- Lunar megaregolith mixing by impacts: Evaluation of the nonmare component of mare soils
- 3 Tiantian Liu^{1,3}, Greg Michael², Wilhelm Zuschneid², Kai Wünnemann^{2,3}, Jürgen Oberst^{1,4}
- 4 ¹ Institute of Geodesy and Geoinformation Science, Technische Universität Berlin, 10623 Berlin,
- 5 Germany (tiantian.liu@tu-berlin.de)
- 6 ² Freie Universität Berlin, Malteserstr., 74-100, Haus D, 12249 Berlin, Germany
- ³ Museum f
 ür Naturkunde, Leibniz Institute for Evolution and Biodiversity Science, 10115 Berlin,
 Germany
- ⁹ ⁴ Institute of Planetary Research, German Aerospace Center (DLR), 12489 Berlin, Germany
- 10

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12 Moon, surface; Regoliths; Impact processes; Cratering

13 Abstract

14 The source of the non-mare component in the lunar mare soil samples is still an open question. A spatially-resolved numerical model tracing the diffusion of non-mare material was developed by 15 means of the Monte Carlo method to study this issue in more detail. The vertically- and laterally-16 17 transported non-mare components are recorded separately to assess their concentration on the surface. We find a general higher efficiency of lateral transport than of vertical transport, but the 18 19 opposite may occur within small zones of interest. The overall (background) distribution of non-20 mare component is estimated by averaging out regional variations of material composition caused 21 by local impact mixing. We find that almost all the mare surface is mixed with non-mare material. 22 If most of the mare regions have filled basaltic material about 500 m in thickness since the 23 formation of basins, the average non-mare fraction in the top 5 m is about 0.1. The abundance of 24 the non-mare component decreases with increasing distance from the mare-highland boundary, but 25 the slope of the distance-dependence is shallower within ~100 km of the boundary than further 26 away.

27 By comparing the background composition derived from our model with the geochemical analysis and geological interpretation of the lunar mare soil samples, we infer the most plausible geologic 28 29 processes that have significantly altered the material composition at the sampling sites: for the 30 Apollo 15 and 17 mare soil samples, the large fraction of non-mare material is likely to have 31 resulted from the downslope slumping or lateral transport of the nearby massifs. The Apollo 12 32 sampling site that is located interior domains of Oceanus Procellarum has a component of 33 Copernicus ejecta. A mixing of both Copernicus ejecta and excavated local underlying material by high-velocity ejecta has altered the composition at the surface. The non-mare material contained 34 35 in the Apollo 11 and Luna 24 mare soil samples could have been built up gradually by both long-36 time lateral and vertical mixing. The mare deposit at the Luna 16 landing site is likely to be

37 relatively thin resulting in the abundant vertically transported non-mare component. Since the

- 38 history of the lunar volcanic eruptions deposit thicknesses, flux curves and onset of activity -
- 39 have not been well constrained, we make simple estimates with the first-order accuracy. In addition,
- 40 it was found that the vertically transported non-mare abundance in the top surface is influenced by
- 41 both the cessation time of the major mare fillings and the total amount of mare deposit; the
- laterally-transported non-mare abundance in the top surface is mainly dependent on the cessation
 time of the major mare fillings. The peak time of eruption would not change the abundance of both
- 44 the laterally and vertically transported non-mare component.

45 **1. Introduction**

46 **1.1. Non-mare component in the lunar mare soil samples**

47 The returned lunar mare soil samples consist not only of local mare basalts but also of highland anorthosites (Wood, 1970). We use the term *non-mare* to refer to the components that are clearly 48 49 not of volcanic mare origin but for which the relationship to the highlands is not clear or firmly 50 established (e.g., Warren & Wasson 1977). The amount of non-mare material in Apollo 11, 12, 51 and Luna 16 mare soil samples is about 20% on average; some of the Apollo 12 soil samples have 52 more than 70% non-mare material (Rhodes, 1977). In addition, data from the orbital X-ray 53 experiments (the measurement depth for the X-ray data is about 20 µm; Lucey et al. 2006) 54 indicated that, for most mare surfaces, the chemical compositions, such as Al/Si concentration 55 ratios, are more comparable with that obtained from the analyzed mare soils than with that of the 56 local basalts. Thus it was suggested that the mixture of non-mare component in mare soils was not 57 only restricted to the sampling sites that are near the mare margins, but may be a mare-wide 58 phenomenon (Trombka et al., 1974; Rhodes, 1977; Prettyman et al., 2006).

59 The source of the non-mare component contained in the mare samples has been debated since the Apollo epoch (Rhodes, 1977; Laul and Papike, 1980; Simon, Papike and Laul, 1982; Farrand, 60 1988; Fischer and Pieters, 1995; Li and Mustard, 2000; Huang et al., 2017). Two hypotheses have 61 62 been proposed: It could result mainly from the mixing of material transported laterally by impacts 63 from the distant highland regions (De Hon, 1974; Head, 1982; Budney and Lucey, 1998; Evans et 64 al., 2016). It could also primarily be result from the mixing with local material that originally 65 occurred beneath the mare deposit and was excavated by impact events (Arvidson et al., 1975; 66 Rhodes, 1977; Hörz, 1978; Laul and Papike, 1980; Fischer and Pieters, 1995). Studies of both 67 remote sensing data and collected samples have been used to evaluate the efficiency of lateral and 68 vertical transport. The material mixing process across mare-highland contacts driven by a great 69 number of impact events results in a mixing zone inside mare regions where the surface material 70 has been mixed with a significant amount of non-mare component displaying relative high albedo 71 in optical images. The observed ~4-5 km wide mixing zone across the mare-highland boundary is relatively narrow, indicating inefficient lateral mixing by impact events, while the abundance of 72 73 highland material in the soil samples far from this boundary suggests more efficient lateral 74 transport.

Huang et al. (2017) implemented a regolith material transport model into the Cratered Terrain
 Evolution Model (CTEM) to investigate the mixing of material across the mare-highland boundary.

- 77 The effects of local and distal ejecta were systematically studied and their relatively importance
- 78 was quantified. It was found that both local and distal material transports were important to the
- 79 material diffusion across the mare-highland boundary. In addition, the superposition of crater rays
- 80 was proposed to be a likely cause for the elevated non-mare abundance in some mare soil samples.
- For example, the Apollo 12 soil samples that had a significantly higher concentration of non-mare 81
- 82 component were suggested to be influenced by the emplacement of the Copernicus Crater ray
- 83 material.

84 While the research of Huang et al. (2017) has improved our understanding of non-mare diffusion

- 85 over the lunar surface, there remain several issues to resolve. First, Huang took the mare deposit as an intact layer on top of a highland layer. But the mare flooding occurred intermittently. Earlier 86 transported non-mare material could become buried decreasing its abundance on the surface. 87
- 88 Furthermore, the non-mare components beneath the mare deposit were not traced. Therefore, the
- 89 relative magnitude of lateral versus vertical transport could not be evaluated.
- 90 In this study, we aim to develop a numerical model to trace the diffusion of non-mare material 91 with the cumulative effect of impact mixing and intermittent filling of mare material. Questions

92 that need resolving are: (a) Was the non-mare component transported over large distances on to 93 the mare surfaces following the emplacement of mare material? Or (b) did a significant fraction of

- 94 it originate beneath the mare surface? (c) How do spatial and temporal variations in mare
- deposition affect the expected surface abundances of non-mare materials today?
- 95
- 96 **1.2. Mare volcanic flooding**

Besides the impact mixing process, the volcanic eruptions are a key process to model. Previous 97

- 98 work, nevertheless, has shown the difficulty to establish a well-constrained eruption flux (e.g.,
- 99 Head & Wilson 1992; Wilson & Head 2017; Hiesinger, Head, et al., 2011).

