

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

3 Kaveh Pahlevan^{1,2}, Charles F. Gammie³
4
5
6
7

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.
32
33
34
35
36

¹ Corresponding author kpahlevan@seti.org

² Carl Sagan Center, The SETI Institute, 189 Bernardo Ave, Suite 200, Mountain View, CA 94043 USA

³ Department of Astronomy, University of Illinois at Urbana-Champaign, 1002 West Green Street, Urbana, IL 61801, USA

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 ν , 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.

117

118 2. DISK MODEL

119 Currently considered scenarios for a single Moon-forming giant impact (Canup
120 2004, 2012; Canup & Asphaug 2001; Āuk & 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.

132

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 ρ 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, σ_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 ($\equiv \sigma_v / \sigma_T$), Ω 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:

190

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

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 μ the vapor mean molecular weight. By
 194 substituting nominal parameter values for the proto-lunar disk ($\sigma_T = 5 \times 10^6$ g/cm²,
 195 $\Omega = 2 \times 10^{-4}$ s⁻¹, $c_s = 10^5$ cm/s) into both terms in Equation (2), the dominance of
 196 central gravity in determining disk vertical structure can be confirmed.⁴ 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:

206

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

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

⁴ 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 SiO_2 , SiO , Si , FeO , Fe , MgO , Mg , O_2 , and 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 (≈ 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 , ρ_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:
 297

298

$$\sigma_v \frac{\partial c_{i,v}}{\partial t} = - \nabla \cdot (\sigma_v D_v \nabla c_{i,v}) \quad (7)$$

299

300

301

302

303

304

305

306

307

308

309

310

311

312

313

314

315

316

2.5. Thermal Evolution

317

318

319

320

321

322

323

324

325

326

327

328

329

330

331

332

333

where $\sigma_v (=f_v \sigma_T)$ is the atmospheric surface density [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.

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 ~ 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 σ_{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 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 SiO₂ 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 ³⁰Si/²⁸Si between the
 400 liquid and vapor, as previously calculated using standard procedures (Pahlevan,
 401 et al. 2011) and δ_L , δ_V , δ_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, Δ_{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 δ_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.

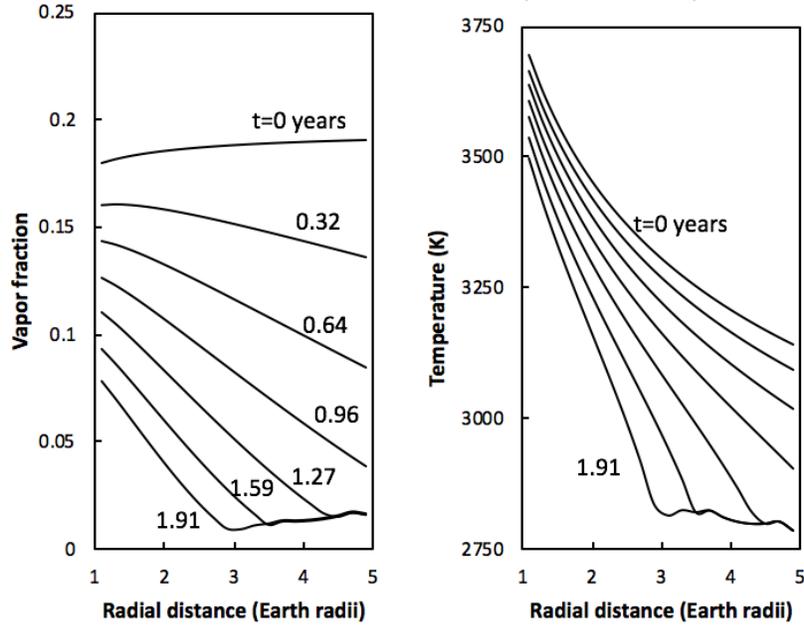


Figure 1. Disk thermal state and evolution. (a) Disk vapor fraction (atmospheric mass / total disk column mass) versus radial distance. This cooling history corresponds to an initial condition ($f_v \sim 0.2$) generated via the “standard” impact. The simulations are halted at $f_v \sim 0.01$ for numerical 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 $[(\text{Fe}, \text{Mg})_2\text{SiO}_4]$ thermodynamic model adopted has one variable
 446 for chemical composition, $x_{\text{Fe}} (\equiv \text{Fe}/\text{Fe}+\text{Mg})$, 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_2SiO_4) has a higher vapor pressure than forsterite
 450 (Mg_2SiO_4) 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 ($x_{\text{Fe}}=0.12-0.35$), while the liquid layer is FeO-depleted ($x_{\text{Fe}}=0.02-0.06$),
 453 relative to the composition of the column as a whole ($x_{\text{Fe}}=0.1$, 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.

