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5 **Volcanically-Induced Transient Atmospheres on the Moon:**
6 **Assessment of Duration, Significance and Contributions to Polar Volatile**
7 **Traps**
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19 **Key Points:**

- 20 • A transient lunar atmosphere from peak volcanic degassing lasting up to ~70 Ma
21 was recently proposed as a source of lunar polar volatiles.
- 22 • We forward-model individual eruption volume, degassing patterns, and duration
23 of periods between eruptions (repose periods), finding that:
- 24 • Transient, volcanically-induced atmospheres are inefficient sources for volatile
25 delivery to permanently shadowed lunar polar regions.
26

27 **Abstract**

28 A transient lunar atmosphere formed during a peak period of volcanic outgassing and
29 lasting up to about ~70 Ma was recently proposed. We utilize forward-modeling of
30 individual lunar basaltic eruptions and the observed geologic record to predict eruption
31 frequency, magma volumes, and rates of volcanic volatile release. Typical lunar mare
32 basalt eruptions have volumes of $\sim 10^2$ - 10^3 km³, last less than a year, and have a rapidly
33 decreasing volatile release rate. The total volume of lunar mare basalts erupted is small
34 and the repose period between individual eruptions is predicted to range from 20,000-
35 60,000 years. Only under very exceptional circumstances could sufficient volatiles be
36 released in a single eruption to create a transient atmosphere with a pressure as large as
37 ~0.5 Pa. The frequency of eruptions was likely too low to sustain any such atmosphere
38 for more than a few thousand years. Transient, volcanically-induced atmospheres were
39 probably inefficient sources for volatile delivery to permanently shadowed lunar polar
40 regions.

41

42 **Plain Language Summary**

43 Could gas emitted from volcanic eruptions during the most intense and voluminous
44 period of lunar mare volcanism produce a temporary lunar atmosphere? Could the
45 presence of such an atmosphere enable volatiles to reach the cold traps in the
46 permanently shadowed regions at the lunar poles? We use information from lunar
47 geology and sample analyses to predict the number of eruptions with time, the volume of
48 individual eruptions, the rates of volcanic gas release during each eruption, and the time
49 between eruptions. We find that only under rare circumstances could a single eruption or
50 two eruptions closely-spaced in time release enough gas to create a transient atmosphere
51 with a pressure as large as ~0.5 Pa. Furthermore, it is difficult to sustain such an
52 atmosphere for more than a few thousand years. These results suggest that volcanically-
53 produced atmospheres are inefficient source mechanisms for delivery of volatiles to form
54 deposits in permanently shadowed polar regions of the Moon; this favors volatile-rich
55 impactors as the major source of polar ice.

56

57 **1 Introduction**

58 The current atmosphere of the Moon is a stable, low-density surface boundary
59 exosphere ($\sim 10^{-12}$ mbar) (Stern, 1999; Cook et al., 2013; Benna et al., 2015) and is
60 thought to have changed little in the last several billion years. Volcanism, a significant
61 source of volatile supply to planetary atmospheres throughout planetary history, is known
62 to have been much more important in early lunar history (mare basalt volcanism; Shearer
63 et al., 2008; Head and Wilson, 2017), spanning from over 4 billion years ago (beginning
64 with cryptomaria; Whitten et al., 2015a,b), reaching peak fluxes between 3 and 4 Ga, and
65 declining to much lower levels between 3 and 1 Ga (Hiesinger et al., 2011; Pasckert et
66 al., 2015; Head et al., 2020).

67 Needham and Kring (2017) assessed lunar mare basalt volcanic flux estimates and
68 volatile release abundances to address whether these patterns might lead to a transient or
69 sustained lunar atmosphere early in lunar history. Using the distribution and quantity of
70 mare basalt fill, and estimates of its age, they calculated the magma flux (the volume of
71 mare basalt erupted as a function of time), and then estimated the corresponding release

72 rate of volatiles on the basis of estimates derived from the analysis of lunar samples (e.g.,
 73 Saal et al., 2008; Rutherford and Papale, 2009; Hauri et al., 2011; Kring, 2014). Using
 74 estimates of mare basalt unit ages (e.g., Hiesinger et al., 2011) and thicknesses (Weider et
 75 al., 2010), Needham and Kring (2017) concluded that during a period of peak mare
 76 emplacement and volcanic volatile release at ~3.5 Ga (Figs. S1a,b), the maximum
 77 atmospheric pressure at the lunar surface could have reached ~1 kPa (~1.5 times greater
 78 than the current atmospheric surface pressure of Mars) (Fig. S1c) and that this lunar
 79 atmosphere could have persisted for ~70 million years before fully dissipating (Fig. S1c).
 80 They further pointed out that even though most of the volcanically-released volatiles will
 81 have been lost to space, if only 0.1% of the water released during these eruptions
 82 migrated to the permanently shadowed polar regions of the Moon, then the resulting
 83 hydrogen mass could account for the entire currently observed hydrogen deposits located
 84 there (Eke et al., 2009; Livengood et al., 2018).

85 We adopt a different approach, using improved models of the generation, ascent
 86 and eruption of lunar basaltic magma (Wilson and Head, 2017), to predict flow volumes,
 87 eruption frequencies (Head and Wilson, 2017), and temporal magmatic volatile release
 88 patterns in individual eruptions (Rutherford et al., 2017; Wilson and Head, 2018). Key
 89 components of this analysis are 1) the range (and mean value) of magma volumes erupted
 90 in individual eruptions, 2) the masses, and hence volumes, of the various gases released
 91 in any one eruption, 3) the duration of the eruption and the gas release rate (varying
 92 significantly as the eruption progresses), 4) the typical time intervals between eruptions
 93 (repose periods) as a function of geologic time and 5) the timescale for the dissipation of
 94 an atmosphere once one is emplaced. We review the geological basis for the first four
 95 components, examine the potential time-dependence and variability of gas release in an
 96 individual typical eruption, and finally address the question: *Are these gas-release values*
 97 *sufficient to form a transient atmosphere and, if so, for what duration?* We then compare
 98 our findings with the broad-scale, time-averaged peak flux estimates of Needham and
 99 Kring (2017) (see the Supporting Material), and address similarities and differences and
 100 their causes, and how estimates might be refined in the future. We conclude by assessing
 101 whether the forward-modeling predictions of gas-release rates are sufficient to: 1) act as a
 102 significant supply of volatiles to the permanently shadowed lunar polar cold-trap regions
 103 and 2) form a transient lunar atmosphere for a period sufficient to favor astrobiological
 104 activity as suggested by Schulze-Makuch and Crawford (2018).
 105

106 **2 Forward Modeling Lunar Mare Basaltic Eruptions**

107 Wilson and Head (2017) and Head and Wilson (2017) improved earlier
 108 theoretical models for the generation, ascent and eruption of basaltic magma on the Moon
 109 (Wilson and Head, 1981; Head and Wilson, 1992) by using new data on crustal thickness
 110 and density (Wieczorek et al., 2013), magma volatile inventories (Rutherford et al.,
 111 2017), and surface morphology, topography and structure (from Lunar Reconnaissance
 112 Orbiter). They showed that ongoing partial melting in buoyant diapirs deep in the mantle
 113 overpressurizes the source regions, producing sufficient stress to cause brittle fracturing;
 114 a magma-filled crack grows, disconnects from its source and propagates to the surface as
 115 a blade-shaped, convex-upward dike. The typical turbulent magma rise speeds that result
 116 are ~10 to a few tens of m s^{-1} , dike widths are ~100 m, and eruption rates from 1-10 km
 117 long fissure vents are $\sim 10^5$ to $10^6 \text{ m}^3 \text{ s}^{-1}$. Lunar eruption volume fluxes derived from lava

118 sinuous rille lengths and depths or flow thicknesses and surface slopes are $\sim 10^5$ to 10^6 m³
 119 s⁻¹ (volume-limited lava flows) and $>10^4$ to 10^5 m³ s⁻¹ (rilles). The volume of magma
 120 released in one event is predicted to be in the range 10^2 - 10^3 km³ (Wilson and Head, 2017;
 121 Head and Wilson, 2017). Thus, if all the magma were extruded from these dike events
 122 and spread evenly across the surface in 25 m thick flows, they would occupy areas of
 123 4000 to 40,000 km², well within the range of thicknesses and areas of mapped and dated
 124 (e.g., Hiesinger et al., 2011) lunar mare lava flows. We now summarize aspects of these
 125 treatments to derive the relevant parameters.

126 2.1) Individual mare basalt eruption volumes: Range and typical values:
 127 Individual eruption volumes of typical visible, and therefore most recent, lava flow
 128 deposits, are at least ~ 200 - 300 km³ (Head and Wilson, 2017) (Table 1a). In addition,
 129 Head and Wilson (1981) estimated the minimum volume of lava, ~ 100 km³, needed to
 130 thermo-mechanically erode the preserved sinuous rille channels. Applying the same
 131 method to the largest lunar rille, Schroeter's Valley, implies a volume of 2000 km³ and a
 132 duration of ~ 150 days. On the basis of these predictions and observations, we adopt a
 133 range of individual eruption volumes, V , of 100 - 2000 km³ (Table 1a), with typical values
 134 in the range 100 - 300 km³.

135 2.2) Total mare basalt erupted volumes: Using mare basin lava fill depth
 136 estimates, the total volume, V_t , of all volcanic products erupted on the Moon over its
 137 lifetime is $\sim 10^7$ km³ (Head and Wilson, 1992; Evans et al., 2016). The absolute dates of
 138 specific eruptions are unknown, but crater size-frequency distribution-derived dates of
 139 units mapped from orbit, and stratigraphic relationships, imply that the overall time span
 140 of the vast majority of lunar volcanic activity was ~ 2 Ga (Hiesinger et al., 2011; Head
 141 and Wilson, 2017; Head et al., 2020).