100 In order to constrain the flux of mare volcanism, the thicknesses of individual flows are required,

- 101 which are found on the basis of their morphometric characteristics neglecting the problem that 102 most flows are covered by others. Such measurements, nevertheless, are complicated due to the
- 103 limited availability of suitable data necessary for the recognition of flow fronts (i.e. high-resolution
- 104 topography and low-sun images), and the obliteration of flow fronts caused by impact cratering.

105 The onset and duration of mare volcanism are not well understood (summarized by Niquist et al. 2001). The returned samples revealed that mare volcanism was active between \sim 3.9 and 3.1 Ga 106 (Head, 1976; Nyquist and Shih, 1992). The ages of some basaltic clasts in older breccias point to 107 108 an onset of mare volcanism prior to 3.9 Ga (Ryder and Spudis, 1980), perhaps as early as 4.2-4.3 109 Ga (Taylor et al., 1983; Dasch et al., 1987; Nyquist, Bogard and Chi-Yu, 2001). Dark halo craters 110 found in remote sensing data may support the presence of the older volcanism. These craters are 111 interpreted to be impacts into basaltic deposits that are now buried underneath a veneer of basin 112 ejecta. These underlying basalts might be among the oldest mare material on the Moon (Schultz and Spudis, 1979; Hawke and Bell, 1982). In addition, crater counts on mare surfaces suggest that 113 114 some mare volcanism erupted as late as $\sim 1-2$ Ga ago (Hiesinger, Head, et al., 2011), which is 115 much younger than that was determined in the collected samples (~3 Ga).

116 The total thickness/volume of the volcanic deposits is an important quantity in defining the extent 117 of mare volcanism. A variety of techniques have been applied to estimate the total volume, but no 118 consistent results have been obtained. Crater-geometry techniques using pristine crater 119 morphometric relationships and the diameter of partially to almost entirely flooded craters yield 120 values of mare thickness up to 2 km, ranging 200-400 m on average (De Hon, 1974, 1979; De Hon and Waskom, 1976; Hörz et al., 1991). Hörz (1978) further refined the crater-geometry 121 122 technique by considering the average degradational state of impact craters, and argued that the 123 mare thicknesses estimated by De Hon and coworkers should be decreased by a factor of ~2. Other 124 constraints are derived from "mascons" (i.e. the positive gravity anomalies observed over many of 125 the basins filled with mare material). When the excess mass required to produce the observed 126 gravity anomaly is assumed to be entirely mare filling, estimates for mare thickness of 1–2 km are obtained (Phillips et al., 1972; Phillips and Lambeck, 1980; Solomon and Head, 1980). However, 127 128 these are minimum estimates, because it is not known whether there is additional mare fill below 129 the equilibrium gravitational surface used in the modeling. In addition, there would be at most ten 130 times larger estimates of the total volume if one assumes a specific geometry for the original impact 131 basin cavities, modeled after the Orientale basin, and assumes similar cavity geometries for the all

132 basins (Head, 1977).

133 Since higher-quality image data are now available, more studies have been done aiming to estimate

134 the basaltic thickness. For example, by using recent high-resolution topographic data, Du et al

135 (2019) revisited the previous studies of De Hon and Hörz. They identified 661 buried craters over

136 the lunar surface and concluded that the basaltic thicknesses vary from 33-455 m with a medium

value of 105 m. Using the high-resolution gravity and topography data, Gong *et al.* (2016)

estimated the total thickness of basalts on the nearside hemisphere, which yield an average thickness of 740 m. In addition, a recent study from Needham & Kring (2017) summarized the

thickness estimate of Williams & Zuber (1998) and some other recent estimates to calculate the

141 volume of mare deposit. They derived a thinner average mare thickness of ~250 m.

Both the duration of emplacement and the thickness of mare deposits are not precisely known making it difficult to estimate the eruption flux. Our goal is thus to estimate the diffusion pattern of the non-mare material from different sources. In section 2, the main aspects of the previously developed model are outlined and the integration of the lunar volcanic history into it is described.

146 In section 3 and 4, two cases including the rapid and continuous mare fillings are present. First, 147 and more simply (section 3), we take the volcanic material to be emplaced instantaneously as single layers. Secondly (section 4), we investigate a probably more realistic scenario, where the 148 mare material slowly and continuously fills the basins. To investigate the diffusion features of the 149 150 lateral and vertical mixing, the abundances of the laterally and vertically transported components 151 are traced separately, and the relative rates of lateral versus vertical transport are evaluated. In 152 section 5, the overall distribution of non-mare material is presented. Based on the model-predicted 153 non-mare abundance, the origin of the non-mare component in the mare samples is discussed in

154 section 6.

155 **2. Model**

156 In our previous work (Liu *et al.*, 2020), a spatially-resolved numerical model was developed to 157 investigate the diffusion of impact melts through impact mixing. Using the Monte Carlo method,

158 a sequence of impact events was generated, where the occurrence time and the size of impact

- 159 craters were calculated on the basis of the chronology and production functions of Neukum (1983).
- 160 In the model, impact events of different sizes occurred in sequence. The occurrence of each impact
- 161 event would exhume some local (target) material. The excavation depth, d_{exc} , was approximated
- 162 to one third of the transient crater depth. The volume of the excavated materials was estimated to
- 163 be one-third of a disc with d_{exc} in thickness and the transient crater size in diameter. The excavated
- 164 material would be emplaced outside the crater cavity as ejecta layers, the thickness of which 165 decreases with distance from crater center. In addition, the high-velocity ejecta would mix with
- 166 local material while emplacing. The degree of mixing was quantified using a mixing ratio of local
- material to ejecta, $\mu = 0.0183r^{0.87}$ (Oberbeck *et al.* 1975). When μ is larger than 5.0, the value of μ
- 168 was modified by roughly half: $\mu' = \mu/2 + 2.5$ (Petro & Pieters 2006).

169 The evolving distribution of material composition on the surface and with depth caused by each 170 impact event was recorded by the uniformly distributed one million points on a sphere with the 171 Moon's radius (1737.4 km). Each point was related to a certain area of $\sim 5 \times 5$ km². In addition, the 172 increasing number of deposition layers as the simulation progresses made it necessary to 173 periodically simplify the layer sequence to one: 0.2×1.9^{a} m, where a = 0, 1, ..., 20. Materials 174 within set layers would be mixed together resulting in a more average composition. The thinner 175 layers close to the surface were chosen to preserve a fine resolution of material composition in the 176 near surface. A more detailed explanation of the model is given in Liu et al. (2020).

In this study, we extend the capabilities of the model and take into account the volcanic basin infilling forming the lunar mare regions. The minimum crater diameter considered is 5 km. In the following section, the models used to simulate the ejecta distribution and the mare fillings are presented in detail. In all model runs of sections 3 and 4, the simulated cratering is identical in timing, position, and crater sizes, so all changes can be attributed to the varying parameters of the lava emplacement process.

183 **2.1. Ejecta distribution**

184 During the formation of an impact crater, the majority of the excavated material is deposited near 185 the crater rim forming a continuous blanket (i.e. the proximal ejecta which is generally distributed within 2-3 radii from the crater center). Further away from the crater, the continuous ejecta is 186 187 replaced by the patchily distributed distal ejecta, usually in the form of bright rays and grouped 188 secondary craters. Although the volume of distal ejecta is not large (~25% deposits further than 189 five radii away from the transient crater rim; Liu et al., 2019, 2020), previous studies have shown 190 its significance to the material composition of mare regions. For example, the study of Huang et 191 al. (2017) focused on the diffusion of non-mare material across the mare-highland contact. It 192 showed that if only the proximal ejecta contributed to the soil composition, the mare surface would 193 be clearly different from what is actually seen on the Moon, and the mare-highland mixing zone

194 would be rather narrower than observed.

For proximal ejecta, the thickness distribution with distance from crater center (*r*) conforms to a power law, typically: $\delta(r) = Ar^{-3}$ (A is constant but varies for differently-sized craters, Melosh 197 1989). However, so far, no systematic research has been performed to obtain a scaling law for the thickness distribution of distal ejecta. In addition, distal ejecta may occur much further out than where we see bright rays and secondary craters. This fraction of the distal ejecta emplaced with high velocity and fine grain size is difficult to distinguish from the background surface.

