experience divergent fates, with one phase being preferentially reaccreted to  
687 the Earth (Canup, et al. 2015; Charnoz & Michaut 2015; Lock, et al. 2018; Nie  
688 & Dauphas 2019; Wang et al. 2019). Whether these evolutionary scenarios can be  
689 reconciled with the silicon isotopic homogeneity of the EM system remains to be  
690 investigated. These results open a new quantitative window into the evolutionary  
691 aftermath of the lunar-forming giant impact.

692

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696 like to dedicate this paper to the memory of Bill Ward, who pioneered the study  
697 of the proto-lunar disk.

698

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