142 2.3) Number of eruptions, average eruption rates, and estimated repose periods:
 143 Using the 100 - 300 km³ average eruption volume, the $\sim 10^7$ km³ total erupted volume of
 144 mare basalts, and the ~ 2 Ga duration of volcanism, we calculate a total of $\sim 30,000$ to
 145 $100,000$ eruptions with an average repose period of $20,000$ to $60,000$ years. These repose
 146 times assume that eruptions occur randomly in space and time, in which case two
 147 eruptions might occur with a much smaller time interval; however, we show in the
 148 Supplementary Material that eruptions at intervals close enough in time to influence our
 149 conclusions will be rare. Accounting for lunar thermal evolution (conductive cooling and
 150 lithospheric thickening) in terms of mare mantle production rates and the evolving
 151 lithospheric stress state and magnitude (Head and Wilson, 2017), we would predict
 152 decreasing volumes of magma with time. If three times as much magma was erupted in
 153 the 4 - 3 Ga period than in the 3 - 2 Ga period, for example, the earlier eruptions would have
 154 occurred every $13,000$ to $40,000$ years.

155 2.4) Eruption durations: Analyses of the dynamics of lunar eruptions allow us to
 156 estimate the volume fluxes, F_1 , of lava forming surface flows and sinuous rilles (Wilson
 157 and Head 2017; Head and Wilson, 2017); coupled with the typical erupted volumes
 158 described above, these give values for the typical durations, τ_e , of these eruptions (Table
 159 1a), all less than 6 months, with most eruption durations in the 1 - 3 month range.

160 2.5) Magmatic volatiles and volatile release patterns: Analyses of lavas and
 161 pyroclastics sampled by the Apollo missions (Saal et al., 2008; Rutherford and Papale,
 162 2009; Hauri et al., 2011; Chen et al., 2015; Rutherford et al., 2017; Renggli et al., 2017;
 163 Ni et al., 2019) provide estimates for the compositions and amounts of released volatiles.

164 The highest amount is that for picritic magmas, ~3400 ppm (Rutherford et al., 2017). At
 165 the other extreme, Head and Wilson (2017) found that the radii of the lava ponds feeding
 166 the lava flows eroding sinuous rille channels imply total magma volatile contents of no
 167 more than 700 ppm. We adopt 2000 ppm, close to the average of these extremes. The
 168 Rutherford et al. (2017) volatiles are CO, H₂O, SO₂, H₂S, COS and F present in amounts
 169 n_i of 1395, 1133, 327, 168, 327 and 50 ppm, respectively, and with molecular masses m_i
 170 of 28.0, 18.0, 64.1, 34.1, 60.1 and 19.0, respectively, the mean molecular weight is $\Sigma (n_i$
 171 $m_i) / \Sigma n_i = 31.4 \text{ kg kmol}^{-1}$. The corresponding values for alternative compositions
 172 suggested by Renggli et al. (2017) and Newcombe et al. (2017) are 48.9 and 22.2 kg
 173 kmol^{-1} , respectively; we adopt the Rutherford et al. (2017) value as typical for subsequent
 174 calculations.

175 2.6) Volatile input to the atmosphere: We first calculate the *total volume of gas*
 176 *released* from an eruption of a specific volume, and then analyze the *time-history of gas*
 177 *release in the several phases of an individual eruption* (Rutherford et al., 2017; Wilson
 178 and Head, 2018). Multiplying the dense-rock-equivalent erupted volume V by the typical
 179 density of lunar basaltic magma, $\rho_m = \sim 3000 \text{ kg m}^{-3}$, yields the magma mass erupted, and
 180 multiplying that by the total released gas mass fraction $n_t = \Sigma n_i$ gives the total gas mass
 181 released, M_g . Finally dividing M_g by τ_e yields the average gas mass input rate to the
 182 atmosphere, F_g . Table 1a summarizes these values. However, gas release during an
 183 eruption is non-linear, and typically declines with time (Wallace et al., 2015). Speciation,
 184 relative abundances and fluxes of specific volatiles can vary during a single eruption.
 185 Could such variations in individual volcanic eruptions result in spikes in volatile output
 186 contributing to an atmosphere that might be underestimated by deriving an average value
 187 for the entire eruption? We now employ an updated version of a recent model of the
 188 typical phases of a lunar eruption to assess these questions.

189 Wilson and Head (2018), using data from Rutherford et al. (2017), assessed mare
 190 basalt gas release patterns during individual volcanic eruptions as a basis for predicting
 191 the effect of sequential gas production, bubble nucleation and growth, magma and gas
 192 rise rates, bubble coalescence, and magma disruption processes. Subdividing typical
 193 lunar eruptions into four phases (Fig. 1a), they showed how these phases of mare basalt
 194 eruption, together with total dike volumes, initial magma volatile content, vent
 195 configuration, and magma discharge rate, could assist in relating the wide range of
 196 seemingly disparate volcanic features to a common set of eruption processes. Figure 1
 197 updates the values given by Wilson and Head (2018) using a more detailed integration of
 198 the eruption rate model based on work in progress.

199 In Phase 1, which is very short-lived, the rising dike penetrates to the surface, and
 200 initiates *the transient gas release phase*. This very explosive phase is due to
 201 concentration of volatiles into a low-pressure area near the upper propagating dike tip
 202 (Wilson & Head, 2003). Pure gas may extend 100–200 m down from the top of the dike,
 203 above a high vesicularity foam layer extending downward ~10 km. Eruption of this gas-
 204 rich magma dike tip takes as little as a few minutes, resulting in an extremely thin but
 205 very widespread deposit, consistent with volcanic glass beads ubiquitous in lunar soils.

206 During Phase 2, *the high-flux hawaiian eruptive phase*, the dike continues to rise
 207 toward a neutral buoyancy configuration. This phase is characterized by the highest
 208 magma discharge rate during the eruption, $\sim 10^6 \text{ m}^3/\text{s}$, involving a near-steady explosive
 209 magma eruption; the volatile content is representative of the bulk of the magma. This

210 phase is characterized by formation of a relatively steady hawaiian fire fountain, largely
 211 optically dense. Submillimeter-sized pyroclastic droplets lose gas efficiently and
 212 accumulate with negligible cooling within a few to 10 km of the fissure, forming a lava
 213 lake deficient in gas bubbles. For a short-lived eruption, lava that is largely degassed
 214 flows away from the lake, initially turbulently, to form the distal part of the final lava
 215 flow deposit. For a sufficiently long-lasting eruption, the lava will feed a flow eroding a
 216 sinuous rille. More than 80% of the total dike magma volume would have been erupted
 217 during this phase; the erupted magma volume flux decreases from $\sim 10^6$ to $\sim 10^5 \text{ m}^3 \text{ s}^{-1}$
 218 over its typical 2-3 day duration.

219 Phase 3, the lower flux *hawaiian to strombolian transition phase* begins when the
 220 positive buoyancy of the lower part of the dike in the mantle balances the negative
 221 buoyancy of the upper part in the crust, and the eruption-feeding dike approaches an
 222 equilibrium. The lower dike tip stops rising, and fixing the vertical extent of the dike. The
 223 main driving process in this phase becomes the horizontal reduction in dike thickness due
 224 to: 1) decrease in internal excess pressure, and 2) relaxation of forced host rock
 225 deformation due to initial dike intrusion (Wilson & Head, 2017). Shallow crust host rock
 226 deformation is probably elastic and rapid; hotter mantle rock deformation (surrounding
 227 the lower part of the dike) is more likely to be visco-elastic or viscous; this results in a
 228 much longer closure timescale. Magma vertical rise speed in the dike decreases greatly
 229 (to less than 1 m/s) during this period; this implies that the magma volume flux leaving
 230 the vent decreases similarly to a few $\times 10^4 \text{ m}^3 \text{ s}^{-1}$ during ~ 3 -5 days. Reduction in vertical
 231 magma flow speed means that nucleating gas bubbles throughout the dike vertical extent
 232 can now rise through the liquid at an appreciable rate. There is ample time for larger
 233 bubbles to overtake smaller bubbles (especially CO bubbles being produced at great
 234 depths). This leads to coalescence and even greater growth; this in turn leads to very
 235 large bubbles—gas slugs—filling almost all of the dike width and producing strombolian
 236 surface explosions (Parfitt & Wilson, 1995). The transition from hawaiian activity (Phase
 237 2) to strombolian (Phase 3) occurs rapidly.

238 Phase 4, the *dike closing, strombolian vesicular flow phase*, begins when the
 239 activity becomes entirely strombolian. Horizontal dike closure continues due to tectonic
 240 stresses and magma is extruded at a low flux. Magma from the deepest dike parts
 241 continues to be forced upward to lower pressure levels thus continuing to produce some
 242 CO at all depths; the result is very minor, but continuing strombolian explosive activity
 243 above the vent. For magma still emerging from the vent, a stable crust will form and flow
 244 away as lava.

245 Two different pathways might occur during Phase 4 activity. In *low flux eruptions*
 246 (Phase 4a), Phase 4 might begin following of eruption of most of the magma in the dike
 247 and the volume flux has decreased to a very low level (Fig. 1a). Wilson and Head (2017)
 248 predict that the result will be the emplacement of vesicular lava in the vicinity of the vent
 249 as a series of cooling-limited flows potentially building a small, low shield. Erupted
 250 magma consists of liquid which contains bubbles (a mixture of gases and volatile
 251 elements) (Gaillard & Scaillet, 2014; Renggli et al., 2017; Saal et al., 2018). These are
 252 determined by the thermodynamic equilibrium between the products of interactions
 253 (mainly between H_2O and sulfur species) released over the last <500 m of magma flux.
 254 Such gas bubbles would nucleate with diameters of ~ 10 – $20 \mu\text{m}$ and grow to ~ 20 – $30 \mu\text{m}$
 255 at the surface; they remain stable within the lava (surface tension forces impose a

256 retaining pressure of ~30 kPa; Wilson & Head, 2017). Lavas exsolving ~1,000 ppm of
257 such gases would leave the vent as lava foams with vesicularities >90% by volume. The
258 topmost bubbles would likely have exploded into the overlying vacuum; this should
259 produce a layer of bubble wall shards, and gas escapes easily through this accumulating
260 debris layer until welding of particles and accumulated debris weight inhibited further
261 foam disintegration. If the underlying lava still contained dissolved volatiles, the
262 unvesiculated layer could become important during further lava cooling and
263 crystallization if volatile concentration into the remaining liquid resulted in second
264 boiling and additional post-emplacement vesiculation. Volatile contributions to the
265 atmosphere of these latter-stage processes would be minimal, however, as the rates of
266 diffusive volatile loss from vesiculated cooling and cooled lavas are extremely low.

