1	Lunar megaregolith mixing by impacts: spatial diffusion of basin melt
2	and its implications for sample interpretation
3	Tiantian Liu ^{a,b} , Greg Michael ^b , Kai Wünnemann ^{b,c} , Harry Becker ^b , Jürgen Oberst ^{a,d}
4	^a Institute of Geodesy and Geoinformation Science, Technische Universität Berlin, 10623 Berlin,
5	Germany (tiantian.liu@tu-berlin.de)
6	^b Freie Universität Berlin, Malteserstr., 74-100, 12249 Berlin, Germany
7	° Museum für Naturkunde, Leibniz Institute for Evolution and Biodiversity Science, 10115 Berlin,
8	Germany
9	^d Institute of Planetary Research, German Aerospace Center (DLR), 12489 Berlin, Germany
10	Key Words
11	Moon, surface; Regoliths; Impact processes; Cratering
12	Abstract
13	The formation ages of lunar impact basins are critical to understanding the late accretion history
14	of the inner solar system. Furthermore, the correct interpretation of the provenance and isotopic
15	dates of basin-derived impact melt ('basin melt') is essential for the calibration of lunar chronology
16	function. However, abundances of basin melt in the lunar near-surface are not well understood.
17	Basin melt has been gardened by a long sequence of subsequent impact events, altering its
18	abundance and size distribution. We developed a numerical model to investigate this process by

19 means of the Monte Carlo method in a spatially resolved model. The fraction of melt in ejecta was

20 tracked globally and at the Apollo 14-17 and Luna 20 sampling sites and was compared with K-21 Ar age distributions of lunar impact melt breccias. It was found that melt produced by the very 22 large SPA basin as well as the relatively late-forming Imbrium basin should be dominant in the 23 near-surface (top one meter). The simulation shows that the melt component at the Apollo 14-17 24 and Luna 20 sites is strongly affected by nearby mid- to late-forming basins. Imbrium melt should 25 be abundant in Apollo 14-17 samples; Crisium melt is the most significant component of basin-26 sourced melt in Luna 20 samples; all the Apollo 14-17 and Luna 20 samples could include melt 27 from Serenitatis and the SPA basin; Nectaris melt should occur in Apollo 16, Apollo 17 and Luna 28 20 samples; and Orientale melt has no significant mixing in the Apollo 14-17 and Luna 20 29 sampling sites. In general, besides a prominent age peak at 3.9 Ga (related to the Imbrium basin), 30 the model predicts pronounced abundance peaks of older basin melt (>3.9 Ga) which tend to be 31 absent from distributions of K-Ar ages of impactites from landing sites. The diffusion 32 characteristics of basin melt suggest that for future sampling aimed at collecting early basin melt, 33 the re-excavation zones of late impact craters larger than tens of kilometer in diameter inside basins 34 may provide the highest abundances of melt from early basins.

35 1 Introduction

The lunar surface preserves a record of impacts in the inner solar system. Having a rigid crust and no atmosphere, its crater population is the best preserved among the solar system's planetary bodies, providing an essential record of late accretion in the inner solar system. Nevertheless, knowledge of the impact rate between 4.5 and 3.7 Ga, which may be related to different impactor populations, remains contested. Based on radioisotope ages (U-Pb system) of lunar samples that show clustered ages at ~3.9 Ga, Tera et al. (1974) suggested that there may have been an impact spike in the early lunar history ('lunar cataclysm'). This hypothesis was later further substantiated 43 after larger numbers of K-Ar ages on impact rocks became available (Fernandes et al. 2013; 44 Michael et al. 2018). The Nice model (Gomes et al. 2005; Morbidelli et al. 2012), which suggested 45 that late migration of the giant planets could have destabilized part of the asteroid belt injecting 46 projectiles into the inner solar system, was developed as a possible mechanism that might explain 47 the occurrence of an impact spike. In contrast, based on studies of crater size-frequency 48 distributions of surfaces in basins, Neukum (1983) and Hartmann (1995) argued that the Moon 49 was hit only by one impactor population (the collisional remnants of planetesimals). In their view, the impact flux decayed monotonically and the dominant isotope ages of ~3.9 Ga just reflect 50 51 sampling bias caused by the mixing of Imbrium ejecta.

52 The lunar basins can be used to constrain the impact rate in its early history if their ages are known. 53 All lunar basins, including the youngest Orientale basin, formed before ~3.8 Ga by hypervelocity 54 impact events. If the age of melt rocks that originate from a particular basin event can be 55 determined reliably (e.g., by the K-Ar or other chronometers), it provides a calibration point for 56 the lunar cratering chronology (Hiesinger and Head 2006). Basin melt also provides information 57 on the regional chemical makeup of the crust, possibly providing clues to the lunar differentiation 58 history. In addition, determining the distribution of basin melt is significant for lunar exploration, 59 being a prime target for future robotic or human sample return missions (Ryder et al. 1989).

Melt from a specific basin is affected by the long-term gardening processes by later impact events. Its volume and composition is changed by re-melting, and its spatial distribution is altered by entrainment in younger ejecta materials (Michael et al. 2018; Liu et al. 2019, the characteristics of melt evolving distribution are described in supplementary section 2). The lunar meteorites and the lunar highland rocks collected by the Apollo and Luna missions have provided some information on the abundance of impact melt both in an average sense and at specific sites. However, there remains a considerable level of ambiguity in the interpretation of the origin of lunar meteorites and
returned samples (Spudis et al. 2011).

In this study, we developed a numerical model to investigate the diffusion of basin melt by impact gardening. We use "diffusion" to describe the movement of impact melt down its concentration gradient by repeated impact events. The aim is to provide a picture of the potential distribution of basin melt by using a new modeling approach, and thus provide clues for the interpretation of returned samples regarding which age distributions of basin melt should be expected at previous and future landing sites. These data may also be used to assess interpretations of isotopic age distributions in the literature and in future studies.

In section 2, we present the structure of the model. In section 3, the melt component in the nearsurface both on a global scale and at specific sampling sites is estimated and compared with distribution patterns of K-Ar ages. The spatial distribution of melt from three mid- to late- basins (Serenitatis, Crisium and Imbrium) in variable proximity to lunar landing sites is also examined. In section 4, we describe the implications for the interpretation of collected lunar samples and discuss sources of uncertainties in the model.

81 2 The Model

This paper is the third in a series investigating the cumulative melt diffusion by impact gardening. Michael et al. (2018) presented the surface-averaged model, where the volume of un-heated material and melt with different ages at different depth was traced. On this basis, Liu et al. (2019) investigated the diffusion behavior along a lateral path as well as by depth. Then, not only the distribution with depth, but also the lateral distribution along a path of a great circle was further recorded. This work extends the previous models, providing a more complete understanding of the 88 melt evolving characteristics in three spatial dimensions, where the volume of unheated material 89 and melt with different ages at all the depth over the global surface will be traced. We outline the 90 main elements of the model here where we focus in particular on the new components of the model. 91 Further detail is given in the Supplementary Material. In addition, in this model we do not consider 92 the maria, because mare deposits cover parts of surface and occurred late, and hence would not 93 significantly affect basin melt redistribution.