201 Places superposed by the patchy distal ejecta locally have thicker transported material than their 202 surrounding background. This clustered material would be redistributed by the subsequent impact 203 events. As time passes, the clustered material diffuses over the larger area decreasing the thickness. 204 That is, the patchy distal ejecta would fade into the background with continuous impact mixing, 205 and the thickness distribution of the patchy ejecta material becomes more uniform. The average effect on distal ejecta looks similar to a uniform distribution. To estimate the distribution of distal 206 207 ejecta, we therefore assume that the distal ejecta is also continuously distributed, and the thickness 208 is calculated based on the extrapolated power law mentioned above. Although the distal ejecta of 209 recently-formed larger impact craters, such as Copernicus crater, could still remain patchy, their 210 regional influence could be easily recognized from their background. If it occurs over the target 211 regions, such as sampling sites, the composition of regional surface could be significantly different

- from its background.
- 213 The power-law distribution of ejecta thickness leads to a thinner deposit at locations further from 214 the impact center. The farthest ejecta that the model would trace should thus have the minimum 215 deposit thickness (h_{\min}) , based on which the ejecta coverage range of each impact event can be calculated. All the excavated material would be distributed within the coverage range to maintain 216 217 the total volume. The value of h_{\min} is taken to be 0.01 m in this study. Our simulations showed that 218 a smaller value for the minimum thickness, which would relate to a larger traced range of ejecta, 219 would lead to a similar composition of surface material since the volume of ejecta at greater ranges 220 becomes too small to significantly augment the non-mare component. For example, when the value 221 of h_{\min} is ten times smaller (i.e., 0.001 m), the estimated composition of surface material is comparable with that using h_{\min} of 0.01 m, and the difference of non-mare fraction in the near 222 223 surface is generally smaller than ~0.001. On the other hand, a larger value of h_{\min} would relate to 224 a smaller range of ejecta deposit. The simulation results then display significantly less mixing of the non-mare component over the mare regions. For example, when the value of h_{\min} is ten times 225 larger (i.e., 0.1 m), more than half of the mare surface, especially the central part of mare regions, 226 227 has no mixing of non-mare material. This is clearly not in agreement with the observed composition of the lunar surface, so that we exclude this value. The simulation results with the 228 229 varying values of h_{\min} are presented in the supplementary file.
- 230 Craters smaller than 5 km are not involved in the model. The smaller impact events have high
- 231 frequency. They have well mixed the material in the top surface producing a fragmental regolith
- 232 layer. However, since they hardly excavate fresh material from the beneath maria, they would not
- significantly change the abundance of the material components in the top surface. Given that the
- typical thickness of regolith layer over the maria is about 5 m (McKay et al., 1991; Fa and Jin,
- 235 2010; Fa et al., 2014), in the following discussions, we present the average abundance of non-mare

component in the top 5 m. These results can be regarded as the average composition of the regolith

- where the material has been frequently mixed by smaller impact events and hence the material is
- homogeneously distributed.

239 **2.2. Mare volcanic filling**

240 As indicated by the diverse basalt ages over the mare regions (e.g., 30 units over Mare Imbrium; 241 Hiesinger, Head, et al., 2011), the lunar basins were filled with multiple flow units. The 242 intermittent mare flooding would decrease the distribution of early-transported non-mare 243 component on the surface. Therefore, as well as the impact mixing process, the sequential mare volcanic filling is also described by the model. The way to simulate both processes is illustrated in 244 245 Figure 1a. Before the flooding by volcanic material, both the highland and mare region surfaces 246 are non-mare material. To better trace the vertical and lateral mixing of the non-mare component, 247 the non-mare component originating from the highland regions is tracked as "laterally" transported 248 non-mare material, and that from beneath the mare infill as "vertically" transported non-mare 249 material. The labels "laterally" and "vertically" are used to specify the spatial origin, but not 250 necessarily a different mineralogical composition.

251 At the time of a_1 (Figure 1 a1), an eruption occurred, burying the surface of the mare region with 252 volcanic material m_1 . Subsequent impact mixing redistributes mare material regionally, and 253 excavated some non-mare materials from beneath the mare layer (i.e. vertically transported non-254 mare component). Some highland material is transported from the highlands to the mare region 255 (indicated by the arrow in Figure 1 a2). At the time of a_3 (Figure 1 a3), another volcanic eruption 256 occurs, the generated mare material deposits m_2 on the top of volcanic deposit m_1 . The impact mixing keeps laterally transporting the non-mare material to the mare regions. Some of the early-257 258 formed volcanic deposit m_1 are re-excavated to the surface (the open arrow in Figure 1 a4); some 259 of the early-transported highland could be delivered to the surface (the thick closed arrow in Figure 260 1 a4); some mare material might be transported to the highlands (the thin closed arrow in Figure 1 a4). On the lunar surface, the long-term impact mixing excavates various amounts of both mare 261 262 and non-mare material to the surface, resulting in the mixing of mare/non-mare components to

263 different degrees in the present day.





Figure 1 (a1-a4) A schematic of the filling of volcanic material. (b) The thickness distribution of mare
material (after De Hon 1974; De Hon 1979; De Hon & Waskom 1976; Hörz et al. 1991) over the Oceanus
Procellarum (P), and the Mare Frigoris (Fr), Fecunditatis (Fe), Humorum (H), Imbrium (I), Serenitatis (S),
Tranquillitatis (T), Nectaris (N) and Crisium (C). The red stars indicate the locations of Apollo ("A") and
Luna ("L") sampling sites.

- 270 In this study, the volcanic filling of the larger mare regions on the lunar near side are simulated,
- 271 including Oceanus Procellarum (P), and the Mare Frigoris (Fr), Fecunditatis (Fe), Humorum (H),
- 272 Imbrium (I), Serenitatis (S), Tranquillitatis (T), Nectaris (N) and Crisium (C). Other regions are
- taken to consist of non-mare highland material. The pre-existing topography that is covered over
- by successive mare layers is not considered beyond what is shown in Figure 1b.
- The eruption flux has not been well constrained. We approximate *the peak time of eruption, the thickness of mare deposits*, and the *cessation time of the major eruptions* for modeling.
- 277 *Peak time of eruption:* The qualitative eruption features of volcanic history of the Moon were 278 pictured previously (Head and Wilson, 1992, 2017; Wilson and Head, 2017). Head & Wilson 279 (1992) compared the flux of volcanic filling derived from different approaches (i.e. the volume of 280 mare material emplaced on the surface as a function of time), all of which pictured a similar trend 281 of eruption flux from high to low but with different peak times. Based on the returned samples, a peak prior to ~3.5 Ga was obtained. The recent study from Needham and Kring (2017) that 282 283 combined the estimated of mare thickness using different methods also indicted the volume peak 284 at ~3.5 Ga. Using the age of the surface exposure of mare units, the flux curve was shifted toward 285 younger ages with the peak at ~3.2 Ga because the older mare material was covered by the uppermost young deposits. Investigations of the stratigraphy of mare material showed the 286 287 volumetric significance of early mare deposits yielding a flux curve with a peak at older ages of ~ 288 3.8 Ga. The peak of 3.8 Ga is taken for simulations, and the influence of the younger peak would
- be discussed in section 4.3.
- 290 Thickness of mare deposits: There is no direct measurement of the thickness of lava deposits over
- the mare regions. The thickness distribution of volcanic deposits was systematically studied using
- 292 crater-geometry techniques pioneered by De Hon. This approach relies on impact craters flooded
- 293 by mare material. By using the ideal crater shapes, one can calculate the height of the fresh crater

294 rim. The thickness of mare deposits is then calculated by subtracting the present height from the 295 estimated initial rim height. This approach has been used all observable flooded craters, and 296 isopach maps of the thickness of volcanic filling (Figure 1b) were constructed (De Hon 1974; De 297 Hon 1979: De Hon & Waskom 1976; Hörz et al. 1991). We use this result as an estimate of the 298 total volcanic deposit over the mare regions (H_{DeHon}). Note that, in Figure 1b, the contour of a 299 certain color represents a range of mare thickness. The upper limit (instead of the median value) 300 of each contour is taken for simulations. In this way, the major mare areas are filled with mare 301 material of 500 m in thickness, which is about the median value of the previous estimates (~100 302 $m - \sim 1$ km). The effect of the varying mare thickness on the non-mare distribution is discussed in 303 section 4.3. For each mare region, the areas showing the thickest mare deposit are flooded first. 304 As time goes on, mare material accumulates according to the eruption flux. The surrounding areas 305 of thinner mare deposit are subsequently flooded. After the deposit of the youngest volcanic 306 eruption, the thickness of volcanic deposit corresponds to H_{DeHon} . A time step of 0.01 Ga is used 307 to simulate such continuous eruption. At each step, the mare filling occurred within a certain 308 contour region is taken to be uniformly emplaced.