267 A second potential outcome is predicted to occur in dikes that are vertically more
268 extensive (Phase 4b, high flux). If a large fraction of total dike magma remains available
269 for extrusion as vesicular lava, this lava can readily intrude into the still-hot interiors of
270 the previously emplaced nonvesicular flows and cause flow inflation. The shallow parts
271 (<400 m depth) of a dike feeding such intruding/inflating flows would contain not yet
272 exsolved water and sulfur compounds. The resulting inflated flows would cool on a
273 timescale of weeks: volatile concentration into the residual liquid as crystallization
274 occurred would then lead to second boiling. The new population of gas bubbles could
275 cause a possibly extensive further inflation episode (Wilson et al., 2019). The resulting
276 magmatic foam and gas could escape through cracks in the lava crust caused by inflation,
277 but again the gas flux into the atmosphere would be minimal.

278 Eruption Phase 4 duration is controlled by the global stress state of the lithosphere
279 (both its nature and magnitude), influencing host rocks visco-elastic relaxation, and by
280 magma cooling in the dike. Lunar thermal history (Solomon & Head, 1980) suggests
281 extensional lithospheric stresses during the first ~1 Ga, followed by compressive stresses
282 at ~3.6 Ga as the interior cooled. This would encourage more closure of dikes in
283 geologically more recent eruptions. Dike models (Wilson & Head, 2017) predict that
284 Phase 4 dikes had initial widths of at least 10–20 m. Cooling and solidification by
285 conduction alone of near-stagnant magma in such dikes would occur 1-2 years after the
286 end of an eruption.

287 In summary, the majority of the volume of magma erupted during a typical lunar
288 eruption occurs in Phase 2 and 3 (Fig. 1a). The rise speed of magma during these phases
289 is so large that gas bubbles stay locked to the magma, and so the vast majority of gas
290 release into the atmosphere during a lunar eruption also occurs during Phases 2 and 3,
291 phases that take place over about 5-10 days, less than about 25% of the total eruption
292 duration. We now turn to a discussion of the implications of the 1) total gas release
293 patterns and 2) gas release patterns in individual eruptions, for the formation of a
294 transient lunar atmosphere.

295

296 **3 Discussion**

297 The relevant parameters (lava volume, eruption rate, duration, total gas released,
298 gas mass release rate, etc.) for several types and scales of lunar eruptions (short flow,
299 long flow, sinuous rille, and Cobra Head/Schroeter's Valley, the largest known lunar
300 eruption) are shown in Table 1a. For each of the released gas masses we find the
301 properties of the lunar atmosphere that would be created if the gas release rate from the

erupted magma was much greater than the total loss rate of the atmosphere into space by whatever mechanisms were relevant (which we shall show shortly is the case). Using the mean molecular mass $m = \sim 31.4$ kg/kmol described above, we find the scale height of the resulting atmosphere, $H = (Q T) / (m g)$ where Q is the universal gas constant, 8.314 kJ kmol⁻¹ K⁻¹, T is the mean lunar surface temperature, ~ 270 K assuming radiative equilibrium and a 25% dimmer Sun ~ 3.5 Ga ago, and g is the acceleration due to gravity at the lunar surface, 1.62 m s⁻². These values give $H = 44.1$ km. The surface density of the atmosphere, ρ_s , is equal to its mass, M , from Table 1a, divided by the volume equivalent to the surface area of the Moon multiplied by the scale height, i.e. $\rho_s = M / (4 \pi R^2 H)$ where R is the lunar radius, 1738 km. Finally, the surface pressure is $P_s = \rho_s g H$. Table 1b lists the values of ρ_s and P_s corresponding to the eruption types in Table 1a. Assuming the most extreme alternative volatile species mixture suggested in the literature, the sulfur-dominated mixture of Renggli et al. (2017), would increase m by a factor close to 1.5. This would decrease the scale height and increase the surface density of the atmosphere by the same factor, and leave the surface pressure unchanged.

The implied atmospheric gas masses due to the typical types of lunar volcanic activity in Table 1a are of order 10^{12} to 10^{13} kg. As part of an extensive review of three possible types of lunar atmosphere, Stern (1999; his section 5.2.2) treated a hypothetical volcanically-induced atmosphere with a total gas mass of 10^{11} kg and adopted the loss rate calculated by Vondrak (1974) of 10 kg s⁻¹. The same loss rate is estimated in a recent more general analysis by Aleinov et al. (2019) treating much more massive, at least $\sim 10^{15}$ kg, atmospheres with surface pressures > 100 Pa. Using a 10 kg s⁻¹ loss rate leads to the typical timescales for atmospheric decay, τ_d , shown in Table 1b, between $\sim 2,000$ and $\sim 6,000$ years. These values need to be compared with the likely intervals between eruptions on the Moon. As shown earlier, with a total volume of volcanics of $V_t = \sim 10^7$ km³ (Head and Wilson, 1992; Evans, 2016), a typical erupted volume of 200 ± 100 km³ (Table 1a), and a total duration of volcanism of $\tau_d = \sim 2$ Ga, the shortest average interval between eruptions is $\sim 13,000$ to $40,000$ years in the early part of the mare volcanism era if eruptive activity decreases with time. Increasing the 2000 ppm magmatic volatile mass fraction used here to the 3400 ppm suggested by Rutherford et al. (2017) would increase the atmospheric mass values in Table 1a by a factor of 1.7, but this would still make the timescale for atmosphere loss a factor of ~ 4 less than the average time between eruptions.

What effect does the non-linear release of gas during the four phases of a typical volcanic eruption (Fig. 1a) have on the peak loss of volatiles during an eruption? To address this question, we first look at the magma volume eruption rate as a function of time for an eruption releasing 250 km³ of magma (a medium-scale volume in the ~ 100 - 300 km³ average eruption volume range described above) and lasting 46 days (about average for the 1-3 month range discussed above) (Fig. 1b). Magma volume flux is clearly highest in the first ten days (Phase 1 and 2), decreasing two orders of magnitude from an initial peak flux of 10^6 m³ s⁻¹, to 10^4 m³ s⁻¹ after ~ 10 days. Magma volume flux remains at this low value for the next 30 days (Phases 3-4) before falling to zero in the last 4 days at the end of the eruption. Thus, $\sim 90\%$ of the total volume of magma erupted is emplaced in Phase 2, the hawaiian phase characterized by maximum magma degassing and volatile loss.

Using the magma volatile species proposed by Rutherford et al. (2017), the percentages of the magma, water, and CO released as a function of time in the same

348 eruption are shown in Fig. 1c. Released water closely mimics the erupted magma, unless
349 a significant amount is left trapped in late-stage magma (Phase 4) intruded into earlier
350 flow lobes during flow inflation. If significant inflation occurs, and the inflating gas does
351 not escape, about 95% of the water would be released instead of 100% as shown in Fig.
352 1c. The CO in the magma, preferentially released at very great depth, does not all escape:
353 CO released in Phase 4 does not have time to reach the surface before the conduit freezes,
354 even allowing for bubble coalescence and rise. However, this only represents a few
355 percent of the total CO and so almost all of the total is released.

356 In summary, the implied intervals between typical lunar eruptions, ~13,000 to
357 40,000 years, are 6-7 times greater than the likely durations of the vast majority of
358 individual transient atmospheres, between ~2,000 and 6,000 years. Only for the single,
359 extreme example of Cobra Head/Schroeter's Valley are the time scales comparable.
360 Otherwise, only if all of the Moon's $\sim 10^7$ km³ of basaltic volcanism were to have taken
361 place within a 300 Ma interval would the time scales generally be comparable. The non-
362 linear release of gas during the four phases of a single eruption do not alter this
363 conclusion; even though volatile release is concentrated in the first 25-35% of the
364 eruption, the long repose periods between eruptions preclude sufficient buildup to create
365 an enduring atmosphere. The same is true of leakage of gas from magma reservoirs
366 between eruptions: if half of a typical magma volatile inventory is released uniformly
367 over the ~40 ka average interval between eruptions, the leakage rate is somewhat less
368 than 1 kg s⁻¹, an order of magnitude less than the atmospheric loss rate.

369

370 **4 Conclusions**

371 On the basis of our analysis of the generation, ascent and eruption of lunar mare
372 basalt magmas and forward-modeling individual eruptions, we conclude that it is very
373 unlikely that the Moon had a semi-permanent (as long as ~70 Ma) volcanically-driven
374 atmosphere as proposed by Needham and Kring (2017), even during a period of peak
375 volcanic flux in early lunar history. We attribute the differences between our estimates
376 and those of Needham and Kring (2017) (see discussion in Supporting Material) to their
377 use of maximum impact basin depths as average depths, and assignment of all excess
378 volumes below datable units to one age (e.g., 5.9×10^6 km³ assigned to 3.5 Ga in the case
379 of Imbrium).

380 We also conclude that these low volatile release volumes and rates are not
381 conducive to optimizing the transport of released volatiles from the eruption site to the
382 poles to enhance the accumulation of volatiles in polar cold traps (see also Aleinov et al.,
383 2019), nor of creating temporary environments that might favor astrobiological activity
384 (Schulze-Makuch and Crawford, 2018). Our results suggest that most volatiles in lunar
385 polar cold traps originated from volatile-rich impacts, rather than volatile release from
386 volcanic eruptions, similar to findings about polar cold-trap volatile deposits on Mercury
387 (e.g., Ernst et al., 2018; Deutsch et al., 2019, 2020). This issue could be clarified for the
388 Moon by in situ D/H ratio measurements. In order to refine our volcanic emission
389 estimates, future lunar exploration goals should include further analysis of detailed lava
390 flow thicknesses, ages, volumes, volatile contents and repose periods, as well as better
391 determination of the interior structure of mare deposits in large impact basins.