94 We used the Monte Carlo method to simulate the impact gardening process over the history of the 95 observable surface of the Moon. The size-frequency distribution of generated craters conforms to 96 the production function (PF) of Neukum (1983). The occurrence time of impacts was calculated 97 combining the chronology function (CF) and PF (Neukum 1983). The minimum crater diameter 98 considered was chosen as 5 km. The transient crater size, D_t , was calculated based on scaling laws 99 (Melosh 1989; McKinnon et al. 1997). The volume of the generated melt markedly increases with 100 Dt according to a power law (Cintala & Grieve 1998). The thicknesses of both ejecta and melt 101 decrease with distance from crater center (r in km) following a power law, but the fraction (i.e. 102 concentration) of melt in the ejecta linearly increases with distance (Liu et al. 2019; Melosh 1989). 103 We note that our model considers the re-melting of the excavated material for individual impact 104 events, but the possible resetting of the K-Ar clock in hot ejecta blankets was not incorporated 105 because the detailed behavior of the K-Ar system in feldspars from impactites is poorly understood. 106 (See Liu et al. 2019 and supplementary section 1 for details).

107 Materials ejected from craters have high kinetic energy resulting in mixing with local materials on 108 re-impact. The degree of mixing is expected to increase with distance from the ejection point. We 109 considered such local mixing in this new model. Oberbeck et al. (1975) proposed a mixing ratio 110 of local material to ejecta, $\mu = 0.0183r^{0.87}$. However, these values, particularly at larger scales, 111 were suggested to be overestimated in comparison with laboratory cratering experiments (Schultz 112 & Gault 1985). The value of μ was thus modified by roughly half when μ is larger than 5.0: $\mu' =$ 113 $\mu/2 + 2.5$ (Petro & Pieters 2006).

The model used point sets on a sphere to record the material component. The locations of these points were calculated based on the Fibonacci Lattice (González 2010) to make them uniformly distributed on the surface. Each point was related to a cell. For each impact event, the points within tracing ranges (five radii of transient craters) will be found first. In each of these points, the melt volume, the total volume including both melt and un-melted component, and the thickness of ejecta (and melt lens within craters) will be recorded. When projecting the point set onto a plane, we used a Voronoi diagram (i.e. Thiessen polygon) to represent the cell of each point.

Because of the increasing number of deposition layers as the simulation progresses, the layer sequence was periodically simplified to one: 0.2×1.9^a m, where a = 0, 1, ..., 20. While simplifying, all the material within set layers will be mixed together leading to a more average composition. The shallower layers are thinner to preserve a fine resolution of the melt component in the near surface; deeper layers are thicker yielding a more averaged composition.

126 3 Results

Lunar meteorites and the immediate source of current lunar samples are from the near-surface down to a few or tens of meters. Understanding the source and age distribution of the melt component in the surface layer is critical to our understanding of the lunar chronology, and key in the selection of future sampling sites. This study therefore focuses on the distribution of impact melt in the near-surface (top one meter).

132 3.1 Timing of basin-forming events

133 To investigate the melt distribution of the lunar impact basins, a model was created using both the 134 known sizes and positions of the basins and randomly distributed craters smaller than 300 km. 135 Thirty basin-forming events, the crater populations of which were reinvestigated using the buffered 136 non-sparseness correction technique, are included in our simulations (Orgel et al. 2018). The 137 buffered non-sparseness correction technique takes crater obliteration into account for the 138 superposed crater densities, providing more accurate measurements of the occurrence times of 139 basins (Riedel et al. 2018). Neukum et al. (2015) present a more complete list of lunar basins where 140 some potential large lunar basins, such as Aestuum, Lamont, and Hertzsprung basins, are not 141 included in the list in Orgel et al. (2018). The occurrence times of these basins, however, have not 142 been calculated. They are therefore not included in this model. The erased structures of these basins 143 indicate their early formation times. The consequences of excluding these basins are discussed in 144 section 3.3.

Since the scaling laws used to calculate transient crater diameter (D_t , supplementary section 1) do not extrapolate well to basin sizes, we adopt the D_t values of 11 basins from Wieczorek & Phillips (1999) where the transient basin size was determined using gravity models. The D_t of the other basins was interpolated. The locations of the basins showing D_t and age is presented in Figure 1, and Table S1 presents the age, location and size of all the included basins.

150 It should be mentioned that the formation times of basins included in the simulations are taken as 151 a plausible configuration of impact times, and are not claimed as being in any way 'well known'. 152 Our goal is to address the question "What are the consequences of such a configuration of impacts 153 for the sample record?" and to use the comparison with the actual record of isotopic ages to inform 154 our understanding of both the period and the process of basin melt migration into lunar impactite





156



162 Simulated impacts start slightly before the estimated formation time of the SPA basin at 4.31 Ga. 163 In one simulation run there were 162517 impact events in total, including basin-forming events. 164 Many older impacts have no significant influence on the present near-surface, since the majority 165 of their melt was buried deeply by the ejecta of younger impacts. We improved the efficiency of 166 the model by filtering out those impacts which we are certain to have negligible contributions to 167 the near-surface melt abundance (the filtering procedure is described in supplementary section 3). 168 After the filtering, 29176 impacts remain, while 133341 have been removed. As a consequence, 169 the time for one run is decreased from five days to four hours. Of those removed, 96.9% are smaller than 30 km. Because of the greater impact flux during the early period, the majority of craters filtered out were old (Figure S 4b), with 99.97 % being older than 3.5 Ga. The difference of the relative expected melt component at different depths is smaller than ~0.02, indicating that the features of melt component distribution are well-preserved by the filtering procedure.

174 3.2 Average melt component in the lunar near-surface layer

175 If lunar meteorites provide a spatially stochastic sample of lunar highland materials, the statistical 176 radioisotope age record of melt-bearing impact rocks among lunar meteorites could reflect the 177 average distribution of the melt component. We estimated the fraction of impact melt in the top 178 one meter for comparison with the radioisotope age record (Figure 2).

179 The predicted distribution of melt with different ages is shown in Figure 2c. Basins are the main 180 contributors of melt older than ~3.8 Ga. As indicated by the dashed arrows in Figure 2, the largest, 181 SPA basin (~2500 km) is predicted to provide the most abundant melt in the near-surface; The 182 late-forming Imbrium basin (~1320 km) is the second largest contributor of melt. Other basin 183 events also left distinct traces: the melt of the youngest Orientale basin (3.81 Ga) is $\sim 0.4 \times$ as 184 abundant as Imbrium melt leading to the secondary peak at 3.81 Ga; several basins forming 185 between 4.10 and 4.25 Ga generate a bulge during this period. To predict the contribution of basin-186 sourced melt, Table S2 presents volumes of melt from basins and smaller impacts. In general, 187 younger basins leave more melt in the near-surface today.

We expect that there would have been basin-forming impacts prior to those which we are able to observe today (e.g., Neumann et al. 2015; Zhu et al. 2019). It can be expected that impacts occurring in the period between crustal solidification and the time of the presently observable oldest surfaces would leave a melt component, which could persist within the crust until now. These early impact events would heat and alter more crust material and the generated melt ought to leave a trace from depth up to the surface until the present day (see Michael et al. 2018). We recognize, however, that this material could be highly comminuted by impact gardening and partially remelted by younger impacts near the surface, which may leave it unrecognizable there (see further discussion in section 4.1).