309 For mare deposits with a thickness of 500 m, a 5 km diameter crater would begin to tap the

310 underlying non-mare material, the evidence of which could be found through remote sensing data.

311 Such craters were proposed to possess a ring of low FeO ejecta, where the FeO abundance could

be calculated using UV-VIS data from the Clementine mission. For example, Thomson et al. (2009)

313 examined 23 craters on Mare Imbrium >10 km in diameter. Six craters were suggested to penetrate

the mare deposit indicated by their surrounding halos of low FeO ejecta. However, due to the small

volume of the excavated mare component, the ejecta of a crater that just begins to tap the sub-mare

316 material hardly displays obvious low FeO evidence on the surface.

317 *Cessation time of the major eruptions:* The mare regions are the result of volcanic infilling after 318 the formation of the giant basins. The absolute age dating of specific rock samples shows that 319 significant periods of time elapsed between basin formation and extrusion of mare material into 320 the basin, but both the basin ages and the onsets of volcanic infilling are only roughly constrained 321 (Wilhelms, 1987; Head and Wilson, 1992; Hiesinger et al., 2011; Whitten and Head, 2015a, 322 2015b). At one extreme, volcanic eruptions may be assumed to initiate soon after the formation of 323 the respective basin. On the basis of superposed crater density, Wilhelms (1987) documented the 324 relative ages of the lunar basins. Oceanus Procellarum, Frigoris, Fecunditatis, and Tranquilitatis 325 basins were distinguished to be pre-Nectarian, and their formation time was suggested to be around 326 4.1 Ga. Humorum and Nectaris basins were classified into the Nectarian Period, with formation 327 times around 3.9 Ga. The age of Serenitatis, Crisium and Imbrium could be determined from the 328 returned lunar samples. Although their radioisotopic ages are still debated, the generally accepted 329 formation times of both Crisium and Serenitatis basin are around 3.9 Ga, with the Imbrium basin

about 0.05 Ga younger (Stöffler, 2006).

Each volcanic eruption in the model is taken to cover the entire surface within the contour line.

332 This simplified approach can be considered consistent with the early mare filling events, but not

333 with the last flows due to their smaller volume. Head & Wilson (1992) estimated the volume of

mare material during different periods, and showed that the volume of Upper Imbrian deposits was

335 about thirty times greater than those later than of the Eratosthenian Period. In addition, the total 336 coverage area of these younger deposits was six times smaller than that of the earlier deposits. 337 With the aim to investigate the general features of non-mare diffusion, the irregularly distributed 338 young mare deposits are not taken into account in this model. However, the cessation time of the 339 mare filling that could cover the entire mare surface (i.e. the cessation time of major mare flooding) 340 is difficult to estimate. The age of the youngest flows help to constrain the infill age. To determine the rough cessation time of the main filling (t_{end}) among different mare regions, we assess the 341 342 fraction of area of mare deposits younger than a certain age relative to the whole mare region. We 343 take the value of the fraction to be $\frac{1}{2}$, that is, the age t_{end} marks the age when only half of the 344 exposed area would be covered by younger flows, where the age of mare units was estimated by 345 Hiesinger et al. (2011). For Oceanus Procellarum, Mare Frigoris, Humorum, Nectaris, Fecunditatis, Tranquillitatis, Serenitatis, Crisium, and Imbrium, tend is calculated to be 3.4, 3.5, 3.5, 3.6, 3.6, 3.6, 346 347 3.5, 3.3, and 3.3 Ga, respectively. The influence of the varying t_{end} value is discussed in section 348 4.3.

349 **3. Rapid basin filling**

In the first case, the volcanic material is taken to be emplaced instantaneously as a single layer at t_{end} over the different mare regions. The thickness of mare material over all the mare regions is taken to be 500 m, the mare thickness in most mare regions (Figure 1b).

353

3.1. Mixing of the laterally transported non-mare component

354 The excavated non-mare component is entrained in the ejecta of impact craters that occur on the 355 highlands, a part of it travels across the mare-highland boundary, and then falls on the mare regions 356 (i.e. laterally transported non-mare component). As time goes on, the laterally transported non-357 mare component on the mare surface builds up. But the early-transported non-mare material could 358 become buried, being covered by a rapid filling of volcanic material. The size and frequency of 359 later-formed impact events would then determine the abundance of the laterally transported non-360 mare component of the mare surface. Only impacts occurring near the mare-highland boundary 361 could bring a significant amount of laterally transported non-mare material to the mare regions; 362 the ejecta of those occurring far away hardly reaches the mare regions except from some really 363 large craters. Especially at the central areas of the mare regions, little mixing with laterally 364 transported non-mare material occurs.

365 Figure 2a represents the spatial distribution of the laterally transported non-mare component in the 366 near surface (the top 5 m). It shows that in almost all the mare regions some laterally transported 367 non-mare component is present. Near the margin of the mare regions, laterally transported non-368 mare material is more abundant than in the inner part that can only be reached by the ejecta of 369 larger impact events. In addition, because of the longer exposure time after the volcanic flooding, 370 the mare regions with the older t_{end} , such as Mare Tranquillitatis and Nectaris, accumulate more 371 laterally transported non-mare material. But the regional context of the mare region also influences 372 the amount of the accumulated laterally transported non-mare component. A mare region 373 surrounded by highland regions would have a higher accumulation rate. For example, although the 374 volcanic cessation time of Mare Crisium is 0.3 Ga later than that of Mare Tranquillitatis and Fecunditatis, the surface of those regions displays a comparable abundance of laterally transported non-mare component.

377 **3.2. Mixing of the vertically transported non-mare component**

378 Impact events occurring within the mare regions can penetrate through the mare deposit and 379 excavate underlying non-mare material. These large-scale impact events can exhume a great 380 amount of non-mare material to the near surface (i.e. vertically transported non-mare component), 381 forming non-mare-enriched zones. The smaller impact events with shallower excavation depth do 382 not contribute a significant amount of non-mare material to the near surface, but locally garden the 383 surface material diluting the regional concentration of the vertically transported non-mare 384 component.

Figure 2b presents the spatial distribution of the vertically transported non-mare component in the near surface (the top 5 m). We see that vertically transported non-mare is also widely mixed in the near surface but is less abundant, as indicated by the bluish color in Figure 2b. The distribution is relatively heterogeneous marked by some striking non-mare-enriched zones shown in red. These zones are well-preserved indicating their slow fading rate as a result of the lower impact cratering

390 rate during the recent period (Neukum 1983).

391 **4. Continuous basin filling**

392 In a possibly more realistic simulation, the mare material continuously fills the basins until t_{end} .

393 The value of t_{end} is the same as previously, but the total thickness of the basaltic deposit is equal

to H_{DeHon} (Figure 1b). The peak time of mare fillings is 3.8 Ga. Note that the following discussions concerning the abundance of non-mare component is on the basis of those given conditions.