392

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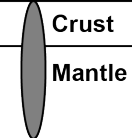
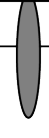
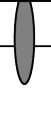
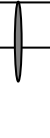
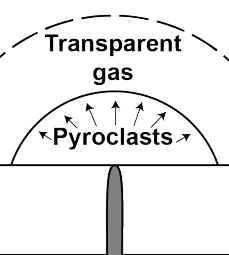
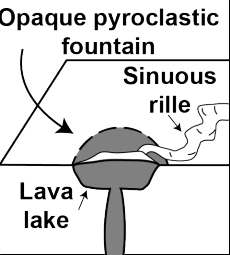
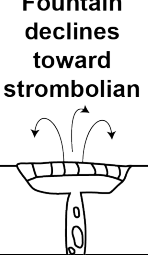
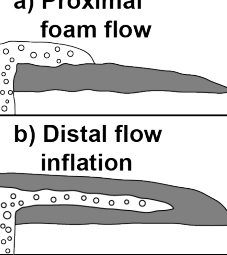
Table 1a. Parameters of various types of lunar eruption. Cobra Head is the source vent of Schroeter's Valley (Head and Wilson, 2017). Released volatiles assumed to have molecular mass $31.4 \text{ kg kmol}^{-1}$ and to form $n = 2000$ ppm by mass of a magma that has a liquid density $\rho_m = 3000 \text{ kg m}^{-3}$. V = lava volume; F_l = lava volume eruption rate; τ_e = eruption duration; M_g = total gas mass released; F_g = gas mass release rate. Typical values for parameters are quoted but individual eruption values may vary by a factor of at least 2 to 3.

Feature	V/km^3	$F_l/(\text{m}^3 \text{ s}^{-1})$	τ_e/days	M_g/kg	$F_g/(\text{kg s}^{-1})$
Cobra Head	2000	1.4×10^5	150	1.2×10^{13}	9.3×10^5
Long flow	300	$\sim 10^5$	30	1.8×10^{12}	6.9×10^5
Small flow	200	$\sim 10^4$	100	1.2×10^{12}	1.4×10^5
Sinuuous rille	100	$\sim 3 \times 10^4$	50	$\sim 6 \times 10^{11}$	1.4×10^5

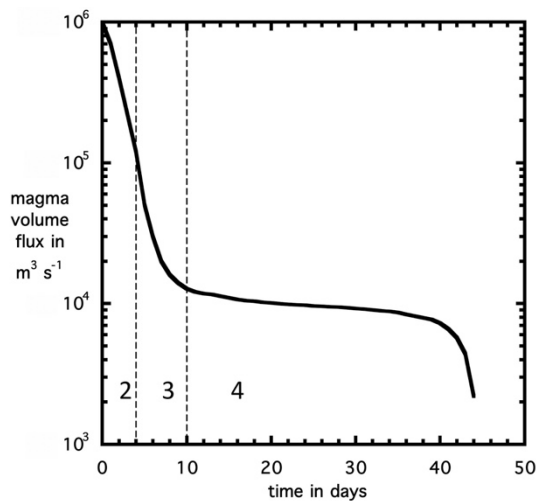
Table 1b. Initial values of the surface density, ρ_s , and surface pressure, P_s , in a transient atmosphere produced by the four types of volcanic activity listed in Table 1a. The maximum duration of the atmosphere, τ_d , is indicated.

Feature	$\rho_s/(\text{kg m}^{-3})$	P_s/Pa	τ_d/years
Cobra Head	7.2×10^{-6}	0.51	38,000
Long flow	1.1×10^{-6}	7.7×10^{-2}	5,700
Small flow	7.2×10^{-7}	5.1×10^{-2}	3,800
Sinuuous rille	3.6×10^{-7}	2.6×10^{-2}	1,900

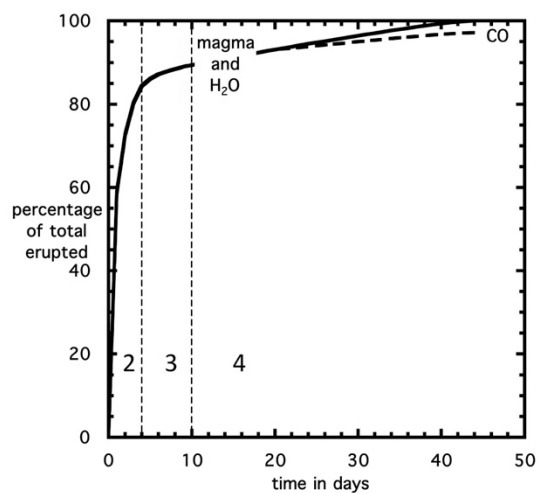
Figure 1a-c.

Eruption Phase	PHASE 1	PHASE 2	PHASE 3	PHASE 4
	Dike penetrates to surface, transient gas release phase	Dike base still rising, high flux hawaiian eruptive phase	Dike equilibration, lower flux hawaiian to strombolian transition phase	Dike closing, strombolian vesicular flow phase
Dike Configuration				
Surface Eruption Style				
Magma Rise Speed	30 to 20 m/s	20 to 10 m/s	5 to <1 m/s	< 1 m/s
Magma Volume Flux	$\sim 10^6$ m ³ /s	10^6 to 10^5 m ³ /s	10^5 to $\sim 10^4$ m ³ /s	$\sim 10^4$ m ³ /s
Percent Dike Volume Erupted	<5%	$\sim 80\%$	$\sim 5\%$	$\sim 10\%$
Phase Duration	~ 3 minutes	~ 4 day	~ 6 day	~ 30 days

1a



1b



1c

Supporting Material for

“Volcanically-Induced Transient Atmospheres on the Moon: Assessment of Duration, Significance and Contributions to Polar Volatile Traps”

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Analysis and Assessment of Procedures and Assumptions:

In this supplementary material, we provide commentary on the major sources of uncertainty in assessing the duration and significance of volcanically-induced transient atmospheres and major sources of uncertainty in the approach, as a guide for comparison to our forward-model gas exsolution estimates in the main text. We summarize the procedures and assumptions used by Needham and Kring (2017) (N&K) to determine the mare basalt volcanic flux that, together with their volatile exsolution model, is the input into their lunar atmospheric buildup and retention calculations (Fig. S1). We also discuss how future work could help improve these estimates.

Major steps in establishing the atmospheric buildup and retention include 1) estimating the mare basalt volume, (2) determining the mare basalt volcanic flux (volume as a function of time), and (3) estimating the production of lunar volatiles over time.

S1. Estimating the mare basalt volume in each basin

In order to estimate the total volume of basalt erupted with time, N&K first tabulate the volumes of basalts erupted into each of the major impact basins (e.g., Imbrium, Serenitatis, etc.) or areas of accumulation (e.g., Oceanus Procellarum) (N&K, their Table 1). There are various levels of uncertainty pertaining to basin size (assignment of rings in multi-ring basins), the initial and final basin geometry, and the state of degradation and response to the thermal structure of the Moon at the time of basin formation that influence estimates of basalt volume in any particular basin. As discussed by N&K, there is also a high level of uncertainty in past estimates of the total thickness and volumes of mare basalts in individual basins using different techniques (e.g., Crisium basin estimates differ by $5 \times 10^5 \text{ km}^3$).

For most of the major mare basins (in order of decreasing total volume: Imbrium, Serenitatis, Crisium, Humorum, Nectaris, Grimaldi, Smythii), N&K use the *basin depth-diameter estimates* of Williams and Zuber (1998) as input into their maximum thickness estimates for their volume calculations; they describe these as generally consistent with the more recent data from LOLA and GRAIL. N&K augment this with older data (Horz, 1978: Procellarum, Tranquillitatis; Yingst and Head, 1997: South Pole-Aitken) and a more recent study for Orientale (Whitten et al., 2011). These data then represent the

45 values for the total mare basalt fill thickness in each of eleven lunar basins (N&K, their
46 Table 1) (Fig. S2a).

47 One source of uncertainty with this approach is that the Williams and Zuber (1998)
48 estimates for impact basin geometry and lava fill thicknesses were compiled with low
49 resolution Clementine altimetry data. In addition, N&K make several assumptions about
50 the initial structure of the basins, and together, these factors tend to very significantly
51 overestimate the total basin volumes in several key cases (e.g., Imbrium, Serenitatis).
52 The Williams and Zuber (1998) maximum thickness estimates (N&K, their Table 1,
53 column 3) are then used by N&K as *average thicknesses* to compute the entire mare
54 basalt volume in each of the 11 basins treated by Williams and Zuber (1998) (N&K, their
55 Table 1, column 4). These 11 total volumes (N&K, their Table 1, column 4) then account
56 for 44% of the lunar separate mare accumulation *areas* considered (11 of 25), but >95%
57 of the total global mare basalt *volume* calculated by N&K (Fig. S2b).

58 More specifically, the Williams and Zuber (1998) approach to estimating maximum
59 basalt thickness in each basin is as follows: Clementine LIDAR data are used to calculate
60 the depths of 29 large craters and basins on the Moon; the most well-preserved examples,
61 generally unflooded by mare basalt volcanism, are then compared with previous
62 depth/diameter (d/D) plots, revealing an inflection point in the diameter range
63 characterized by the transition from complex craters to peak-ring basins.

64 From this relationship, an empirical power law fit is derived for basin depth as a
65 function of increasing size, a relationship characterized by a shallower slope than that of
66 complex craters. The definition of this inflection point is provided by seven peak-ring
67 basins ranging in diameter from 100-200 km. Williams and Zuber (1998) then use this
68 empirical power law basin-scale d/D relationship to estimate the thickness of mare basalt
69 fill in each basin by assuming that the unfilled depths of each basin follow this d/D
70 relationship, and the current mare fill elevation represents the upper bound on the
71 thickness. Thus, the Williams and Zuber (1998) estimated basalt thicknesses for each
72 basin are derived from “the difference between the predicted basin depth and the
73 measured depth of the mare surface.” N&K then use these Williams and Zuber (1998)
74 thickness estimates (N&K, their Table 1, column 3) as input to their basin volume
75 calculations.