During the last 3.0 Ga, the impact rate was lower and remains constant, melt diffusion is thus generally constant and the melt production is much less. Such a lower impact rate leads to a lesser amount of older melt being re-melted. Therefore, in the near-surface, younger impacts leave comparable quantities of melt to older basins, as indicated by relatively large impacts seen as random peaks in Figure 2c, such as those at 0.4 and 3.3 Ga.

Fernandes et al. (2013) compiled published K-Ar data and calculated the relative probability of 40 Ar/³⁹Ar impact ages from lunar meteorites, where 65 samples were included and the age was corrected for monitor age and decay-constant. Their plot is shown in Figure 2b. It is seen that, in the radioisotope age record of impact melt breccias in lunar meteorites (Figure 2b), there is abundant melt older than 3.5 Ga as expected, but less than expected with ages >3.8 Ga. In addition, in the distribution of isotopic dates (Figure 2b), the presence of melt from later impact events is more significant. The possible cause of these differences will be discussed in section 4.1.



210 Figure 2 (a) Size distribution of basins normalized to the diameter of Imbrium basin. The formation times of SPA and Imbrium basin from Orgel et al. (2018) are $\mu 4.31^{+0.019}_{-0.021} \mu 3.87^{+0.035}_{-0.046}$ Ga, respectively. (b) 211 Relative probability plot of ⁴⁰Ar/³⁹Ar from 65 impact melt clasts in lunar meteorites (after Fernandes et al. 212 213 2013). The vertical scale is linear. The peak near 3.8 Ga is commonly interpreted to reflect the age of the 214 Imbrium impact. (c) Fraction of melt component (i.e. relative melt abundance among all the generated melt) 215 in the top one meter of lunar near-surface (left scale). The expected melt generated by SPA, Imbrium and 216 Orientale basins are indicated by three vertical arrows. To show the small melt quantity from smaller 217 impacts, melt fractions have been normalized to the absolute melt fraction derived from the second most 218 abundant melt reservoir, the Imbrium basin (i.e. its relative abundance is equal to 1.0). The crater formation 219 rate (right scale) shows that the high impact rate in the early lunar history is accompanied by a greater melt 220 production. Note that if melt fractions were normalized to SPA melt fractions, the relative quantity of other

melt fractions would be so small that specific distribution features are easily masked. Thus, the relativeabundance of SPA melt fraction exceeds 1.0, and the exact value is not presented.

3.3 Present-day distribution of melt from basins formed between 4.2 and 3.8 Ga
Due to the weaker gardening after 3.0 Ga, abundant melt of basins formed between 4.2 and 3.8 Ga
(mid- to late-forming basins) could survive in the near surface, and is thus very likely to be sampled.
As representative examples, we present the global near-surface melt distribution of the Serenitatis,
Crisium and Imbrium basins (Figure 3).

228 Besides the mare-related volcanism, which is not taken into account in this model, the only process 229 that can significantly modify the distribution of basin melt is gardening by other basin-scale impact 230 events. Smaller impact events can only re-melt small portions of older melt. Because nearly no 231 disturbance by younger basin forming impacts occurred, Imbrium melt is well-preserved and 232 predominant in the near surface (Figure 3a). Imbrium ejecta covered the northwest part of Crisium 233 ejecta, decreasing the abundance of Crisium melt in the near surface. Serenitatis is located between 234 the Imbrium and Crisium basins (Figure 1). The excavation zones of both the Imbrium and Crisium 235 basin exhumed the earlier Serenitatis ejecta. In the model, both the Crisium and the Imbrium ejecta 236 entrained Serenitatis melt, transporting it to more distant locations. Therefore, the coverage of 237 Serenitatis melt is almost equal to the merged zone of both Crisium and Imbrium ejecta (the main 238 coverage by Imbrium and Crisium melt is outlined by a dashed curve in Figure 3). Nevertheless, 239 the quantity of entrained Serenitatis melt is low. The average melt fraction from Imbrium is about 240 ten times larger than from Serenitatis and double that of Crisium.

Subsequent smaller impact events garden the basin melt locally, making the distribution patchy.
They diminish or even remove the melt from the excavation zones. The fraction of the basin melt

entrained in the ejecta of smaller impacts depends on their scale and location. If an impact event is large enough to excavate older basin melt, it can form melt-enriched zones in the near-surface, especially if it excavates melt from the basin interior or the melt near the rim of the basin. Two regions enriched in Serenitatis melt are indicated by the black arrows in Figure 3c. Most smaller impacts occur on the ejecta areas of basins, excavating only a limited amount of basin melt and distributing it further, slightly reducing the proportion of basin melt in the near-surface. These zones are shown as faded red and yellow in Figure 3a and b.

250 The results have some important implications for future sampling aimed at collecting the melt from 251 these three basins. Within the range of several basin radii from Imbrium's center it is very likely 252 to find its melt. Crisium melt could also be abundant, but will be mixed with that of Imbrium 253 within the Imbrium ejecta field. To limit the influence from the Imbrium basin, it would be better 254 to collect samples from the eastern part of Crisium basin. Sampling Serenitatis melt without an 255 admixture from other basins is difficult. It is the most wide-spread, but due to its low probability 256 of preservation at the surface, re-excavation zones may be the best candidates for samples 257 possessing a higher fraction of its melt. It should be noted that, most of regions within the basins 258 were flooded by mare basalts, but if impact events penetrated mare basalts and excavated 259 underlying impact melt, this basin melt should be the most abundant component in the ejecta, 260 besides mare basalt.



261

Figure 3 The global present-day distribution of melt from Imbrium (a), Crisium (b), and (c) Serenitatis basin in the near-surface (top five meters). Warmer colors indicate higher melt fractions (i.e. fraction of basin melt among all impact melt). Red stars indicate Apollo 14-17 and Luna 20 sampling sites. The main coverage of Imbrium and Crisium ejecta is indicated by the dashed regions. The black arrows in (c) indicate re-excavation zones of Serenitatis melt. (d)-(f) are the expected Imbrium, Crisium and Serenitatis melt abundances without local mixing processes.

268 3.4 Basin-sourced melt abundances at Apollo and Luna sampling sites

Without knowledge of their source, the context of lunar samples is difficult to evaluate. We calculated the probable fraction of basin-sourced melt in the near-surface at Apollo 14-17 and

Luna 20 landing sites. These simulation results can be helpful for the interpretation of sample data and radioisotope ages in future work (Figure 4a-e). The volumes of melt from basins and smaller impacts are shown in Table S3. Note that, since no mare basalt or other late volcanic deposits are taken into account in this study, the following discussions concerning the fractions of basin melt do not consider admixtures of mare basalt which were found to be abundant in some returned samples (e.g., Shervais et al. 1985).