595 concerning the abundance of non-mare component is on the basis of those given

4.1. Diffusion features of non-mare material

When the volcanic material is continuously filling the basins, the volume and therefore thickness of each eruption step is much less. Although the surface laterally transported non-mare material would be covered over repeatedly, the smaller thickness of coverage increases the probability that impact events re-excavate the covered laterally transported non-mare material. Some of the laterally transported non-mare component deposited there earlier can become entrained in the ejecta of subsequent impact craters. This way, it could be repeatedly excavated and migrate upwards and finally to the surface layer.

Given the crater diameters of km-scale in simulations, impact events occur on mare regions can easily penetrate through the mare deposit of hundreds of meters in thickness leading to a comparable picture of the concentration of vertically transported non-mare component with that shown in Figure 2b. But on the regions with thicker mare deposit, the abundance of the mare component on the surface should be significantly increased, so that the relative fraction of the vertically transported component becomes less.

410 **4.2. Magnitude of lateral versus vertical mixing**

Figure 2c displays the laterally transported non-mare component in the near surface (the top 5 m).
It appears essentially the same as in the case of a rapid filling, indicating that the step-wise transport

413 of the early-deposited non-mare component to the near surface is too inefficient to result in a 414 significant difference in composition. The total volume of laterally transported non-mare 415 component over the mare surface is only $\sim 2\%$ larger relative to that with rapid flooding (Figure 2a 416 and the upper sub-plot of Figure 2e), that is, $\sim 2\%$ more the buried component is re-excavated to 417 the mare surface. As we can see that, the histogram distribution of the difference caused by the 418 continual filling (the lower sub-plot of Figure 2e) is smaller than 1.

419 Figure 2d shows that there are fewer zones enriched in non-mare material caused by vertical 420 transport over the Oceanus Procellarum and Mare Imbrium regions. This deficiency of vertically 421 transported non-mare material is caused by the thicker mare deposit, as indicated in Figure 1b. 422 Fewer impact events are able to penetrate the mare material, and thus in those cases, the fraction of the vertically transported non-mare component is lower. The total volume of vertically 423 424 transported non-mare component over the mare surface (the top 5 m) is $\sim 20\%$ smaller relative to 425 that with rapid flooding mainly (Figure 2b and the upper sub-plot of Figure 2f) due to the missing 426 of non-mare zones caused by the vertical transport at the south and southeast of Oceanus 427 Procellarum (Figure 2d). The histogram distribution of the difference caused by the continual filling (the lower sub-plot of Figure 2f) displays the maximum difference is about 5%. 428

The average fraction of laterally and vertically transported non-mare component is 0.07 and 0.03, respectively (Figure 2c and d). The greater average abundance of laterally transported non-mare component indicates the generally higher efficiency of lateral mixing. But it is notable that for a given area of interest, the evaluation of the relative magnitude of the lateral versus vertical mixing needs to take its geologic context into account. For example, if the area is close to the recentlyformed impact crater, lateral mixing could become dominant; if the mare material is known to be only thin, the influence of vertical mixing may increase.





Figure 2 The distribution of non-mare material in the top 5 m. In the first scenario the mare material is emplaced instantaneously (first column, a and b); in the second scenario, it is emplaced by continuous

- flooding with time (second column, c and d). (a) and (c) present the abundance of non-mare material that is laterally: (b) and (d) show the abundance of non-mare component that was vertically excavated from the
- is laterally; (b) and (d) show the abundance of non-mare component that was vertically excavated from the beneath. The upper sub-plots of (e) and (f) present histograms of laterally and vertically transported non-
- 442 mare abundance of the mare regions when the mare material is emplaced instantaneously, and their lower
- 443 sub-plot the absolute differences of the percentage caused by the continual filling

444 **4.3.** Varying parameters concerning mare filling

Given the uncertainty in the flux of mare filling, simulations with varying setting of mare fillings are run to investigate the major factors affecting the non-mare mixing. The parameters studied include the thickness of mare deposits, the cessation time of the major mare filling and the peak time of eruption. To better compare the results, the sequence of impact craters remains the same. Only the studied parameter is varied, the values of all other parameters are identical to those described in section 2.2 in each simulation. Table 1 summarizes the simulations shown in this section.

| Parameters studied | Mare thickness | Cessation time of the major mare fillings | Peak time of eruption (Ga) |
|--|------------------------|--|-------------------------------|
| | H_{DeHon} | | 3.8 |
| Mare thickness | 1/5 $H_{ m DeHon}$ | $t_{\sf end}$ | |
| | 1.5 H_{DeHon} | | |
| | or H _{DeHon} | $t_{ m end}$ | |
| Cessation time of the major mare fillings | | t' _{end} | 3.8 |
| mare minigs | | t" _{end} | |
| Peak time of eruption | $H_{\rm DeHon}$ | 4 | 3.8 |
| | | Lend | 3.3 |

452 Table 1 Simulations with varying parameters concerning mare fillings.

453 Parameter I: Mare thickness

Estimates of total volume of mare deposit vary significantly. The implicit assumption of the cratergeometry approach that De Hon applied is that all the flooded craters were originally formed on pristine basin floors before any lava had been erupted. The volume estimates are therefore minimum values. Du et al. (2019) found that mare thickness derived from measurements of buried craters were systematically over-estimated because the topographic degradation of the impact craters was not considered. After taking degradation into account, they found that the median thickness was 105 m, which is nearly $5 \times$ thinner than H_{DeHon} described in section 2.2 (Figure 1b).

- 461 In contrast, Gong et al. (2016) used GRAIL gravity data to estimate average mare thickness and
- 462 found it to be 740 m, which is close to $1.5 \times$ thicker than H_{DeHon} .
- 463 Both simulations with the thinner $(1/5 H_{DeHon})$ and thicker $(1.5 H_{DeHon})$ maria are performed. The
- 464 statistical non-mare abundance is shown in Figure 3 (the spatial distribution is presented in Figure
- 465 S4 in the supplementary file). It is seen that the abundance of laterally transported material is not
- 466 affected by the mare volume, but the vertically transported non-mare component is significantly
- 467 altered. Since the underlying non-mare component is more easily excavated from the thinner mare

468 deposit, the average abundance of vertically-transported non-mare component on the surface

increases by 110%. For the thicker mare infill, the abundance of vertically transported materialdecreases by 40%.

471 *Parameter II: cessation time of the major mare filling*

472 Given the same total deposit of mare material, as the onset of the mare basalt is taken to be the 473 basin formation age (the upper limit of volcanic eruptions, Ivanov & Melosh 2003), a later 474 cessation time of volcanic eruptions leads to a lower lava eruption flux (i.e., the thinner mare 475 deposit of each mare eruption). To investigate its influence, the simulations with the later cessation 476 time (t'end and t''_{end} in Table 2) are performed. The value of t'_{end} is obtained when the fraction of area of mare deposits younger than a certain age relative to the whole mare region is taken to be 2/3477 (rather than $\frac{1}{2}$, t_{end}); The value of t''_{end} is taken from the age of the youngest mare units of each 478 479 region representing an extreme simulation.

480

Table 2 The cessation time of the main flooding among different mare regions.

| Mare ¹ | $t_{\rm end}, {\rm Ga}^2$ | $t_{ m end}^{\prime}$, Ga 3 | $t_{\rm end}^{\prime\prime},{\rm Ga}^4$ |
|---------------------|---------------------------|---------------------------------|---|
| Oceanus Procellarum | 3.4 | 3.0 | 1.2 |
| Frigoris | 3.5 | 3.5 | 2.6 |
| Humorum | 3.5 | 3.5 | 2.9 |
| Nectaris | 3.6 | 3.5 | 3.4 |
| Fecunditatis | 3.6 | 3.5 | 3.4 |
| Tranquillitatis | 3.6 | 3.5 | 3.4 |
| Serenitatis | 3.5 | 3.3 | 2.4 |
| Crisium | 3.3 | 3.1 | 2.7 |
| Imbrium | 3.3 | 3.1 | 2.0 |

¹ The ages of geologic units of the Mare Crisium, the Mare Nectaris and the Mare Fecunditatis are derived from Hiesinger, van der Bogert, *et al.*, 2011 and Hiesinger *et al.* 2008, and Hiesinger *et al.* 2006, respectively. The others are taken from Hiesinger, Head, *et al.* 2011; ² More than half of each mare region is covered with volcanic material older than t_{end} ; ³ More than two thirds of each mare region is covered with volcanic material older than t'_{end} . ⁴ t''_{end} is the age of the youngest mare unit of each mare region.