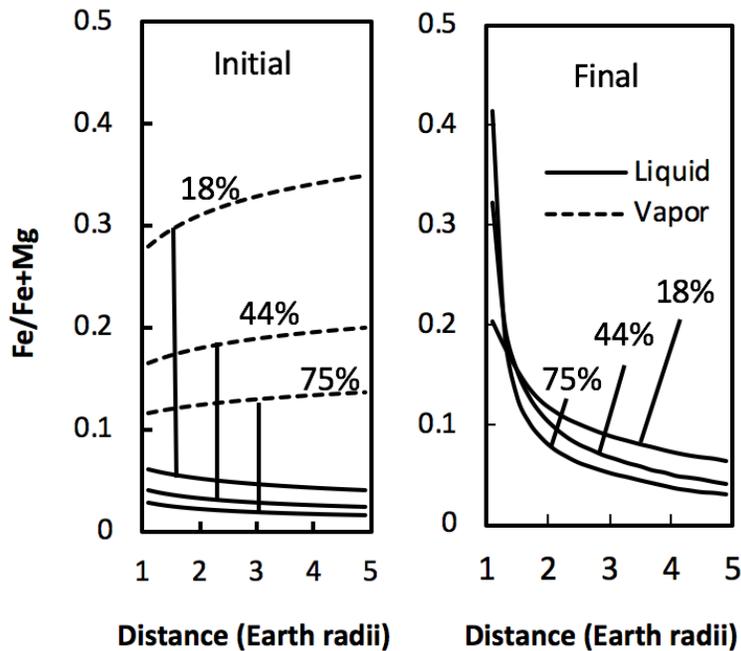


Figure 2. Fe/Mg redistribution in the proto-lunar disk. (a) Initial liquid-vapor partitioning in a disk with bulk $x_{\text{Fe}}(\text{Fe}/\text{Fe}+\text{Mg})=0.1$. The percentage labels on curves correspond to initial disk vapor fraction. (b) Final compositional structure of the disk when the vapor 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.

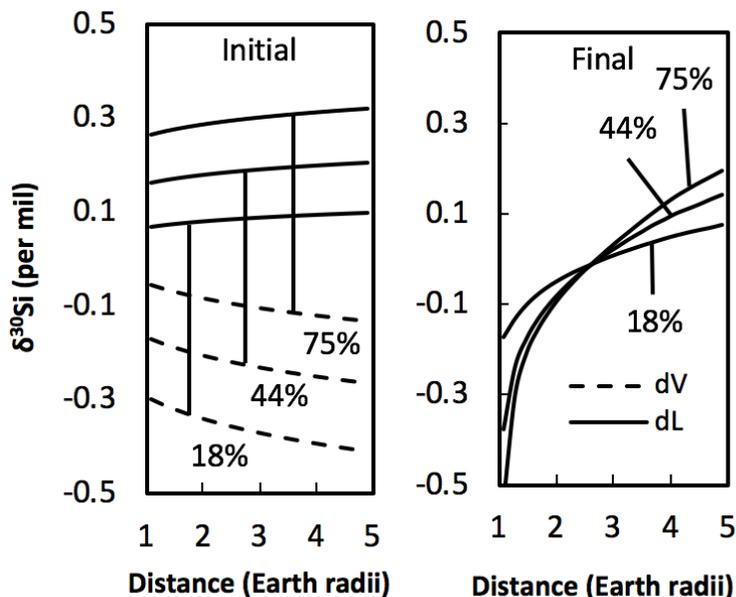


Figure 3. Silicon isotopic fractionation in the proto-lunar disk. (a) The initial radial isotopic structure of the proto-lunar disk atmosphere and liquid layer, with disks assumed to have a radially uniform initial composition set to zero for reference ($\delta^{30}\text{Si}=0$). The number labels on 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 α) 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.