277 The melt component at these sites is strongly influenced by the ejecta of the youngest and closest 278 basin. As seen from Figure 3, the Apollo 14-17 sampling sites were covered by Imbrium ejecta, 279 and the Luna 20 samples were extensively mixed with Crisium ejecta. Therefore, impact melt 280 components in the Apollo 14-17 samples are expected to be dominated by Imbrium melt. The 281 fraction of Imbrium melt is expected to be ~ 1.0 in the melt with ages of 3.87 Ga (Table S2), which 282 is suggested by its high relative abundance at 3.87 Ga in Figure 4a-d highlighted by red color. 283 Luna 20 samples would contain abundant Crisium melt with a fraction of 0.92 in the melt with an 284 age of 4.07 Ga (Table S2), indicated by the pronounced peak with this age (Figure 4e) highlighted 285 by green color. Since both Imbrium and Crisium excavated the older Serenitatis material, all the 286 Apollo 14-17 and Luna 20 samples should be mixed with at least some Serenitatis melt. Although 287 the volume is small, Serenitatis melt is still the major contributor in melt with an age of 4.22 Ga 288 (Table S2). Traces of the nearby older basin events should also be found in impactites collected 289 near the surface, but their contribution would be small due to dilution by the later events. For 290 example, Nectaris melt could be found at the Apollo 16-17 and Luna 20 landing sites (4.17 Ga in 291 Figure 4c), but its quantity is small. The melt of the youngest Orientale basin has no significant 292 mixing in the Apollo 14-17 and Luna 20 sampling sites.

293 Not all large peaks in the melt age distribution were derived from basins. Younger melt generated 294 by smaller impacts (i.e., non-basin) also leaves traces, as those younger melts have experienced 295 less gardening. Taking Luna 20 as an example, according to its location, the high quantity of melt 296 at 3.87 Ga (Figure 4e) is derived from smaller impacts at this time, rather than a basin forming 297 event. For the Apollo 14 sampling site, the model predicts only the peaks at 4.31 Ga, 4.22 Ga, 4.10 298 Ga and 3.87 Ga to be caused by basin forming events. Smaller impacts are randomly distributed 299 over the lunar surface, and thus the simulation will not exactly match what is found at a specific 300 site. The expected peaks caused by younger impacts are therefore different from some sample ages. 301 This might also be the case for the record from lunar meteorites (Figure 2, Fernandes et al. 2013).

Michael et al. (2018) aggregated 117 K-Ar age measurements of Apollo 14-17 and Luna 20 impact melt breccias in highland rocks. The relative probability of K-Ar ages is plotted above each histogram in Figure 4. By comparing the expected melt abundance with the distribution of K-Ar radioisotopic ages, the issue of whether the peaks displayed in the radioisotopic ages is related to the basin forming events could be further evaluated.

307 Note that the recently proposed age of the Imbrium basin derived from U-Pb dates of phosphates 308 in breccias from the Apollo 14 landing site is 3.9±0.02 Ga (Snape et al., 2016). Crisium age was 309 proposed to be 3.95-3.85 Ga (Stöffler 2006). The apparent age of the Imbrium basin (3.87 Ga) and 310 Crisium basin (4.07 Ga) from crater size-frequency distribution (CSFD) and the Neukum (1983) 311 chronology function, may be slightly inaccurate because of a combination of factors. Earlier decay constants used to calculate ⁴⁰K-⁴⁰Ar ages and ⁸⁷Rb-⁸⁷Sr ages during the 1970s and early 1980s are 312 313 slightly different from recent determinations (Jourdan 2012; Naumenko-Dèzes et al. 2019; 314 Rotenberg et al., 2012). Furthermore, uncertainties of ages of neutron fluence monitors used in Ar-315 Ar dating and uncritical use of data from disturbed isochrons and complex Ar release spectra are additional biases on presumed basin ages that use isotopic dates and such biases also affect age
distributions such as Figure 2 and 4 (see Jourdan et al., 2009; Jourdan, 2012; Norman et al., 2010;
Fernandes et al., 2013). In the following, we will use the 3.87 Ga age as representing the age of
Imbrium, even though the true age may be 30-50 Ma older. The reasoning is that both age
distributions (Figure 2 and 4) and the impactor flux and basin ages used as a basis for our model
were affected by the biases mentioned above.

322 We note that all isotopic age distributions from landing sites of the present study reveal the 323 Imbrium peak of 3.87 Ga. However, additional 'age' peaks near 3.87 Ga are also sometimes 324 present and may either result from smaller impact events or may reflect the presence of disturbed 325 age spectra in the distributions. Both the Apollo 17 and Luna 20 sampling sites are expected to 326 possess abundant Crisium melt (green histograms of 4.07 Ga in Figure 4), but the distribution of 327 radioisotope ages does not display the corresponding peaks, and only few ages occur near 4.1Ga. If the proposed Crisium age of 3.95-3.85 Ga is taken (Stöffler 2006), which is indicated by the 328 329 semitransparent-green bars in Figure 4, the simulation results would be consistent with the 330 radioisotopic dates shown as the peak of \sim 3.9 Ga. It may suggest that the melt of \sim 3.9 Ga in the 331 Apollo 17 and Luna 20 samples could be mainly derived from the Crisium melt. If so, then the 332 formation time of Crisium basin is more likely to be ~3.9 Ga which is close to the age of Imbrium 333 basin, and the overlapping of semitransparent-red and green bars at ~3.9 Ga (Figure 4d) indicates 334 that Apollo 17 samples may be a mixture of both Imbrium and Crisium melt.



336 Figure 4 Relative distribution of K-Ar dates obtained on impact melt breccias at the Apollo 14-17 and Luna 337 20 sampling sites from samples (upper diagrams, Michael et al. 2018) in comparison with relative melt 338 abundances at the Apollo 14-17 and Luna 20 sampling sites based on simulations (lower histograms; 339 relative abundance among all the generated melt). The simulation data reflect melt abundances over a radius 340 of 50 km at each landing site in the top 1 m of the near-surface. Model histograms are shown in black 341 (absolute scale, left) and in grey (logarithmic scale, right). In the black histograms, the peaks caused by 342 Imbrium (3.87 Ga) and Crisium (4.07 Ga) melt are highlighted by red and green color, respectively. Both 343 ages were calculated based on the Neukum (1983) crater chronology and decay constants of that time, which are also the basis of the impactor flux used in our model (see text for further explanation). Note that 344 345 Pb-Pb ages of phosphates suggest that the true age of the Imbrium basin is probably 3.92 ± 0.02 Ga (the 346 semitransparent-red bar, Snape et al., 2016). Stöffler (2006) suggested that Crisium-derived melt was 3.95-347 3.85 Ga (the semitransparent-green bars). Overall, the abundance of Imbrian 'ages' near 3.87 Ga at the 348 Apollo landing sites agree with predictions from the model, whereas older K-Ar ages are much less common 349 or do not occur (Apollo 15).

350 4 Discussion

351

4.1 Implications for the interpretation of melt provenance at lunar landing sites

The model predicts less young melt and more older melt (>~3.8 Ga) than implied by the K-Ar age distributions (Figure 2 and Figure 4). In addition, some of the expected peak values of melt

abundance show different ages compared to the K-Ar age peaks. The main assumption in comparisons between age peaks in the model and published K-Ar ages is that the latter represents a statistical measure of the abundance of melt rock at the particular landing site and also globally. However, this assumption is not certain, and may be incorrect. A second assumption is that the model ages of basins are correct, but they could be inaccurate due to the inherent uncertainties in the lunar chronology function.