The later cessation time extends the time when laterally transported non-mare material is being buried by the emplaced volcanic material. The volume of laterally-transported non-mare component in the near surface would therefore be less. As seen from Figure 3, in the near surface, when the cessation time of mare flooding is taken to be t'_{end} and t''_{end} , the abundance of the laterally transported non-mare component is 45% and 50% less relative to the estimate shown in Figure 2d, respectively (the spatial distributions are shown in Figure S5)

491 respectively (the spatial distributions are shown in Figure S5).

The later emplacement not only buried the more laterally transported non-mare component, but also the more late-excavated non-mare component caused by the vertical transport. The concentration of vertically transported non-mare component is therefore also less. When the cessation time of mare flooding is taken to be t'_{end} and t''_{end} , its average fraction is decreased by 33% and 40%, respectively (Figure 3 and Figure S6).

490 and 4070, respectively (1 igure 5 and 1 igure 1

497 *Parameter III: peak time of eruption*

To investigate the influence from a varying eruption flux, the simulation is performed with a later volcanic eruption peak at 3.3 Ga (instead of 3.8 Ga). Since the abundance of the laterally transported non-mare component depends on the cessation time of the volcanic eruptions, its concentration on the surface is unchanged (Figure 3 and Figure S6).

502 The younger volcanic peak indicates that the volume (i.e. thickness) of the old volcanic eruption

- 503 is less. Therefore, in the early period, there would be more vertically transported non-mare being
- 504 excavated to the near-surface, some of which could be finally delivered to the top surface on
- 505 present day by the cumulative impact mixing. The simulation results (Figure 3), however, show
- that the increased amount of the vertically transported non-mare (<1%) is too less to significantly
- 507 alter the material composition in the top surface.



508

509 Figure 3 The fraction of laterally (a) and vertically (b) transported non-mare material with varying 510 parameters concerning mare filling in the top 5 m. The results based on the different studied parameters are 511 indicated by four grey zones. For better comparison, the first zone shows the results based on the parameters 512 described in section 2.2 (with mare thickness, H_{DeHon} ; cessation time of mare filling, t_{end} ; peak time of mare 513 filling, 3.8 Ga). The second, third and fourth zone show the results for other values of mare thickness, 514 cessation and peak time of mare filling, respectively (the parameters are summarized in the Table 1). The 515 schematics above the grey zones illustrate the differences of the studied parameters. The non-mare 516 distribution is shown as box-and-whisker plots. The inside red lines are the median value and the upper and 517 lower boundaries of the boxes are the first and third quantile. The ends of the whiskers represent 99% of 518 data. The black dots represent the mean values.

519 In summary, on the lateral transport, only the cessation time of the major mare filling has a 520 significant influence. Vertical transport is strongly influenced by both infill thickness variations

and the cessation time. The timing of the peak volcanic activity does not exert a significant

521 and the cessation time. The timing of the peak volcame activity does not exert a significant

522 influence. The non-mare abundance with respective to the simulations of difference parameters is

523 presented in Table S1.

5. Spatial distribution of non-mare component 524

525 5.1. Overall features

526 Because of local impact mixing, the regional distribution of the non-mare component varies with 527 the different random spatial configuration of the generated impact craters in simulations. By taking 528 an average of the results from ten simulations where the spatial configuration of generated craters 529 is different, the regional differences, especially the non-mare enriched zones caused by vertical 530 transport, are averaged out and the general features of non-mare material concentration are 531 obtained (Figure 4).

- 532 The spatial distribution of the total non-mare material including both laterally and vertically
- 533 transported non-mare component in the near surface (the top 5 m) is shown in Figure 4a. Since the
- 534 regional enrichment in the non-mare component is averaged out, the distribution in each mare
- 535 region is relatively homogeneous. In addition, it can be seen that throughout the mare regions the
- 536 non-mare component is present. This is consistent with the results from orbital X-ray instruments,
- 537 which demonstrated that the admixture of non-mare components is a ubiquitous feature over the
- 538 entire mare surface (Trombka et al., 1974; Rhodes, 1977; Prettyman et al., 2006).
- 539 The older mare surfaces accumulate more non-mare material. The non-mare component over the
- 540 Mare Tranquillitatis, Fecunditatis, and Nectaris regions showing the more reddish and lighter color
- 541 is more abundant than that of Oceanus Procellarum and the Mare Serenitatis, Imbrium and Crisium
- 542 regions. In addition, the boundary of mare and highland regions is generally clear but that of the
- 543 mare regions where volcanism ceased early is relatively fuzzy, such as Mare Frigoris,
- 544 Tranquillitatis and Fecunditatis.

545 The histogram of the non-mare fraction of all the mare regions in the top 5 m (Figure 4b) shows 546 that if most of the mare regions have filled mare material about 500 m in thickness since the 547 formation of basins, the average and median fractions of non-mare component over all the mare 548 regions are both about 0.1. Half of the mare regions possess a non-mare component fraction 549 between 0.05 and 0.15. The mare regions with non-mare fraction smaller than 0.1 are mainly 550 distributed in the Mare Crisium region and the inner parts of Oceanus Procellarum and Mare 551 Imbrium. This results not only from their late volcanic filling, but also from the long distances 552 between the rim and the central regions.



553

554 Figure 4 Averaged non-mare material abundance in the top 5 m based on multiple simulations. (a) Spatial 555 distribution of the non-mare component. Red stars indicate the location of Apollo (A) and Luna (L) 556 sampling sites. (b) Histogram of the non-mare component abundance in the top 5m by area over the mare 557 regions. The sum of the bars' value is 100%. (c) Average distribution of non-mare component in the top 1 558 and 5 m with the distance from mare-highland contact. The black curves indicate the average values and 559 the upper and lower bands of the plots (grey zones) are defined by the 95% confidence intervals from the 560 Monte Carlo simulation results. (d) Non-mare material abundance at the Apollo and Luna sampling sites 561 (Bence & Grove 1978; Rhodes 1977), and its distribution with the distance from the mare-highland boundary. The grey bars with slashes represent the chemical estimate of the lunar soil samples. The blue 562 563 diamonds are the median abundance from Huang et al. (2017). The red symbols are the results of this study 564 where the points indicate the mean value. The box-and-whisker plots are presented for the better 565 comparison with Huang et al.'s results, where the inside red lines are the median values and the upper and 566 lower boundaries of the boxes are the first and third quantile. The red triangle and the red inverted triangle 567 are the predicted non-mare fraction at A12 when the very early cessation time of mare filling (0.1 Ga after 568 the basin formation) and when the formation of Copernicus Crater (0.8 Ga, 98 km in diameter) is considered, 569 respectively (all the other parameters are the same as those in section 2.2).

570

5.2. Distribution with the distance from the mare-highland boundary

571 The fraction of non-mare component at varying distance from mare-highland boundary is 572 calculated by taking the average value of the non-mare fraction of each mare region at the given 573 distance. In the top 5m, the average fraction of the total non-mare material, i.e. including both 574 laterally and vertically transported non-mare material, within 100 km is both about 0.2; between

- 575 100 km and 200 km, the non-mare abundance quickly decreases; on the more internal mare regions,
- 576 the fraction is smaller than 0.1.
- 577 The average composition in the top 5 m is regarded to be comparable with that of the lunar surface
- 578 regolith where the material is homogeneously distributed due to the long-term gardening of small
- 579 impacts. But for a young surface, such as the youngest geologic unit in Oceanus Procellarum with
- 580 an age of 1.2 Ga (Hiesinger, Head, *et al.*, 2011), only the very top surface has been well-mixed.
- 581 The calculated average non-mare abundance in the top meter (Figure 4c) indicates that a fresh
- 582 surface without intensive mixing could possess higher abundance of non-mare component.