76 Among the sources of uncertainty in the thickness estimates derived by Williams and
77 Zuber (1998) are:

- 78 1. Loading and flexure of the basin interior can cause basin subsidence (e.g.,
79 Solomon and Head, 1979, 1980), resulting in mare basalt thickness in the basin
80 interior being greater than that predicted by the d/D relationship. Williams and
81 Zuber (1998) accounted for this loading-related subsidence in their calculations
82 (their Table 2), and thus this is accounted for in the thicknesses N&K utilize in
83 their Table 1.
- 84 2. Not all of the basins chosen by N&K to apply the Williams and Zuber (1998)
85 thickness estimates to derive volumes (the 11 basins in their Table 1) were
86 comparable to the most well-preserved and generally unmodified basins used to
87 calculate the empirical power-law fit. A large number of factors can influence
88 basin topography during and subsequent to basin formation: initial lithospheric
89 thermal structure, subsequent viscous relaxation, and impact-induced
90 topographic degradation are all likely to mean that older basins (for example,

91 old Tranquillitatis compared with young Imbrium) were shallower than the
 92 empirical fit when they began to be filled with mare basalt, resulting in
 93 overestimation of the thickness of their fill. Impact degradation can lower rim
 94 elevation, emplace ejecta inside the basin, and shallow the floor, all decreasing
 95 the basin depth prior to filling with mare basalt. For example, the impact event
 96 forming the Imbrium basin emplaced significant thicknesses and volumes of
 97 ejecta inside the Serenitatis basin (Spudis et al., 2011), volumes that would need
 98 to be subtracted from the total thickness, and thus volume, of Serenitatis mare
 99 fill.

- 100 3. Scaling depth/diameter relationships to larger basin-scale diameters implies a
 101 consistent choice for the crater and basin diameter. Unfortunately, the choice of
 102 which ring in multi-ring basins represents the closest approximation to the crater
 103 or peak-ring basin rim crest is controversial (see Head, 1974; Howard et al.,
 104 1974; Spudis, 1993; Neumann et al., 2015; Zuber et al, 2016; Johnson et al.,
 105 2016, for discussion of this problem) and thus the choice of basin diameter to
 106 input into the extrapolation of depth/diameter relationships to larger basin-scale
 107 diameters could vary by several tens of percent.
- 108 4. Depth/diameter relationships provide data on the *maximum* depth of the basin,
 109 and thus the *maximum* thickness of the fill, not the *average* thickness. For
 110 example, the Orientale basin has a maximum depth of >8 km but the majority of
 111 the basin is less than ~2 km deep (Fig. S3a). The geometry of the interior of
 112 multi-ringed basins differs significantly from a cylindrical plug (Fig. S3b). For
 113 example, the thickness of mare basalts may differ by a factor of four between
 114 the deepest part of the basin interior and the outer area overlying the terrace
 115 inside the topographic ring. Volcanic flooding models of the essentially
 116 unfilled Orientale basin (Head, 1982) showed that the area between the Inner
 117 Rook ring and the Cordillera ring, an area making up more than 70% of the total
 118 area of the basin, had lava thicknesses of less than 2 km, a thickness about 25%
 119 of the deepest part of the basin.
- 120 5. There are uncertainties in extrapolating d/D data from the diameter range of
 121 peak-ring basins to the much larger scale of multi-ringed basins. Using much
 122 improved d/D data from the Lunar Orbiter Laser Altimeter (Smith et al., 2010)
 123 and GRAIL gravity data (Zuber et al., 2013), Baker and Head (2013) and Baker
 124 et al. (2012) showed that there is a possible further shallowing of the slope of
 125 the d/D trend at the multi-ringed basin scale, the diameter of most of the 11
 126 basins considered by Williams and Zuber (1998) and N&K. This predicted
 127 shallowing would result in the thicknesses calculated by the Williams and Zuber
 128 (1998) method (N&K, their Table 1, column 3) being overestimates.

129 *Summary of the N&K tabulation of mare basalt volumes in the 11 major mare basins:* In
 130 order to derive the total volume of mare basalt in each basin, N&K use the *maximum*
 131 *thickness* estimates of William and Zuber (1998) (N&K, their Table 1, column 3). They
 132 assume that this is the *average thickness* (Fig. S3b), and multiply this maximum
 133 thickness by the area inside the basin to derive a *total volume of mare basalt in each*
 134 *basin* (N&K, their Table 1, column 4). For example, in the Imbrium basin, N&K use the
 135 thickness of mare basalts determined by Williams and Zuber (1998) derived as follows
 136 (Fig. S3b: Imbrium basin extrapolated d/D, minus depth to current mare surface, yields a

137 thickness of 4.70 km; adjusting this for loading and subsidence yields a *maximum*
 138 thickness of 5.24 km for the lava at the center of the Imbrium basin. N&K then take this
 139 *maximum* thickness, which includes all of the caveats described above predicting that the
 140 sign of this value will be an overestimate, and then take two additional steps: 1) they
 141 calculate the area of the Imbrium basin to cover 1,010,400 km², and then assume that the
 142 *maximum* thickness estimate of 5.24 km is the *average* thickness over the entire
 143 1,010,400 km² area of the basin, yielding a *total volume* of the Imbrium basin mare fill to
 144 be 5.295 x 10⁶ km³ (Fig. S3b). On the basis of using a *maximum* thickness estimate
 145 (very likely to be an overestimate for the reasons stated above) and assuming that it
 146 represents the *average* thickness for the whole basin, we believe that this value
 147 *significantly overestimates* the volume of mare basalt in the Imbrium basin. A similar
 148 approach is utilized for each individual basin (N&K Table 1) suggesting that these values
 149 will also be overestimates.

150

151 ***S1.1 Opportunities and prospects for current and future improved estimates***

152 The major uncertainties in the total volume estimates for each basin derive from lack
 153 of detailed knowledge of the underlying geometry of impact basins at the time of their
 154 initial mare fill, and their detailed response to topographic filling (loading, flexure and
 155 subsidence). New Lunar Orbiter Laser Altimeter (LOLA) and Gravity Recovery and
 156 Interior Laboratory (GRAIL) data are the types of data that can help reduce this
 157 uncertainty. The acquisition of much higher resolution altimetry data (Lunar Orbiter
 158 Laser Altimeter; Smith et al., 2010) and gravity data (GRAIL; Zuber et al., 2013)
 159 permitted better understanding of the topography of craters, peak-ring and multi-ring
 160 basins (e.g., Baker and Head, 2013, 2015) and an understanding of the three-dimensional
 161 structure and lava filling histories of impact craters and basins. For example, Whitten
 162 and Head (2013) provided detailed modeling of lava flooding and progressive filling
 163 estimates for typical peak-ring basins and degraded multi-ring basins. Evans et al. (2016)
 164 used GRAIL gravity data to assess the presence of craters buried by lava filling and the
 165 thickness and volume of their fill. LOLA and GRAIL data have also been used to
 166 estimate the average thicknesses of mare basalts on the lunar nearside (0.74 km) (Gong et
 167 al., 2016). For the future, there is a compelling need for detailed basin-wide geophysical
 168 traverse surveys to assess the depth to the mare-basin floor interface, the depth and
 169 geometry of the crust-mantle interface, and variations in basin geometry and fill as a
 170 function of initial basin age.

171

172 **S2. Determining the mare basalt volcanic flux (volume of lava extruded as a** 173 **function of time)**

174 To convert total estimated mare basalt volumes derived for each basin (as described
 175 above) into a volcanic flux (volume as a function of time), the ages of the various mare
 176 basalt units need to be determined and their relative abundance assessed. Six steps are
 177 needed to accomplish this task: 1) definition of a volcanic unit, and then determination of
 178 the 2) area covered by the unit, 3) thickness of the unit, 4) volume of the unit, 5) age of
 179 the unit, and finally, 6) duration of its emplacement. Obviously, deriving each of the six
 180 factors in these estimates becomes more and more difficult for older and older deposits,
 181 as the stratigraphically younger flow units mask the older flow units (Fig. S4). Needham
 182 and Kring (2017) use the following set of steps to accomplish this task:

183 ***S2.1. Estimate mare unit area and age***

184 N&K use the mare unit boundary mapping and crater size frequency analyses data
185 from Hiesinger et al. (2011) (and others). On the basis of these data they conclude that
186 mare basalt provinces were emplaced from ~3.9 Ga to as recently as ~1.1 Ga.

187 ***S2.2. Estimate mare unit thickness***

188 N&K note that “observations of specific mare eruptive units indicate an average mare
189 unit thickness of ~250 m (Weider et al., 2010) within Serenitatis and Oceanus
190 Procellarum...” with this thickness “...expected to incorporate an integrated sequence of
191 thinner flows....and is assumed to be the average thickness for all surface mare units in
192 the absence of other thickness measurements...” In the analysis of Weider et al. (2010),
193 they identify eight units (their Table 4) with estimated average unit thicknesses ranging
194 from 80-600 m, and derive an average of 250 m derived by averaging the total of the
195 individual values in their Table 4.

196 As N&K acknowledge, spectrally defined flow units can easily be composed of
197 multiple lava flows of similar composition. In addition, the thickness of a flow unit is
198 related to the nature of the underlying topography: flows emplaced in rough terrain such
199 as the interior of craters or the highlands will pond and be much thicker than flows
200 emplaced on a flat or sloping mare plain (e.g., Head, 1982; Whitten and Head, 2013). A
201 wide range of mare basalt flow and unit thickness have been observed or inferred in the
202 relatively flat lunar maria, as follows: 1) 3-5 m from near-terminator images of flow
203 fronts (Head and Lloyd, 1971; Lloyd and Head, 1972); 2) average of 30-35 m in the flow
204 fronts in the observed young Eratosthenian-aged flow fronts in Mare Imbrium (Schaber,
205 1973); 3) 30-60 m average thickness (range of 20-220 m) from basalt flow units exposed
206 within the nearside maria, using inflections in impact crater size-frequency distributions
207 (Hiesinger et al., 2002); 4) ~10 m average flow thicknesses estimated from exposed
208 sections in impact crater and pit crater walls (Robinson et al., 2012).