360 The radioisotope age distributions show prevalent younger melt rock. Many previous studies have 361 investigated plausible causes of the apparent deficient of the old melt rock (e.g., Chapman et al. 362 2007; Cohen 2000; Hartmann 1975; Zellner 2017). It could be caused by the complete or partial 363 Ar loss of older melt rocks and mineral clasts due to heating and shock, whereas inheritance of Ar 364 from undigested older feldspar clasts in melt rocks might yield apparent ages that cannot be related 365 to specific events (Korotev 1994; Norman et al. 2006; Norman & Nemchin 2014). Older clasts in 366 melt breccias and their minerals may have been contaminated by younger (Imbrian) KREEP rich 367 impact melt which may influence Ar budgets. In addition, older melt clasts may be less abundant 368 than predicted because they were digested in large impact melt sheets or incorporated in warm 369 ejecta blankets and all previously accumulated radiogenic Ar was lost because of degassing 370 (Fernandes et al. 2013; Norman et al. 2006; Norman et al. 2010). Furthermore, as the grain size of 371 melt rocks decreases with time due to gardening, older melt is more likely to survive near the 372 surface in the form of tiny clasts or fine-grained minerals in breccia matrices. The larger clasts of 373 melt rocks from younger basin forming impacts such as Imbrium may have been preferably chosen 374 for dating, and thus young ages show up more frequently.

A significant number of published ⁴⁰Ar-³⁹Ar ages on lunar melt rocks or feldspars are based on
 perturbed age spectra, most of which do not satisfy current statistical tests for a point-like

377 geological event (at least ~70% of the age spectrum (Cassata et al. 2009)). Such samples may yield 378 apparent dates that are shifted towards younger or older dates by tens and even hundreds of 379 millions of years. A review of the geochronology of impact melt breccias by Grange et al. (2010) suggests that, when using strict data filtering (Jourdan et al. 2009), only a few tens of ⁴⁰Ar/³⁹Ar 380 381 data of almost 300 sample analyses yield well-behaved and reliable age spectra (e.g. ⁴⁰Ar/³⁹Ar 382 plateau ages given by Norman et al. 2006, Norman et al. 2010, and Fernandez et al. 2013). A better 383 understanding of the chronology of major basin-forming impacts would require a much more 384 systematic study of a larger number of samples, both from existing collections and from sample 385 sets brought back by future missions.

386 Although a comparison of detailed aspects of the age distributions with the model results are 387 difficult (e.g. the number of recorded impact events or ages of specific basins, Niihara et al. 2019; 388 Norman et al. 2006) some robust general features may help to advance interpretations in future 389 studies. According to our results, basin ejecta from the late-formed Imbrium and also the Crisium 390 basin, should be the most significant components which should show up in the distribution in the 391 K-Ar ages. In case of Imbrium, K-Ar ages near 3.87 Ga are common and match the model (Figure 392 4). For Crisium, a model age of 4.07 Ga could reflect data from some Apollo landing sites, whereas 393 data from Luna 20 (which is closer to Crisium than the Apollo landing sites and relatively far away 394 from Imbrium) would be more supportive of a relatively young age (3.85-3.95 Ga; Stöffler 2006). 395 Both the model of ejecta ages and isotope age distributions at landing sites may be affected by 396 locally produced impact melt from small impacts (see Figure 2 and 4). Such biases may be 397 evaluated by multi-chronometer studies of samples, combined with petrological and geochemical 398 information. A key result of the data in Figure 4 is the absence (Apollo 15) or rarity of > 4.1 Ga 399 old impact melt (i.e. melt from basins such as Serenitatis and South Pole Aitken) among isotope 400 ages which is in contrast to the predicted minor abundance of such melt at the landing sites. As 401 discussed above, this could reflect efficient loss of Ar from old melt clasts by younger heating 402 events, biases during sampling of melt breccias, for instance if ancient melt rocks have a small 403 grain size, or inaccuracies in our model.

404 4.2 Implications for future basin-melt targeted sampling

Linking the lunar samples to specific basins is a critical step in understanding the lunar history.
Other than Imbrium, there is little consensus on which melt rocks or clasts retrieved from the
Apollo and Luna missions were derived from specific basin events (Stöffler 2006).

Theoretically, the best locations to collect basin melt would be the basin floor where the majority of melt is deposited after basin formation. However, old basins were mostly later filled with mare basalt, burying the impact melt by up to several kilometers.

411 Later impacts that are larger than tens of kilometer in diameter with excavation depths of several 412 kilometers are likely to penetrate the mare basalt cover and form regional domains enriched with 413 older melt rocks. Such impact craters are even more promising when located near the basin rim, 414 because of the higher melt abundance there. For example, although the majority of Serenitatis melt 415 was buried, the model predicts that there are still some regions in the near-surface possessing a 416 large fraction of Serenitatis melt as indicated by arrows in Figure 3c. The potential for excavation 417 of deeper ejecta blankets by young impacts has been also discussed in geochronological and 418 geochemical studies of impactites from the Apollo 16 landing site. For instance, recent data 419 support arguments that the 50 Ma old North Ray Crater excavated deeper stratigraphic ejecta units 420 that were mainly derived from the Imbrium impact (Norman et al. 2010; Norman et al. 2016). Clast 421 proportions, age distributions and geochemical characteristics in the deep units are different from

the more shallow Cayley Plains Formation in that area, which is also interpreted to be Imbrium-derived (Norman et al. 2010).

424 The SPA region is also one of hot targets of future lunar missions (e.g. oft-suggested MoonRise 425 mission). Figure 5 presents the predicted melt composition inside the SPA cavity (top one meter). 426 Among all the generated impact melt older than 3.8 Ga, the SPA melt could be far more than those 427 of later-formed impact events. In addition to the SPA melt, impact melts of Pointcare and Apollo 428 basins would be the most prominent components regarding to the giant-basin forming events. In 429 addition, due to the high impact flux in the early history, the impact melt of smaller impact events 430 could also leave significant abundance until now, which would make the interpretation of surface 431 material difficult.



432

433

Figure 5 Predicted melt component in the top one meter within the SPA region.



435 To estimate the sensitivity of the model, here, we describe the effects of varying input parameters.

436 For each tested parameter, we keep the other values as previously.

437

i) Melt produced in individual impacts

438 The scaling law applied to calculate the total volume of generated melt of individual impacts is 439 derived from observations and numerical models for materials heated above melting point after 440 release from shocked state (Cintala & Grieve 1998). However, the K-Ar closure temperature of 441 feldspars is significantly lower than the solidus temperature of basic lunar rocks, although the 442 applicability of the closure temperature concept in rapid heating and cooling processes during 443 impacts is uncertain. Therefore, the term "melt" used here should be regarded as "hot ejecta" that 444 include all material heated above the K-Ar closure temperature of lunar rocks (Liu et al. 2019). 445 The total volume of "hot ejecta" could be greater than of the estimate for "melt". To check its 446 influence, simulations with slightly (two times) and significantly (ten times) more melt production 447 in each impact were performed. We found that (Figure S5 in supplementary material), in the top 448 500 m, when the melt of individual impact is two times more, the difference of relative melt 449 fraction is generally smaller than 0.05, which results in $\sim 15\%$ difference of melt fraction compared 450 with the result shown in Figure 2. When the melt of individual impact is ten times more, in the top 451 ten meters, the difference of relative melt fraction compared with the result shown in Figure 2 is 452 generally smaller than 0.1; at greater depth, the difference is larger, but is generally smaller than 453 0.2 compared with the result shown in Figure 2. In both cases, however, old basin melt, 454 predominantly remains in the near-surface: it is not erased by later events

455

ii) Ejected melt volume

456 Only a minor portion of the generated impact melt would be ejected outside the craters. In this 457 model, 75% of generated melt stays within the crater, with 25% being ejected. Simulations with 458 instead 20% and 30% ejected melt of individual impacts were performed. The results (Figure S6 459 in supplementary material) show that, in the top 500 m, the difference of relative melt fraction is 460 <-0.02, which results in ~10% difference compared with the result shown in Figure 2a.