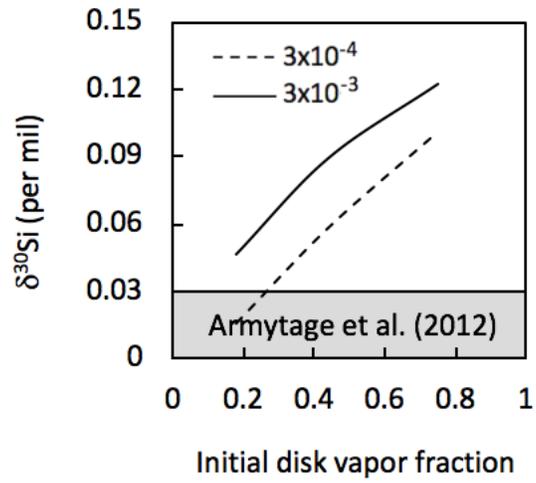


Figure 4. Summary of silicon isotopic redistribution in the proto-lunar disk. The average isotopic composition of the outer Moon-forming disk ($3-5R_E$) is plotted as a function of the initial vapor fraction for two different values of the turbulent α parameter. The initial composition of the disk ($\delta^{30}\text{Si}$) 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 ≥ 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

693 ACKNOWLEDGEMENTS

694 This work was supported in part by the Poincaré Fellowship. K.P. acknowledges
695 discussions with David Stevenson, Robin Canup, and Sebastien Charnoz. We would
696 like to dedicate this paper to the memory of Bill Ward, who pioneered the study
697 of the proto-lunar disk.

698

699 REFERENCES

- 700 Armytage, R. M. G., Georg, R. B., Williams, H. M., & Halliday, A. N. 2012, *Geochimica Et*
701 *Cosmochimica Acta*, 77, 504
702 Atobe, K., & Ida, S. 2007, *Icarus*, 188, 1
703 Cameron, A. G. W., & Ward, W. R. 1976, *Lunar Sci*, 7, 120
704 Canup, R. M. 2004, *Icarus*, 168, 433
705 ---. 2012, *Science*, 338, 1052
706 Canup, R. M., & Asphaug, E. 2001, *Nature*, 412, 708
707 Canup, R. M., Barr, A. C., & Crawford, D. A. 2013, *Icarus*, 222, 200
708 Canup, R. M., Visscher, C., Salmon, J., & Fegley Jr, B. 2015, *Nature Geosci*, 8, 918
709 Carballido, A., Desch, S. J., & Taylor, G. J. 2016, *Icarus*, 268, 89
710 Carballido, A., Stone, J. M., & Pringle, J. E. 2005, *Monthly Notices of the Royal Astronomical*
711 *Society*, 358, 1055
712 Charnoz, S., & Michaut, C. 2015, *Icarus*, 260, 440
713 Čuk, M., Hamilton, D. P., Lock, S. J., & Stewart, S. T. 2016, *Nature*, 539, 402
714 Čuk, M., & Stewart, S. T. 2012, *Science*, 338, 1047
715 Fitoussi, C., & Bourdon, B. 2012, *Science*, 335, 1477
716 Gammie, C. F., Liao, W.-T., & Ricker, P. M. 2016, *The Astrophysical Journal*, 828, 58
717 Genda, H., & Abe, Y. 2003, *Earth Planets and Space*, 55, 53
718 Hartmann, W. K., & Davis, D. R. 1975, *Icarus*, 24, 504
719 Holton, J. R. 1992, *An Introduction to Dynamic Meteorology* (San Diego: Academic Press)
720 Huang, F., Wu, Z., Huang, S., & Wu, F. 2014, *Geochimica et Cosmochimica Acta*, 140, 509