209 On the basis of these estimates, all typically less than ~50 m, we conclude that the
210 average thickness of 250 m may overestimate the average thickness of individual lava
211 flows by a factor of five. Clearly, average thickness in the initial rough topography of
212 the basin floor may have been larger, but as shown by the interior of Orientale (e.g.,
213 Head, 1982; Whitten et al., 2011), these variations will soon tend to smooth out due to
214 emplacement of superposed lavas. An additional factor is that a spectrally defined lava
215 flow unit may be composed of a series of individual lava flows. The values determined
216 by Weider et al. (2010) are for spectrally defined flow units, and thus these are likely to
217 be composed of a series of flow units whose thickness was cumulative. Using this
218 average 250 m flow thickness to estimate the thickness of all previous dated flow units
219 carries with it the interpretation that these previously dated flow units are likely to be
220 composed of multiple flow units of uncertain age range. This is acknowledged by N&K,
221 but needs to be kept in mind as one moves forward to the determination of volatile flux in
222 individual eruptions and the contribution of such individual eruptive events to production
223 and retention of a transient atmosphere. For example, if a dated 250 m thick lava flow
224 unit consists of five separate 50 m thick eruptive lava flows, what are the thicknesses,
225 volumes and volatile fluxes of each of the individual eruptions, and is the repose period
226 between their emplacement sufficient to cumulatively contribute to the buildup of a
227 transient atmosphere, or do the volatiles dissipate between eruptions?
228

229 **S2.3 Estimate mare basalt erupted volume as a function of time**

230 Step 1: Surface Flows: Needham and Kring (2017) then used the individual mapped
 231 mare basalt units and ages (predominantly from Hiesinger et al., 2011) to calculate the
 232 volume of surface mare basalts emplaced in each basin as a function of time (their Fig.
 233 2a; reproduced as Fig. S1a). These mapped and dated mare units are exposed at the
 234 surface, and thus overlie older units for which only relative ages are available (they are
 235 older because they underlie the basalts exposed at the surface) (Fig. S4).

236 Step 2: Lava Flows Underlying Exposed Surface Flows: Needham and Kring then
 237 “assume that the underlying flows were emplaced as older surface flows that were
 238 embayed by younger surface flows, such that the mare units are stacks of superposed lava
 239 units emplaced via effusive surface eruptions. Although ages of underlying basalts, with
 240 volumes taken as the difference between the total mare basalt for a given basin and the
 241 volume of the mapped surface flows, are not identified directly, these deposits are at least
 242 as old as the oldest surface unit (noted in italics in their Table S1).” (Fig. S4). Without
 243 age constraints on the underlying units, it is not possible to accurately describe the timing
 244 of the older eruptions. N&K provide a maximum estimate by assuming that all
 245 underlying units erupted at the same time as the surface units.

246 These underlying units are then dealt with by Needham and Kring in two ways as
 247 shown in the following example for the Imbrium basin:

- 248 1. The oldest dated mare unit in the Imbrium basin is listed as 3.55 Ga (their Table
 249 S1). For the basin maria “with unreported ages of units or of units underlying
 250 surface mare with identified ages” (their Table S1 explanation, italicized entries
 251 as listed in Table S1), N&K assign an age to these *undated and underlying units*
 252 that is equal to the age of the oldest flow (3.55 Ga). For Imbrium, this gives 17
 253 units that are 3.55 Ga, the age of the oldest dated flow, adding an additional
 254 25,383 km³ of lava emplaced at this age (3.55 Ga). This provides a total volume
 255 of “dated flows” (35 basalt units ranging in age from 1.1 to 3.5 Ga with a
 256 volume of 221,217 km³, 4.3% of the total Imbrium basin mare fill of 5,294,497
 257 km³), plus unreported and underlying surface units (17 units, all assigned the
 258 age of the oldest dated unit, 3.55 Ga with a volume of 25,383 km³, 0.5% of the
 259 total Imbrium basin mare fill) (summarized as percentages in parentheses in Fig.
 260 S4). An exact definition of the “underlying units” is not provided, but it
 261 consists of these 17 units (their Table S1, units with ages in italics).
- 262 2. In order to account for the rest of the mare basalt basin fill that lies below the
 263 dated flows, the undated flows together with the “underlying units”, N&K take
 264 the total volume of these flow units (35 + 17 = 52 units in the case of the
 265 Imbrium basin) and subtract this number (252,600 km³) from their total volume
 266 of the Imbrium basin derived from using the Williams and Zuber (1998)-based
 267 thickness estimate (N&K, their Table 1, column 3) to derive a *total volume*
 268 *estimate* (5,294,497 km³; N&K, their Table 1, column 4), thus identifying
 269 5,041,900 km³ of additional basin fill (95.2% of the total Imbrium basin
 270 volume) as “excess volume”. They then assign all of this remaining “excess
 271 volume” (95.2% of the total Imbrium basin fill) to an age of the oldest dated
 272 surface flow (3.55 Ga), resulting in 95.2% of the total Imbrium basin fill having
 273 a single age (Fig. S4).

274 *Summary:* In summary, N&K take the total Imbrium basin volume from the Williams

275 and Zuber (1998) maximum basalt thickness estimate, assume that it represents the
 276 average mare fill thickness in the basin (Fig. S3b), and then subtract the volumes that
 277 they have accounted for so far with “dated surface”, “undated surface” and “subsurface”
 278 flow units (4.8% of the total Imbrium basin volume), and subtract this total number from
 279 the “grand total” implied by adopting the Williams and Zuber number maximum
 280 thickness number as the average thickness number. N&K then assign *all of this*
 281 remaining “excess volume” (95.2% of the total volume) to an age of emplacement of the
 282 *oldest dated surface flow*, 3.55 Ga (Fig. S2c, S4). This 95.2% of their estimated total
 283 basin volume accounts for virtually all of the $\sim 5.4 \times 10^6 \text{ km}^3$ peak in global mare basalt
 284 flux at ~ 3.55 Ga shown in N&K Figure 2a (Figure S1a).

285 N&K treat the second largest basin fill volume (the Serenitatis basin) in a similar
 286 manner (Fig. S2d). The total Serenitatis basin volume is derived by N&K using the
 287 maximum thickness from Williams and Zuber (1998) as an average thickness (Fig. S3b)
 288 (N&K, their Table 1, column 3), multiplying by the total basin area (N&K, their Table 1
 289 column 2), to obtain a total basin mare basalt volcanic fill volume of $1,473,679 \text{ km}^3$
 290 (N&K, their Table 1, column 4). They define 23 dated basin units, ranging in age from
 291 2.44-3.81 Ga, that make up a volume of $62,262 \text{ km}^3$, 4.2% of the total. They further
 292 identify 12 additional units comprised of “undated surface” and “subsurface” flow units,
 293 that make up a volume of $23,417 \text{ km}^3$, 1.6% of the total Serenitatis basin, and add this to
 294 the 23 dated flows, for a total of $85,679 \text{ km}^3$, a volume making up 5.8% of the total
 295 Serenitatis basin volume estimated by N&K (column 4, their Table 1). Finally, they
 296 subtract this subtotal from the volume “grand total” derived from using the Williams and
 297 Zuber maximum thickness number as an average thickness ($1,473,679 \text{ km}^3$; their Table 1
 298 column 4), yielding an ‘excess volume’ of $1,388,000 \text{ km}^3$, 94.2% of the total volume in
 299 the Serenitatis basin. This entire “excess volume” is then assigned the age of the oldest
 300 dated flow, 3.81 Ga, resulting in $\sim 94\%$ of the total volume in the Serenitatis basin (N&K,
 301 their Table 1, column 4) being assigned to a single emplacement age (3.81 Ga) (Fig.
 302 S2d). This 94.2% accounts for virtually all of the $\sim 1.5 \times 10^6 \text{ km}^3$ peak in global mare
 303 basalt flux at ~ 3.8 Ga shown in Needham and Kring Figure 2a (Figure S1a).

304 *Implications:* The assignment of this huge “excess volume” in individual basins
 305 (95.2% of the total Imbrium basin mare basalt volume; 94.2% of the total Serenitatis
 306 basin mare basalt volume) to one specific age (Imbrium = 3.55 Ga; Serenitatis = 3.81 Ga)
 307 then forms almost the entire $5.5 \times 10^6 \text{ km}^3$ global peak in the mare basalt flux at 3.5 Ga
 308 seen in their Figure 2a and the second peak at ~ 3.8 Ga in their Figure 2a (Fig. S1a).

309

310 ***S2.4 Opportunities and prospects for current and future improved estimates of mare*** 311 ***basalt age assignments and total volumes (flux)***

312 What are the alternative approaches to the N&K assignment of the huge “excess
 313 volume” to the age of the single oldest dated flow? Instead of assigning 95.2% of the
 314 undated volume in Imbrium to one age one could spread this volume evenly out over the
 315 entire time between the approximate formation of the Imbrium basin (about 3.85 Ga; see
 316 discussion in Stoffler et al., 2006), and the 3.55 age (oldest dated surface flow), reducing
 317 the peak down to $< 2 \times 10^6 \text{ km}^3$, spread out over 250-300 Ma (compare this to their Fig.
 318 2a; Fig. S1a here). In another approach, Thompson et al. (2009) used superposed craters
 319 penetrating through the mare basalt to derive a total volume for the Imbrium basin mare
 320 basalt fill of $1.3 \times 10^6 \text{ km}^3$. The Thompson et al. (2009) value is only $\sim 25\%$ of the

321 Needham and Kring total volume value ($5.295 \times 10^6 \text{ km}^3$). Future exploration to address
 322 this uncertainty should involve a sample return mission to the ejecta of crater penetrating
 323 the entire mare fill, and regional geophysical surveys to establish subsurface stratigraphy
 324 and structure.

325

326 **S3. Production of Lunar Volatiles Over Time**

327 N&K now take the total “Volume of erupted basalts as a function of time, indicating
 328 peak volcanic activity primarily in the Imbrium basin ca. 3.5 Ga.” (their Fig. 2a; Fig. S1a
 329 here) and calculate the “Mass of volatiles, primarily CO and S, degassed during mare
 330 emplacement...” (their Fig. 2b; our Fig S1b here). In order to convert the mare basalt
 331 fluxes, as discussed above, to a production function for volatiles released over time, N&K
 332 first estimate the proportion of volatiles degassed during mare emplacement (their Table
 333 2), using maximum and minimum values reported in the literature for five species (CO,
 334 H₂O, H₂, OH and S). They then estimate the percent of each gas species liberated from a
 335 unit mass (CO, 100%; H₂O, 90%; H₂, 100%; OH, 99%; S, 90%), and then convert this to
 336 maximum and minimum degassed masses of gas for each species in ppm (their Table 2).
 337 The “mass of erupted lava was then calculated by multiplying the estimated volume by
 338 the bulk density of typical mare basalt ($\sim 3.00 \text{ g/cm}^3$),” and this mass was then multiplied
 339 by the minimum and maximum contents of each mare basalt volatile species as listed in
 340 their Table 2. This approach enabled the determination of “the mass range of each
 341 volatile released during an eruption”.