461 iii) Sequence of basins

462 Although the sequence of lunar basins is roughly known, some basins display inconsistent 463 sequences based on different measurement techniques (Fassett et al. 2012; Orgel et al. 2018; 464 Stöffler 2006). For example, in the past, the Nectaris basin was suggested to be older than 465 Serenitatis basin, which contrasts with the results of Orgel et al. (2018). To test the influence of an 466 alternative age sequence of basins, we assumed that Serenitatis formed after the Nectaris basin, 467 the ages of other basins remaining unchanged. The simulation result shows that (Figure S7) the 468 difference of the fraction of Serenitatis melt in the near surface in these different age models for 469 Serenitatis and Nectaris is generally smaller than ~ 0.01 . The major difference is the absent of 470 Serenitatis melt surrounding the Nectaris basin since ejecta from the "early-formed" basin is unable to garden the "later-formed" Serenitatis melt. Therefore, given the general sequence of 471 472 lunar basins, especially those formed during the mid- to late- period, the distribution of basin melt 473 in the near surface would not be significantly changed but the melt abundance of early-formed 474 basins surrounding the later-formed basins might be altered.

475 4.4 Other notable uncertainties in the model's prediction of the distribution of basin476 melt

The transient cavity sizes of basins are not well constrained, the estimates being diverse (Hikida & Wieczorek 2007; Miljković et al. 2016; Potter, Kring, et al. 2012; Potts & von Frese 2003). For example, the D_t of Imbrium basin has been estimated to be 744 km (Wieczorek & Phillips 1999) and 895 km (Hikida & Wieczorek 2007). However, although it would change the absolute abundances, the structure of the distribution of basin melt would not be changed (Liu et al. 2019).

In addition, the early Moon, during which time most basins were formed, is thought to have been
much hotter than at later times (Laneuville et al. 2013). The higher internal energy leads to higher

melt production since a warmer target requires less energy to melt (Zhu et al. 2017). The distribution of melt in the ejecta is also affected by the thermal state of the Moon. For a warmer target, Zhu et al. (2017) found that in a basin-forming impact, the melt fraction in ejecta material is almost constant with distance at ~20%. Therefore, on the younger Moon there might be more basin melt near the surface leading to probably even more prominent abundance of these old basin melt in the near-surface (Liu et al. 2019). In addition, for the largest SPA basins, some ejecta may be lost close to five radii because of the high launch velocity.

491 The efficiency of melt diffusion is significantly influenced by the degree of mixing with local 492 material during ejecta emplacement. As seen from Figure 3d-f, without the consideration of this 493 mixing process, the Imbrium melt in the near-surface would be much more abundant. In the 494 western part of the Crisium basin, melt abundance would be reduced due to the coverage by 495 Imbrium ejecta. The fraction of remaining Serenitatis melt would strongly decrease because of the 496 coverage by both Crisium and Imbrium ejecta. Nevertheless, quantitative investigation of the 497 entrainment of local material by ejecta needs to be further studied (Hörz et al. 1983; Zhu & 498 Wünnemann 2013).

Impacts forming craters smaller than 5 km in radii are not taken into account in this model. We expect that the gardening by these smaller impacts would reset the age of a component of the material and mix it to a more homogeneous state. We expect the older basin-derived components to remain essentially the same, minus this younger component.

503 Furthermore, our model did not take into account distal regions beyond five radii. Such processes 504 lead to regional, melt-enriched areas (Hubbard et al. 1971). However, these processes are more 505 sporadic and the volume is small. In addition, most impacts on the Moon occur with an oblique 506 incidence angle. While resulting craters remain mostly near-circular (except for very shallow 507 impact angles), the ejecta distribution can deviate more strongly from concentric symmetry: the 508 ejecta of small craters are asymmetric only at distances where continuous ejecta blankets do not 509 exist whereas the ejecta of large basins are asymmetric even near the crater rim (Shuvalov 2011). 510 Given the large number of the smaller impacts, the asymmetry distribution of ejecta will be 511 averaged out, but the oblique impact angle may significantly affect the ejecta distribution of certain 512 basins.

513 The formation process of the SPA basin is still not fully understood. The large melt volume, 514 resulting from the higher internal energy, may have led to the formation of a deep and broad melt 515 pool within the basin (Potter, Collins, et al. 2012). Our model may underestimate the global 516 distribution of SPA melt, but SPA melt in the near surface should still be predominant on average 517 (but not at landing sites close to Imbrium). However, melts with ages of 4.3 Ga are rare among 518 landing site samples and possible reasons have been identified in the previous discussion. The lack 519 of evidence for SPA melt could imply that there may have been more old basin-forming events 520 that have not been identified.

521 5 Conclusion

The formation time of lunar basins, is crucial for the study of late accretion in the inner solar system. To quantitatively understand the spatial diffusion of basin- and crater-derived impact melt, we developed a numerical model to simulate the long-term impact gardening process for a given impact flux scenario. The abundance and presence of impact melt with different ages, both globally and at specific Apollo and Luna sampling sites, were estimated and compared with K-Ar age distributions of impact melt breccias. Combined with the spatial distribution of basins, the probable distribution of basin-derived impact melt was discussed.

529 On a global basis, the estimated melt component in the near-surface shows, not unexpectedly, that 530 impact melt from the SPA basin should be highly abundant. But very few melt rocks with isotopic 531 ages of 4.3 Ga have been found among lunar samples. Imbrium-derived melt is also dominant in 532 the near surface due to its late formation time and large size and here model and isotopic ages tend 533 to match. Impact melt generated by the other basins was subject to cumulative gardening and thus 534 the remainder in the near surface was significantly diluted, although melt from these basins is still 535 quite abundant at depth. Older basin melt, especially from basins before 3.8 Ga, may have survived 536 in the lunar meteorites, but the expected small fragments, resulting from long-term pulverization, 537 make it difficult to investigate such impact melt clasts. In contrast, because of the low impact rate 538 during the last 3 Ga, the melt of smaller impacts is more easily preserved and the clasts may still 539 be intact, making the occurrence of young impactites more likely among lunar meteorites.