721 Jones, J. H., & Palme, H. 2000, in *Origin of the Earth and Moon*, eds. K. Righter, & R. M. Canup
722 (Tucson, AZ: University of Arizona Press), 197
723 Klahr, H. H., & Bodenheimer, P. 2003, *The Astrophysical Journal*, 582, 869
724 Kruijer, T. S., & Kleine, T. 2017, *Earth and Planetary Science Letters*, 475, 15
725 Lesur, G., & Ogilvie, G. I. 2010, *Monthly Notices of the Royal Astronomical Society: Letters*, 404,
726 L64
727 Lesur, G., & Papaloizou, J. C. 2010, *Astronomy & Astrophysics*, 513, A60
728 Lock, S. J., Stewart, S. T., Petaev, M. I., Leinhardt, Z., Mace, M. T., Jacobsen, S. B., & Cuk, M. 2018,
729 *Journal of Geophysical Research: Planets*, 123, 910
730 Machida, R., & Abe, Y. 2004, *Astrophysical Journal*, 617, 633
731 Nagahara, H., Kushiro, I., & Mysen, B. O. 1994, *Geochimica Et Cosmochimica Acta*, 58, 1951
732 Nakajima, M., & Stevenson, D. J. 2014, *Icarus*, 233, 259
733 Nie, N. X., & Dauphas, N. 2019, *The Astrophysical Journal Letters*, 884, L48
734 Pahlevan, K. 2014, *Philosophical Transactions of the Royal Society a-Mathematical Physical and*
735 *Engineering Sciences*, 372
736 Pahlevan, K., & Morbidelli, A. 2015, *Nature*, 527, 492
737 Pahlevan, K., & Stevenson, D. J. 2007, *Earth and Planetary Science Letters*, 262, 438
738 Pahlevan, K., Stevenson, D. J., & Eiler, J. M. 2011, *Earth and Planetary Science Letters*, 301, 433
739 Reufer, A., Meier, M. M. M., Benz, W., & Wieler, R. 2012, *Icarus*, 221, 296
740 Refu, R., & Canup, R. M. 2020, *Journal of Geophysical Research: Planets*, 125, e2019JE006312
741 Salmon, J., & Canup, R. M. 2012, *Astrophysical Journal*, 760
742 ---. 2014, *Philosophical Transactions of the Royal Society a-Mathematical Physical and*
743 *Engineering Sciences*, 372
744 Stevenson, D. J. 1990, *Astrophysical Journal*, 348, 730
745 Stone, J. M., & Balbus, S. A. 1996, *Astrophysical Journal*, 464, 364
746 Taylor, G. J., & Wieczorek, M. 2014, *Philosophical Transactions of the Royal Society a-*
747 *Mathematical Physical and Engineering Sciences*, 372
748 Tennekes, H., & Lumley, J. L. 1972, *A First Course on Turbulence* (Cambridge, Massachusetts: The
749 MIT Press)
750 Thompson, C., & Stevenson, D. J. 1988, *Astrophysical Journal*, 333, 452
751 Tian, Z., & Wisdom, J. 2020, *Proceedings of the National Academy of Sciences*, 117, 15460
752 Tian, Z., Wisdom, J., & Elkins-Tanton, L. 2017, *Icarus*, 281, 90
753 Touma, J., & Wisdom, J. 1998, *Astronomical Journal*, 115, 1653
754 Urey, H. C. 1947, *Journal of the Chemical Society*, 562
755 Visscher, C., & Fegley, B. 2013, *Astrophysical Journal Letters*, 767
756 Wang, X., Fitoussi, C., Bourdon, B., Fegley, B., & Charnoz, S. 2019, *Nature Geoscience*, 12, 707
757 Ward, W. R. 2012, *Astrophysical Journal*, 744
758 Ward, W. R., & Cameron, A. G. W. 1978, in *Lunar and Planetary Science Conference IX* (Houston,
759 Texas)
760 Ward, W. R., & Canup, R. M. 2000, *Nature*, 403, 741
761 Wisdom, J., & Tian, Z. L. 2015, *Icarus*, 256, 138
762 Zambardi, T., Poitrasson, F., Corgne, A., Meheut, M., Quitte, G., & Anand, M. 2013, *Geochimica*
763 *Et Cosmochimica Acta*, 121, 67
764 Zhang, J. J., Dauphas, N., Davis, A. M., Leya, I., & Fedkin, A. 2012, *Nature Geoscience*, 5, 251