342 The next step taken by Needham and Kring was to assess “Incremental production...
 343 calculated for mare volumes erupted every 0.1 Ga.” Their Table S3 shows that a grand
 344 summed total of $8,900,775 \text{ km}^3$ of mare lava was erupted on the Moon; 61.2% of this
 345 total ($5,446,355 \text{ km}^3$) was erupted at the 100 Ma interval centered at 3.5 Ga, and of this
 346 61.2%, 93.5% was erupted in the Imbrium basin. Finally, 99.5% of this total erupted
 347 volume was “undated” and “excess” lava, underlying the four oldest dated flow units in
 348 Imbrium (3.5-3.55 Ga, $24,031 \text{ km}^3$) (Fig. S2c): on the basis of Needham and Kring
 349 approach outlined above, all of this 99.5% was assigned the age of the oldest flow, 3.5
 350 Ga.

351 Their Table S3 also shows that, of the grand summed total of $8,900,775 \text{ km}^3$ of mare
 352 lava erupted on the Moon, 17% of this total ($1,525,044 \text{ km}^3$) was erupted in the 100 Ma
 353 interval centered at 3.8 Ga, and of this 17%, 92.6% was erupted in the Serenitatis basin.
 354 Finally, 99.5% of this total volume erupted in Serenitatis was “undated” and “excess”
 355 lava, underlying the oldest dated flow unit in Serenitatis (3.85 Ga, 621 km^3) (Fig. S2d):
 356 on the basis of N&K approach outlined above, all of this 99.5% was assigned the age of
 357 the oldest flow, 3.85 Ga.

358 Needham and Kring then use these data in order to derive the “Mass and surface
 359 pressure of volatiles degassed by lunar mare basalt as a function of time“ (their Table
 360 S3). The results are plotted in their Figure 2c (Fig. S1c here), the “Atmospheric surface
 361 pressure resulting from the volatiles released during mare emplacement, with a peak
 362 pressure $\sim 1\%$ of Earth’s current atmospheric pressure corresponding to peak volcanic
 363 activity 3.5 Ga.”

364 *Summary:* On the basis of this assessment and analysis, the Needham and Kring
 365 approach of assigning the “underlying”, “undated” and “excess” mare lavas to a single
 366 age (the age of the oldest flow) (Fig. S4), and the reservations outlined above concerning

367 the use of the “maximum” basin depth from Williams and Zuber (1998) as an “average
 368 depth” for the mare fill, appear to create unrealistically large volumes focused at single
 369 time intervals (3.5 Ga for Imbrium and 3.8 for Serenitatis), that tend to produce and
 370 significantly overestimate the peak flux at these times (their Figure 2b; Fig. S1b here). In
 371 a final step, the resulting production functions were then plotted (their Figure 2b; Fig.
 372 S1b here) and were summarized in their Table S3. Needham and Kring conclude that
 373 these data clearly show that peak volatile releases occurred at 3.8 Ga and 3.5 Ga.

374 Among the uncertainties in this approach, in addition to the total volumes and age
 375 assumptions for individual basins discussed above, are:

376 1) Eruption time period (duration): Volcanic unit ages, volumes, and gas release
 377 masses are binned in 100 Ma intervals (N&K, their Table S3). This binning effectively
 378 serves to reduce any individual eruptive peaks to an average of the 100 Ma period. If the
 379 volumes of units and their individual ages were known with great accuracy, an individual
 380 peak could potentially greatly exceed the average, and its contribution to a lunar
 381 atmosphere could be underestimated.

382 2) Time-dependent volatile input during a single eruption: Gas release during
 383 volcanic eruptions is typically non-linear, decreasing as a function of time during the
 384 eruption. Depending on the eruption duration, this could have a significant effect on the
 385 volumetric contributions of volatiles to an atmosphere and its dissipation history.
 386 Similarly, individual gas species vent at different rates and times during eruptions,
 387 important considerations in potential buildup and retention in an atmosphere. We treat
 388 these factors in more detail in the main contribution.

389 3) Eruption repose period (time between eruptions): Similarly, summing the entire
 390 volume and flux for a 100 Ma period implies that this input was continuous into the
 391 atmosphere for a 100 Ma period. If the repose period was significant, the atmospheric
 392 contribution from a single event may completely dissipate, rather than contribute to the
 393 buildup from a longer term average input. The τ_d values in our Table 1b in the main text
 394 show that (ignoring the one-off Cobra Head event), two eruptions would have to occur
 395 within ~4000 years of one another for there to be a significant effect on prolonging a
 396 temporary atmosphere. Approximating the 20,000 to 60,000 year typical interval by
 397 normal distribution with mean 40,000 and standard deviation 20,000, a 4000 year interval
 398 would have a probability of ~3%.

399

400 ***S3.1 Opportunities and prospects for current and future improved estimates of mare*** 401 ***basalt volatile contributions to an atmosphere***

402 Clearly, improved stratigraphic relationships of dated lava flows in the most
 403 volumetrically significant lunar basins (e.g., Imbrium, Serenitatis) would be essential to
 404 decreasing the uncertainty in the Needham and Kring (2017) estimates, as would more
 405 precise determinations of the absolute ages of individual lunar basins. Also critically
 406 important is the initial volatile content of mare basalt magmas generated at depth and
 407 their global variability, as well as volatile release (e.g., Rutherford et al., 2017) and loss
 408 processes as a function of individual eruptions (e.g., Wilson and Head, 2018).
 409 Information on volumes of individual eruptions, their duration, and the chronology of
 410 volatile speciation and loss would be essential for the reliable determination of loss rates
 411 and contributions to a candidate lunar atmosphere.

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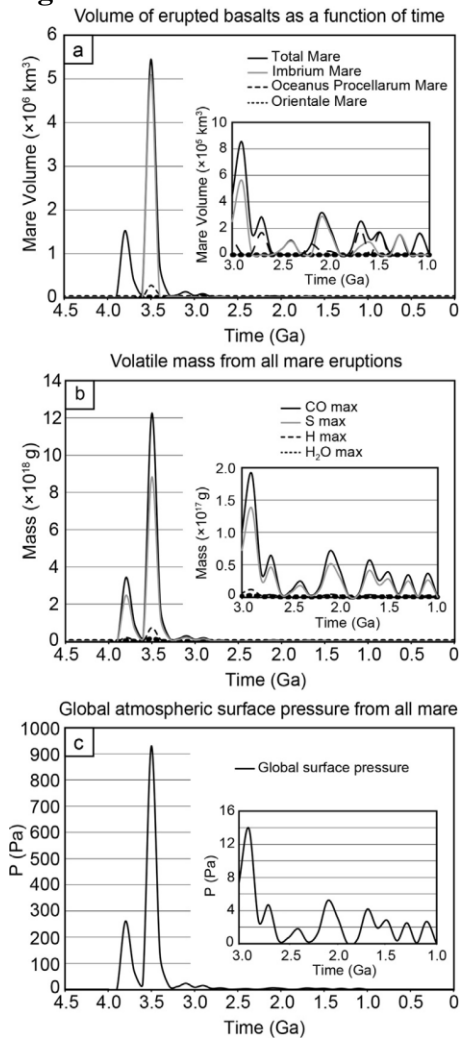
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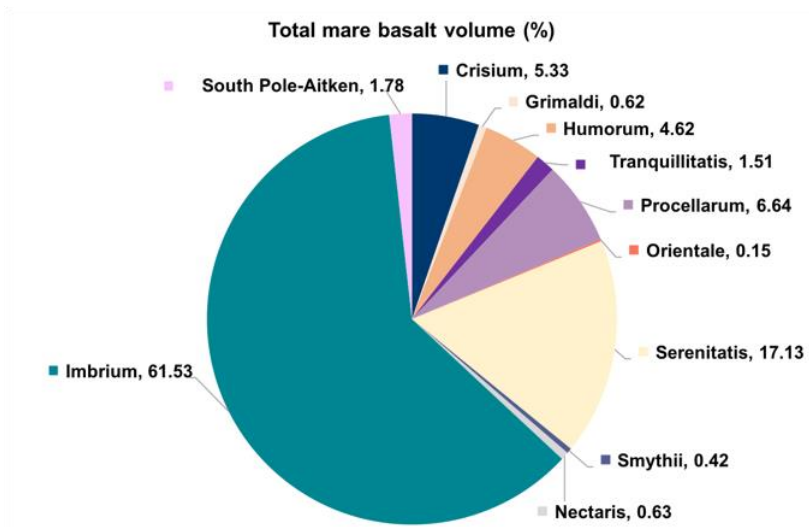
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Figures:

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551 Figure S1. Volumes of erupted basalts and volatiles and lunar atmospheric pressure as a
 552 function of time (from Needham and Kring, 2017). (a) Volume of erupted basalts as a
 553 function of time, indicating peak volcanic activity primarily in Imbrium basin ca. 3.5 Ga;
 554 the inset in each graph shows results for the time period from 3.0 Ga to 1.0 Ga at an
 555 expanded scale. (b) Mass of volatiles, primarily CO and S, degassed during mare
 556 emplacement. (c) Atmospheric surface pressure resulting from the volatiles released
 557 during mare emplacement, with a peak pressure $\sim 1\%$ of Earth's current atmospheric
 558 pressure, corresponding to peak volcanic activity 3.5 Ga. Quantified results for panel (a)
 559 are included in Table S2, and for panels (b) and (c) in Table S3 of Needham and Kring
 560 (2017).