540 The estimated melt abundances at the Apollo 14-17 and Luna 20 sampling sites suggests that the 541 regional impact melt component is closely related to the nearby basins (and occasionally nearby 542 craters). Most of the collected samples at the Apollo 14-17 sites are likely to have been mixed with 543 Imbrium melt, as suggested by previous radioisotope dating and geochemical studies. The model 544 predicts that the modelled melt with an apparent model age of 3.87 Ga (based on the Neukum, 545 1983, chronology function), which appears to correspond to Pb-Pb phosphate ages of ~3.9 Ga 546 (Snape et al. 2016), is almost all derived from the Imbrium basin. This result suggests that the K-547 Ar ages of impact melt breccias near 3.9 Ga in Apollo 14-17 samples represent the mixing of 548 Imbrium melt with un-heated ejecta. In addition, the simulation results indicate that the formation 549 time of Crisium basin could be close to Imbrium basin with the age of \sim 3.9 Ga as suggested by 550 Stöffler (2006), and the impact melt of ~3.9 Ga in the Apollo 17 samples could be a mixture of 551 both Imbrium and Crisium ejecta. The Luna 20 sampling site is close to the Crisium basin. We estimated the fraction of Crisium melt in the melt of 4.07 Ga (the CSFD model age of Crisium
basin; or ~3.9 Ga, assuming the radioisotope age is correct) to be ~0.9.

All the Apollo 14-17 and Luna 20 samples should contain components of Serenitatis melt, but may 554 555 not be mixed with the ejecta of the youngest Orientale basin. The melt of older surrounding basins, 556 including SPA melt, also should occur, however, the content has been diluted by gardening. The 557 Apollo 14, 16, 17 samples may also contain Humorum, Nectaris and Smythii melt, respectively. 558 Little other basin-sourced melt is expected in the Apollo 15 samples than Imbrium, Serenitatis and 559 SPA melt, however, only Imbrian K-Ar ages do occur. Luna 20 samples may contain Smythii and 560 Nectaris melt. Other peaks observed in the K-Ar age distributions more likely derive from smaller-561 scale impact events or reflect disturbed Ar-Ar systematics.

Acknowledgments: We acknowledge discussions about cratering processes and formation of
impact melt with Menghua Zhu, Lukas Manske, and Robert Luther. We thank Wilhelm Zuschneid
for proof-reading and many helpful suggestions. This work was supported by German Research
Foundation (DFG) CRC TRR 170, subproject A4. This is TRR 170 contribution 82.

566 References

567 Cassata, W.S., Renne, P.R. & Shuster, D.L., 2009. Argon diffusion in plagioclase and implications for

thermochronometry: A case study from the Bushveld Complex, South Africa. *Geochimica et Cosmochimica Acta*, 73(21), pp.6600–6612.

Chapman, C.R., Cohen, B.A. & Grinspoon, D.H., 2007. What are the real constraints on the existence and
magnitude of the late heavy bombardment? *Icarus*, 189(1), pp.233–245.

- 572 Cintala, M.J. & Grieve, R.A.F., 1998. Scaling impact melting and crater dimensions: Implications for the
 573 lunar cratering record. *Meteoritics and Planetary Science*, 33(4), pp.889–912.
- 574 Cohen, B.A., 2000. Support for the Lunar Cataclysm Hypothesis from Lunar Meteorite Impact Melt
- 575 Ages. Science, 290(5497), pp.1754–1756.
- 576 Fassett, C.I. et al., 2012. Lunar impact basins: Stratigraphy, sequence and ages from superposed impact
- 577 crater populations measured from Lunar Orbiter Laser Altimeter (LOLA) data. *Journal of*
- 578 *Geophysical Research: Planets*, 117(2), pp.1–13.
- 579 Fernandes, V.A. et al., 2013. The bombardment history of the Moon as recorded by 40 Ar-39 Ar
- 580 chronology. *Meteoritics and Planetary Science*, 48(2), pp.241–269.
- 581 Gomes, R. et al., 2005. Origin of the cataclysmic Late Heavy Bombardment period of the terrestrial
 582 planets. *Nature*, 435, p.466.
- 583 Grange, M.L., Nemchin, A.A. & Jourdan, F., 2010. Review of Ages of Lunar Impact Rocks: Implication
- 584 to the Timing of Serenitatis and Imbrium Impacts and the LHB Model. In *Lunar and Planetary*
- 585 *Science Conference*. Lunar and Planetary Science Conference. p. 1275.
- Hartmann, W., 1995. Planetary cratering I: Lunar highlands and tests of hypotheses on crater populations.
 Meteoritics, 30, p.451.
- 588 Hartmann, W.K., 1975. Lunar "cataclysm": A misconception? *Icarus*, 24(2), pp.181–187.
- 589 Hiesinger, H. and Head III, J.W., 2006. New views of lunar geoscience: An introduction and overview.
- 590 Reviews in mineralogy and geochemistry, 60(1), pp.1-81.
- 591 Hikida, H. & Wieczorek, M.A., 2007. Crustal thickness of the Moon: New constraints from gravity
- 592 inversions using polyhedral shape models. *Icarus*, 192(1), pp.150–166.

- Hörz, F., Ostertag, R. & Rainey, D.A., 1983. Bunte Breccia of the Ries: Continuous deposits of large
 impact craters. *Reviews of Geophysics*, 21(8), pp.1667–1725.
- Hubbard, N.J. et al., 1971. The composition and derivation of Apollo 12 soils. *Earth and Planetary Science Letters*, 10(3), pp.341–350.
- Jourdan, F., Renne, P.R. & Reimold, W.U., 2009. An appraisal of the ages of terrestrial impact structures.
 Earth and Planetary Science Letters, 286(1), pp.1–13.
- Jourdan, F., 2012. The 40Ar/39Ar dating technique applied to planetary sciences and terrestrial impacts.
 Australian Journal of Earth Sciences, 59(2), pp.199-224.
- 601 Korotev, R.L., 1994. Compositional variation in Apollo 16 impact-melt breccias and inferences for the
- 602 geology and bombardment history of the Central Highlands of the Moon. *Geochimica et*
- 603 *Cosmochimica Acta*, 58(18), pp.3931–3969.
- Laneuville, M. et al., 2013. Asymmetric thermal evolution of the Moon. *Journal of Geophysical Research: Planets*, 118(7), pp.1435–1452.
- Liu, T. et al., 2019. Regolith mixing by impacts : Lateral diffusion of basin melt. *Icarus*, 321(0019-1035),
 pp.691–704.
- McKinnon, W.B. et al., 1997. Cratering on Venus: Models and Observations. In *Venus II: Geology, Geophysics, Atmosphere, and Solar Wind Environment*. p. 969.
- 610 Melosh, H.J., 1989. *Impact cratering: A geologic process*, New York: Oxford University.
- 611 Michael, G., Basilevsky, A. & Neukum, G., 2018. On the history of the early meteoritic bombardment of
- 612 the Moon: Was there a terminal lunar cataclysm? *Icarus*, 302, pp.80–103.
- 613 Miljković, K. et al., 2016. Subsurface morphology and scaling of lunar impact basins. *Journal of*
- 614 *Geophysical Research: Planets*, 121(9), pp.1695–1712.