583 6. Non-mare concentration at sampling sites

This section focuses on the material composition at the sampling sites, to test whether our model is able to reproduce these observations. The results on the basis of multiple simulations weaken the influence of local impact mixing. The calculated average non-mare distribution (Figure 4a) provides expectation values for the spatial composition, although we understand any particular impact configuration will show local differences. Based on the composition of the background surface, the non-mare abundance at the sampling sites is predicted (Table 3).

- 590 Figure 4d (red points) shows the expected fraction of the non-mare material of Apollo 11, 12, 15,
- 591 17 and Luna 16, 24 sampling sites in the near surface, and in relation to the distance from the mare-
- highland boundary. All these sampling sites are located near (<~100 km) the mare-highland
- 593 contact where we expected a weak correlation with the distance. Apollo 11, 17 and Luna 16 are
- both located in Mare Tranquillitatis, at the contact of Mare Tranquillitatis and Serenitatis, and in the
- 595 Fecunditatis regions, respectively. The volcanic cessation time of these regions was relatively early,
- resulting in the higher abundance of non-mare component, with a non-mare component fraction of up to ~ 0.3 . The Apollo 15, 12, and Luna 24 sampling sites are located on the Mare Imbrium,
- 597 up to ~0.5. The Apono 15, 12, and Luna 24 sampling sites are located on the Mare Informati, 598 Oceanus Procellarum, and the Mare Crisium surfaces, where the volcanic cessation time was later
- 599 leading to a less non-mare component (~ 0.1).
- 600 The non-mare abundance as a function of distance from the mare-highlands boundary and at the 601 sampling sites were also predicted by Huang et al. (2017). The median values of their results at 602 the sampling site are shown in Figure 4d (blue diamonds). The results of Huang et al. (2017) display no clear relationship between the non-mare abundance and the distance which is consistent 603 604 with this study, but the non-mare abundance calculated by Huang et al. is generally higher and 605 more comparable with the average composition predicted for the very top surface by our model (Figure 4c). For better comparison, the predicted median values of non-mare abundance of this 606 607 study are also presented (the red box-and-whisker plot; Table 3). It is seen that both the average 608 and median abundance of non-mare component are smaller than Huang et al's estimate. This can 609 at least in part be explained by the assumptions in the model of Huang et al.: They divided the 610 lunar surface into mare (4 km in thickness) and non-mare zones and took this as the initial status 611 of the model. They did not consider the varying cessation time of mare flooding among the 612 different mare regions and did not involve the continuous mare filling that would bury the early-613 transported non-mare material, decreasing its abundance. In addition, as indicated in the Figure 4c,
- 614 the top surface is expected to possess a higher abundance of non-mare material. Huang et al.

615 calculated the abundance of non-mare material of the very top surface (the uppermost millimeter(s))

- 616 which could result in the greater non-mare abundance than this study (the upper meters of the lunar
- 617 surface material). The process of regolith gardening was contained in Huang et al's model. Such a
- 618 process would make the material, especially of the top surface (Figure 6 in Huang et al. 2017), 619 homogeneously distributed diluting the non-mare concentration. Even though such a process is not
- 619 nonogeneously distributed diffing the non-mare concentration. Even though such a process is not 620 considered in this study, our predicted non-mare abundance is still less than Huang et al's estimate.
- 621 It may indicate that other factors, such as the sampling depth and the burial by the mare material,
- have a greater influence on the surface non-mare concentrations. Li and Mustard (2000) also
- 623 calculated the non-mare abundance through an anomalous diffusion model, but only a narrow zone
- 624 of the mare/highland boundaries was analyzed. Their results showed that the non-mare fraction
- 625 quickly decreased from ~ 0.5 to ~ 0.2 over 5 km (the mixing-zone). Due to the limit of our model's
- 626 resolution (5 km), although the drop of the non-mare concentration is not presented in this study
- 627 (Figure 4), our estimates both in the top one (~ 0.2) and the top five (~ 0.3) meters near the
- 628 mare/highland boundary are consistent with Li and Mustard's results.

| Sampling sites | Chemically | This study ² | | Huang et al. (2017) |
|----------------|------------|-------------------------|--|------------------------|
| | | Average | Median ³ | Median |
| A11 | 0.2 | 0.19 | $0.17^{0.32}_{0.06}$ | 0.36 |
| A12 | 0.3 - 0.7 | 0.06 | $0.04_{\scriptstyle 0.01}^{\scriptstyle 0.07}$ | 0.36 |
| A15 | 0.2 - 0.8 | 0.12 | $0.04_{\scriptstyle 0.03}^{\scriptstyle 0.11}$ | 0.34 |
| A17 | 0.1 - 0.7 | 0.29 | $0.24_{0.14}^{0.64}$ | 0.32 |
| L16 | 0.2 | 0.12 | $0.07_{0.03}^{0.14}$ | 0.32 |
| L24 | 0 - 0.1 | 0.07 | $0.02_{0.01}^{0.02}$ | 0.26 |

629 Table 3 Non-mare fraction at the sampling sites.

630 ¹ Rhodes 1977, Bence & Grove 1978; ² parameters concerning mare flooding are taken to be: mare thickness:

631 H_{DeHon} ; the cessation time of the major filling: t_{end} ; the peak time of mare eruption: 3.8 Ga;³ the upper and 632 the lower limit indicate the first and the third quantile.

633 Grey bars in Figure 4d present the abundance of the non-mare component in the soil samples of 634 Apollo 11, 12, 15, 17 and Luna 16, 24 (Rhodes 1977). The model-predicted non-mare abundance

635 and the measurements of the mare soil samples are then compared:

636 Apollo 15/17: Both Apollo 15 and 17 samples were collected from multiple stations with the

637 different geologic settings, and thus the non-mare concentrations display widely varying ranges of

638 non-mare concentration. Rhodes et al. (1974) classified the Apollo 17 soils samples into three

639 compositional groups. Each one related to a specific geologic setting: valley floor type, south

640 massif type and north massif type. The south massif type soils contain abundant non-mare material

641 with a fraction up to ~0.75 (e.g. Korotev and Kremser 1992), which is much greater than that of

642 the valley floor type soils (~ 0.2). Similarly to the Apollo 17 soil samples, the Apollo 15 soil 643 samples were found to be dominated by two types (Apennine Front and Apennine base) and the 644 non-mare fraction ranges from about 0.2 to 0.8 (Duncan 1975). As can be seen from Figure 3d, 645 the model-predicted non-mare abundance falls at the lower range of the chemically estimated 646 abundance in the mare soil samples. Given that the sampling site is adjacent to the highland massifs 647 which present the similar chemical composition (KREEPy) with those non-mare samples (Korotev, 648 1976; Laul et al., 1978), downslope transport by slumping of nearby highlands could be the most 649 plausible process enriching the non-mare abundance of the collected samples.