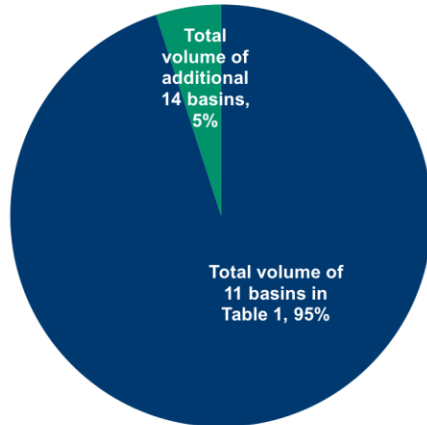
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a) Total mare basalt fill for the 11 basins considered in Table 1 of Needham and Kring (2017).

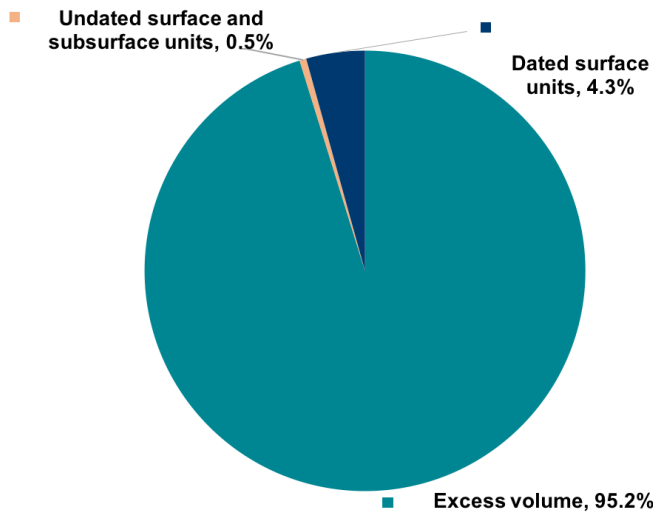
Percentage of total mare basalt volume in 11 major basins and 14 other basins used in the total mare basalt fill calculation of Needham and Kring (2017)



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b) Percentage of total mare basalt volume in 11 major basins and 14 other basins used in the total mare basalt fill calculation of Needham and Kring (2017).

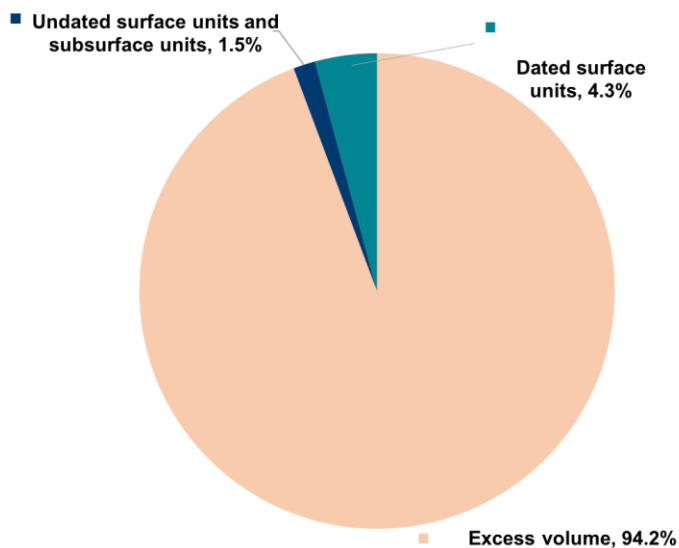
Imbrium basin: Percentage of total mare fill in each of the categories (dated, undated, underlying, and excess) utilized by Needham and Kring (2017)



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c) Imbrium basin: Percentage of total mare fill in each of the categories (dated, undated, underlying, and excess) utilized by Needham and Kring (2017).

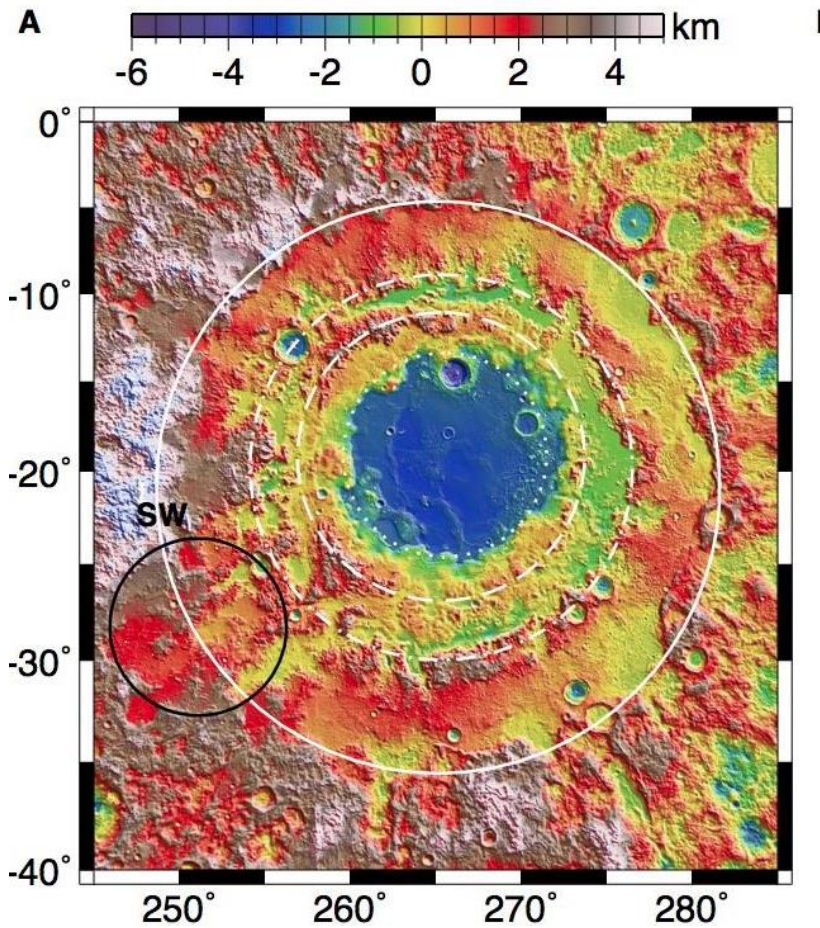
Serenitatis Basin: Percentage of total mare fill in each of the categories (dated, undated, subsurface, and excess) utilized by Needham and Kring (2017)



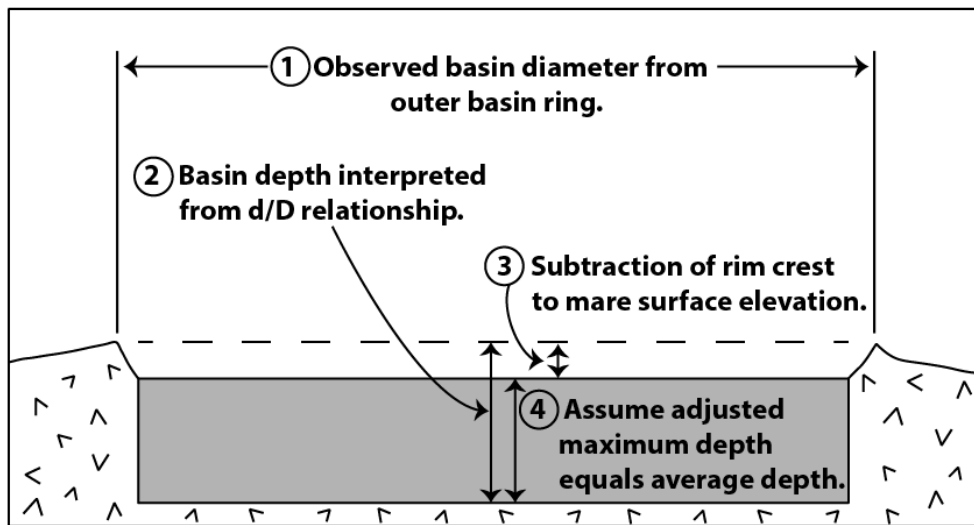
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d) Serenitatis Basin: Percentage of total mare fill in each of the categories (dated, undated, underlying, and excess) utilized by Needham and Kring (2017). See Figure S3 for explanation of Needham and Kring (2017) unit age assignments.

Figure S2. Relative percentages of mare fill.



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582 Figure S3. Topography of a typical multi-ring basin geometry and mare fill: a) Orientale

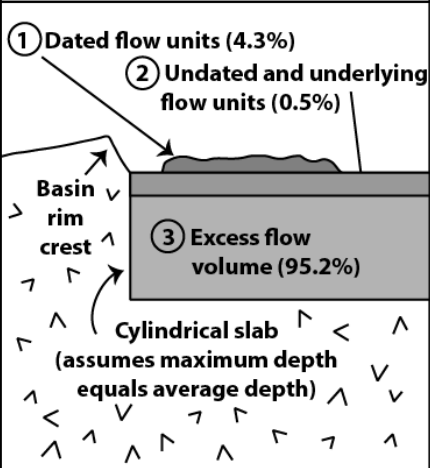
583 basin topographic map showing that the deepest part of the basin (blue) is not equivalent

584 to the average depth. b) Assumption that the maximum basin lava fill thickness is

585 equivalent to the average basin fill depth in the Needham and Kring (2017) model.

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Needham and Kring Nomenclature	Candidate Sources of Uncertainty
 <p>① Dated flow units (4.3%)</p> <p>② Undated and underlying flow units (0.5%)</p> <p>③ Excess flow volume (95.2%)</p> <p>Basin rim crest</p> <p>Cylindrical slab (assumes maximum depth equals average depth)</p>	<p>① Single dated flow unit may have multiple ages, durations.</p> <p>② Units assigned single age of oldest exposed dated flow unit: actual thicknesses, ages and durations unknown.</p> <p>③ "Excess" unit assigned single age of oldest dated flow: this unit certain to be multiple flow units of different ages, thicknesses, volumes and durations. - "Excess" volume overestimated due to assumption that maximum basin thickness = average thickness.</p>

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Figure S4. Cross-section of a multi-ring basin lava fill illustrating assignments of different types of dated units by Needham and Kring (2017) and candidate sources of uncertainty in estimating basin lava fill thicknesses and volumes. Numbers in parentheses show the percentages of each type of unit assigned by Needham and Kring (2017) to age distributions in the Imbrium basin.