- Morbidelli, A. et al., 2012. A sawtooth-like timeline for the first billion years of lunar bombardment. *Earth and Planetary Science Letters*, 355–356, pp.144–151.
- 617 Naumenko-Dèzes, M.O., Nägler, T.F., Mezger, K. and Villa, I.M., 2018. Constraining the 40K decay
- 618 constant with 87Rb-87Sr-40K-40Ca chronometer intercomparison. Geochimica et cosmochimica
- 619 acta, 220, pp.235-247.
- Neukum, G., 1983. Meteoritenbombardement und Datierung Planetarer Oberflaechen. *Habilitation Dissertation for Faculty Membership, Univ. of Munich*, pp.1–186.
- Neumann, G.A. et al., 2015. Lunar impact basins revealed by Gravity Recovery and Interior Laboratory
 measurements. *Science Advances*, 1(9).
- 624 Niihara, T., Beard, S.P., Swindle, T.D., Schaffer, L.A., Miyamoto, H. and Kring, D.A., 2019. Evidence
- 625 for multiple 4.0–3.7 Ga impact events within the Apollo 16 collection. Meteoritics & Planetary
- 626 Science, 54(4), pp.675-698.Norman, M.D. et al., 2016. Crystal accumulation in a 4.2 Ga lunar

627 impact melt. *Geochimica et Cosmochimica Acta*, 172, pp.410–429.

- 628 Norman, M.D., Duncan, R.A. & Huard, J.J., 2006. Identifying impact events within the lunar cataclysm
- from 40Ar–39Ar ages and compositions of Apollo 16 impact melt rocks. *Geochimica et*
- 630 *Cosmochimica Acta*, 70(24), pp.6032–6049.
- 631 Norman, M.D., Duncan, R.A. & Huard, J.J., 2010. Imbrium provenance for the Apollo 16 Descartes
- 632 terrain: Argon ages and geochemistry of lunar breccias 67016 and 67455. *Geochimica et*
- 633 *Cosmochimica Acta*, 74(2), pp.763–783.
- Norman, M.D. & Nemchin, A.A., 2014. A 4.2 billion year old impact basin on the Moon: U–Pb dating of
 zirconolite and apatite in lunar melt rock 67955. *Earth and Planetary Science Letters*, 388, pp.387–
 398.
- 637 Oberbeck, V.R. et al., 1975. On the origin of the lunar smooth plains. *The Moon*, 12(1), pp.19–54.

- 638 Orgel, C. et al., 2018. Ancient Bombardment of the Inner Solar System: Reinvestigation of the
- 639 "Fingerprints" of Different Impactor Populations on the Lunar Surface. *Journal of Geophysical*640 *Research: Planets*, 123(3), pp.748–762.
- 641 Petro, N.E. & Pieters, C.M., 2006. Modeling the provenance of the Apollo 16 regolith. *Journal of*
- 642 *Geophysical Research*, 111(E9), p.E09005.
- Potter, R.W.K., Collins, G.S., et al., 2012. Constraining the size of the South Pole-Aitken basin impact. *Icarus*, 220(2), pp.730–743.
- 645 Potter, R.W.K., Kring, D.A., et al., 2012. Estimating transient crater size using the crustal annular bulge:
- Insights from numerical modeling of lunar basin-scale impacts. *Geophysical Research Letters*,
 39(18), pp.1–5.
- Potts, L. V & von Frese, R.R.B., 2003. Comprehensive mass modeling of the Moon from spectrally
 correlated free-air and terrain gravity data. *Journal of Geophysical Research*, 108(E4), p.5024.
- 650 Riedel, C. et al., 2018. A New Tool to Account for Crater Obliteration Effects in Crater Size-Frequency
- Distribution Measurements. *Earth and Space Science*, 5(6), pp.258–267.
- Robinson, M.S. et al., 2010. Lunar reconnaissance orbiter camera (LROC) instrument overview. *Space Science Reviews*, 150(1–4), pp.81–124.
- Rotenberg, E., Davis, D.W., Amelin, Y., Ghosh, S. and Bergquist, B.A., 2012. Determination of the
- decay-constant of 87Rb by laboratory accumulation of 87Sr. Geochimica et Cosmochimica Acta,
 85, pp.41-57.
- Ryder, G., Spudis, P.D. & Taylor, G.J., 1989. The case for planetary sample return missions: Origin and
 evolution of the Moon and its environment. *Eos, Transactions American Geophysical Union*,
- 659 70(47), pp.1495–1509.

- Schärer, U., 1998. Dating of Impact Events. In D. Benest & C. Froeschlé, eds. *Impacts on Earth*. Berlin,
 Heidelberg: Springer Berlin Heidelberg, pp. 157–183.
- Schultz, P.H. & Gault, D.E., 1985. Clustered impacts: Experiments and implications. *Journal of Geophysical Research*, 90(B5), p.3701.
- 664 Shervais, J.W., Taylor, L.A. & Lindstrom, M.M., 1985. Apollo 14 Mare basalts: Petrology and
- geochemistry of clasts from Consortium Breccia 14321. *Journal of Geophysical Research: Solid Earth*, 90(S02), pp.C375–C395.
- Shuvalov, V., 2011. Ejecta deposition after oblique impacts : An influence of impact scale. *Meteoritics & Planetary Science*, 1718(11), pp.1713–1718.
- 669 Snape, J.F. et al., 2016. Phosphate ages in Apollo 14 breccias: Resolving multiple impact events with
- high precision U–Pb SIMS analyses. *Geochimica et Cosmochimica Acta*, 174, pp.13–29.
- 671 Spudis, P.D., Wilhelms, D.E. & Robinson, M.S., 2011. The Sculptured Hills of the Taurus Highlands:
- 672 Implications for the relative age of Serenitatis, basin chronologies and the cratering history of the
- 673 Moon. Journal of Geophysical Research, 116(12), p.E00H03.
- Stöffler, D., 2006. Cratering history and lunar chronology. *Reviews in Mineralogy and Geochemistry*,
 60(1), pp.519–596.
- 676 Tera, F., Papanastassiou, D.A. & Wasserburg, G.J., 1974. Isotopic evidence for a terminal lunar
 677 cataclysm. *Earth and Planetary Science Letters*, 22(1), pp.1–21.
- Wieczorek, M.A. & Phillips, R.J., 1999. Lunar Multiring Basins and the Cratering Process. *Icarus*, 139,
 pp.246–259.
- 680 Zellner, N.E.B., 2017. Cataclysm no more: New views on the timing and delivery of lunar impactors.
- 681 *Origins of Life and Evolution of Biospheres*, 47(3), pp.261–280.

- Zhu, M.-H. et al., 2019. Reconstructing the late-accretion history of the Moon. *Nature*, 571(7764),
 pp.226–229.
- 684 Zhu, M.-H. & Wünnemann, K., 2013. Modeling of Meteorite Impact-Induced Secondary Mass Wasting
- 685 {\mdash} Case Study by Means of the Bunte Breccia Ejecta Blanket at Ries Crater, Germany. In
- 686 *Lunar and Planetary Science Conference*. Lunar and Planetary Science Conference. p. 1921.
- Zhu, M.-H., Wünnemann, K. & Artemieva, N., 2017. Effects of Moon's thermal state on the impact basin
 ejecta distribution. *Geophysical Research Letters*, 44(22), p.11,211-292,300.

689