650 Apollo 12: The measurements of different Apollo 12 sub-samples also present a wide range of 651 non-mare fraction from 0.3 to 0.7 which is greater than the model-predicted concentration (~ 0.05). Such a great non-mare abundance (the inverted red triangle in Figure 4d) may be obtained when 652 the very early cessation time of mare-filling is considered in our model (e.g., 0.1 Ga after the basin 653 654 formation). However, given the KREEP background of the landing site, such an early cessation 655 time of mare-filling is unlikely. The role of other factors is likely more important. The sampling 656 site is close to the late-formed Copernicus crater (390 km north of the site; Wilhelms 1987). 657 According to the orientation of the bright rays of the Copernicus crater, the ejecta material has been believed to be derived from Copernicus crater. The non-mare material entrained in the ejecta 658 659 of Copernicus could have been spread across the sampling site; in the meanwhile, the high-velocity ejecta could have excavated a certain amount of the underlying non-mare component (Huang et al. 660 661 2017; Quaide et al., 1971; Schonfeld and Meyer Charles, 1972). Multiple simulations with the formation of Copernicus crater (800 Ma, 96 km in diameter) are also performed. The results show 662 663 that if all the other parameters remain the same as those in section 2.2, the non-mare fraction at A12 is increased by a factor of two (the red triangle in Figure 4d). This is still less than both the 664 665 results of the chemical estimate and Huang et al. A plausible explanation is that the distal ejecta is 666 taken to be continuously distributed so that the spatial geometry of crater rays that were included 667 in Huang et al are not considered. The continuously distributed distal ejecta could be regarded as 668 the consequence of prolonged impact mixing. However, given the young age of Copernicus crater, 669 most of its rays are well-preserved, and thus its influence on the A12 sites could be more significant 670 than we predict.

671 *Apollo 11/Luna 24*: The sampling site is far away from the highland-mare boundary and no obvious 672 disturbance of the young ejecta is observed. The model-predicted non-mare fraction of the Apollo 673 11 and Luna 24 soil samples is ~ 0.2 and ~ 0.1 , respectively, which falls within the range of the 674 chemical estimates. It indicates that the non-mare component in these soil samples has been mixed 675 with both laterally and vertically transported non-mare material caused by the cumulative impact 676 mixing.

677 *Luna 16*: The model-predicted abundance of the non-mare component is less than the chemical 678 estimate of the samples. This may indicate either that the Mare Crisium mare deposit is thinner 679 than 500 m or that the mare surface is older than 3.3 Ga (t_{end} in Table 2).

For the Apollo 11, 15 and Luna 16 samples, rays from Theophilus, Aristarchus or Autolycus, and
 Langrunus or Tarunius have been proposed as the sources for the highland non-mare components,

respectively (Duncan et al., 1975; Schonfeld, 1975). Taking the Apollo 11 sampling site as an

683 example, it is ~380 km (~7.6 radii) away from Theophilus crater (99 km in diameter). The ray 684 length of Theophilus crater can be estimated to be \sim 950 km (Elliott et al. 2018), so that some of 685 its distal ejecta might have deposited at the Apollo 11 sampling site. If the scaling law of ejecta 686 thickness distribution is extrapolated to this distance, the thickness of Theophilus ejecta at the Apollo 11 sampling is about 1 m. The ratio of the amount of local material to that of ejecta is 687 688 estimated to be \sim 3, that is, the fraction of ejecta is about 25%. This is comparable with the 689 chemically estimated value of ~20%. However, the bright ray of Theophilus ejecta has been 690 degraded and is hardly detectable on the present day. It is difficult to determine whether the Apollo 691 11 sampling site is really located on the ray deposit. If it is, the calculation of the local ejecta 692 thickness indicates that the non-mare component in the samples is mainly derived from Theophilus 693 ejecta. The new look at the Apollo 11 regolith which suggested the more mixing of non-mare 694 component (>20%; Korotev and Gillis, 2001) may further support the deposit of the Theophilus 695 ejecta.

696 In addition to the impact mixing, some other factors could also affect the non-mare abundance at 697 the sampling sites, such as very young mare lava flows. Due to their small volume, they are 698 typically regionally restricted and can be detected based on spectral data (e.g., the spectral ratios 699 of Clementine imaging data used for the definition of the basaltic units; Hiesinger, Head et al. 700 2011). In addition, thorough studies of soils samples (e.g., Apollo 17; Rhodes 1974) suggested that 701 the less coherent anorthositic (highland) material was preferentially sampled. Therefore, the 702 sampling strategy could also have led to a bias towards a higher abundance of non-mare material 703 resulting in the difference between the predicted abundance and the chemical estimate of the 704 samples.

It should be mentioned that the abundance of non-mare material is predicted on the basis of the given model parameters including the mare thickness (H_{DeHon}), the cessation (t_{end}) and peak time

(3.8 Ga) of mare flooding. Varying these values would change the predicted absolute value of non mare abundance, but would not significantly change relative abundance among the areas covered

- 709 with different amount of mare material.
- For the Apollo 11 and Luna 24 sampling sites, the consistency between the model-predicted and
- the chemically estimated non-mare abundance suggests that the mare thickness at both sites is
- 712 likely to be on the order of 500 m, and that other geologic processes aside from impact mixing did
- 713 not play a significant role.

714 **7. Conclusions**

715 Lunar mare soil samples from the Apollo and Luna missions contain non-mare component (Rhodes,

1977). To investigate the possible origin and distribution of the contained non-mare component, a

717 numerical model was developed to trace the diffusion of non-mare material, where both the vertical

and lateral mixing are recorded to investigate their evolving features. The volcanic flux is the most important parameter, but it is not well constrained (e.g., the total mare volume and the onset and

end of eruptions). The abundance of non-mare material is therefore calculated for various possible

⁷²⁰ end of eruptions). The abundance of non-mare material is therefore calculated for various possible

721 scenarios.

722 The laterally and vertically-derived non-mare component display different diffusion features. The 723 distribution of the laterally transported non-mare material is relatively homogeneous and closely 724 related to the distance to the mare-highland boundary. The majority of it is distributed near the 725 mare-highland boundary, while the central mare areas possess a significantly lower abundance of 726 laterally transported non-mare material. In contrast, the vertically-derived non-mare material is 727 patchily distributed. It is closely related to the distribution of large impact events that could 728 excavate a large amount of the underlying non-mare material to the surface, generating non-mare 729 enriched regions. In addition, we find a higher efficiency of lateral transport than that of vertical transport in general. Those diffusion features of both laterally- and vertically-derived non-mare 730

731 material could be helpful for the choice of future landing sites that aim to collect deep-seated

material and for the interpretation of the source of non-mare component in the collected material.

733 It is found that, across almost the entirety of the mare regions of the Moon, a non-mare component

is present. If the mare thickness is on the order of 500 m, the average non-mare fraction over the

whole mare regions is about 0.1, and half of the areas possess a fraction ranging from 0.05 to 0.15.

736 The mare surfaces with older volcanic cessation times contain more non-mare material, with an

737 average fraction of ~ 0.3 .

738 We compared our results with the analysis of the collected soil samples, and discuss the most 739 plausible geologic processes that may have altered the non-mare material abundance at the 740 sampling sites. In the Apollo 15 and 17 samples, the downslope slumping and lateral transport 741 from nearby high relief regions are a likely cause for the larger non-mare component. At the Apollo 12 sampling site, the ejecta of the nearby crater Copernicus, that not only delivered a non-mare 742 743 component but excavated local material, could increase the regional non-mare concentration. The 744 low amount of non-mare material contained in the Apollo 11, Luna 16 and 24 mare soil samples 745 may result from the gradual accumulation of both the lateral and vertical mixing. In addition, the

region where Luna 16 landed may have a very thin mare deposit.

747 In the future, a model involving topographic relief could be further developed to assess the 748 influence of highland slumping. In addition, with greater understanding of the distribution of distal 749 ejecta, the model could consider some realistic young craters of interest, such as crater Copernicus, 750 and get a more reliable picture of non-mare distribution.

751 Without a well-constrained estimate of volcanic eruption flux, we performed tentative simulations 752 with the varying mare thickness, the peak time of mare eruption and the cessation time of the major 753 mare filling. It is found that on the surface, the total amount of mare deposit can significantly 754 change the abundance of vertically delivered non-mare, but does not influence the abundance of 755 the laterally transported non-mare material; the peak time of mare eruption could not significantly 756 affect the abundance of both laterally and vertically transported component; the cessation time of 757 the major mare fillings is the major factor altering the abundance of laterally transported non-mare material, and it could also affect the fraction of the buried vertically transported non-mare material 758

that had ever been excavated to the near-surface.

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