

## Exposure history of lunar meteorites Queen Alexandra Range 93069 and 94269

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**Abstract**—Cosmic-ray produced <sup>14</sup>C ( $t_{1/2} = 5730$  years), <sup>36</sup>Cl ( $3.01 \times 10^5$  years), <sup>26</sup>Al ( $7.05 \times 10^5$  years), and <sup>10</sup>Be ( $1.5 \times 10^6$  years) in the recently discovered lunar meteorites Queen Alexandra Range 93069 (QUE 93069) and 94269 (QUE 94269) were measured by accelerator mass spectrometry. The abundance pattern of these four cosmogenic radionuclides and of noble gases indicates QUE 93069 and QUE 94269 were a paired fall and were exposed to cosmic rays near the surface of the Moon for at least several hundred million years before ejection. After the meteorite was launched from the Moon, where it had resided at a depth of 65–80 g/cm<sup>2</sup>, it experienced a short transition time, ~20–50 ka, before colliding with the Earth. The terrestrial age of the meteorite is 5–10 ka. Comparison of the cosmogenic nuclide concentrations in QUE 93069/94269 and MAC 88104/88105 clearly shows that these meteorites were not ejected by a common event from the Moon.

### INTRODUCTION

Most lunar meteorites have complex cosmic-ray exposure histories (Eugster, 1989; Nishiizumi *et al.*, 1991), having been exposed both at some depth on the Moon ( $2\pi$  irradiation) before their ejection and as small bodies in space ( $4\pi$  irradiation) during transport from the Moon to the Earth. The exposures, on the Moon and in space, are then followed by a long terrestrial residence time. Measurements of cosmogenic radio- and stable nuclides can potentially unravel the complex histories of these objects. The concentrations of cosmogenic nuclides also constrain the depth of the sample at the time of ejection from the Moon. These exposure durations can then be used to model impact and ejection mechanisms. Generally, unraveling the complex history of these objects requires the measurement of at least four cosmogenic nuclides.

We report here cosmogenic <sup>14</sup>C ( $t_{1/2} = 5730$  years), <sup>36</sup>Cl ( $3.01 \times 10^5$  years), <sup>26</sup>Al ( $7.05 \times 10^5$  years), and <sup>10</sup>Be ( $1.5 \times 10^6$  years) results for the new lunar meteorites Queen Alexandra Range 93069 (QUE 93069) and 94269 (QUE 94269). Partial results were reported earlier by Nishiizumi *et al.* (1995). Noble gas data are now available (Spettel *et al.*, 1995; Swindle *et al.*, 1995; Thalmann and Eugster, 1995) and will be used to obtain the cosmic-ray exposure history of these objects both on the Moon and in space and the terrestrial residence age in Antarctica.

### EXPERIMENTAL METHODS

The anorthositic breccia QUE 93069 was collected in the Queen Alexandra Range, Antarctica. The recovered sample is  $5.0 \times 2.2 \times 2.3$  cm in size and 21.4 g in weight. The top is covered by a thick fusion crust while the bottom has a rather thin fusion crust. Since some lunar meteorites, specifically Calalong Creek, Yamato 791197, and Yamato 793169, contain SCR (solar cosmic ray) produced nuclides, which indicate negligible ablation during atmospheric entry (Nishiizumi *et al.*, 1992; Nishiizumi *et al.*, 1991), the search for SCR effects is an important component of this study. To investigate SCR effects in QUE 93069, we measured <sup>10</sup>Be, <sup>26</sup>Al, and <sup>36</sup>Cl in four subsamples having different shielding depths. Lunar meteorite QUE 93069,14 was chipped into three portions: thick fusion crust (0–1 mm from recovered surface), 1–4 mm, and 5–9 mm. The interior sample, QUE 93069,13 was sampled at 8–11 mm from the surface. Carbon-14 was measured in another subsample QUE 93069,45 for accurate terrestrial age determination.

The four samples of QUE 93069,13 and 14 were crushed and dissolved with a HF-HNO<sub>3</sub> mixture together with Be (2 mg) and Cl (5 mg) carrier

solutions. A small aliquot was obtained for chemical analysis. After the analysis aliquot was taken, Al carrier (7 mg) was added to reduce the <sup>26</sup>Al/Al ratio. Beryllium, Al, and Cl were chemically isolated then from each sample and purified for accelerator mass spectrometry (AMS) measurement using a combination of ion chromatographic and liquid-liquid extraction techniques (Nishiizumi *et al.*, 1984a; Nishiizumi *et al.*, 1984b). The <sup>10</sup>Be, <sup>26</sup>Al, and <sup>36</sup>Cl concentrations were measured using AMS techniques at the Lawrence Livermore National Laboratory (Davis *et al.*, 1990). The concentrations of the target elements Mg, Al, K, Ca, Mn, and Fe were determined by atomic absorption spectrometry.

For <sup>14</sup>C measurements, the sample of QUE 93069,45 was initially cleaned in 100% H<sub>2</sub>PO<sub>4</sub> in order to remove any weathering products (Jull *et al.*, 1993). The CO<sub>2</sub> was extracted then from the cleaned sample (49.4 mg) by fusion with ~4 g of Fe combustion chips in a flow of O<sub>2</sub> using an RF induction furnace at >1600 °C (Jull *et al.*, 1993; Jull *et al.*, 1989). The collected gas was measured and diluted to 0.977 cm<sup>3</sup> STP with <sup>14</sup>C-free CO<sub>2</sub>. This gas was reduced to graphite over an Fe catalyst at 625 °C, and the <sup>14</sup>C concentration was measured by AMS at the University of Arizona (Jull *et al.*, 1993; Jull *et al.*, 1989).

Anorthositic breccia QUE 94269 was collected also in the Queen Alexandra Range, Antarctica. This sample is  $1.9 \times 1.4 \times 1.3$  cm in size and 3.2 g in weight. This lunar meteorite is identical to QUE 93069 and is believed to be paired with QUE 93069. One side of this flat object also has a thick fusion crust, and the other side has a thin fusion crust. To identify the pairing of these two lunar meteorites and to examine the exposure condition in space, <sup>10</sup>Be, <sup>26</sup>Al, and <sup>36</sup>Cl concentrations were measured in QUE 94269. Lunar meteorite QUE 94269,13 was sampled from the thick fusion crust (0–2 mm from the recovered surface) and processed in the same manner as QUE 93069.

### RESULTS AND DISCUSSION

The chemical composition of samples QUE 93069,13,14 and QUE 94269,13 and their corresponding depths are given in Table 1. The measured <sup>10</sup>Be/Be, <sup>26</sup>Al/Al, and <sup>36</sup>Cl/Cl ratios were  $4 \times 10^{-12}$  to  $1 \times 10^{-11}$ ,  $\sim 7 \times 10^{-12}$ , and  $2 \times 10^{-12}$  to  $4 \times 10^{-12}$ , respectively. After background corrections ( $3 \times 10^{-14}$  for <sup>10</sup>Be/Be,  $3 \times 10^{-15}$  for <sup>26</sup>Al/Al, and  $1 \times 10^{-14}$  for <sup>36</sup>Cl/Cl), the measured ratios were normalized to ICN <sup>10</sup>Be, NBS <sup>26</sup>Al, and NBS <sup>36</sup>Cl standards (diluted by one of authors, K. Nishiizumi). The results are shown in Table 2. The errors include a  $\pm 1\sigma$  AMS measurement error and do not reflect the errors in the chemical analysis (1.5–2%), or the uncertainty in the absolute activities of the standards.

The <sup>14</sup>C result for QUE 93069,45 is shown in Table 2 and includes errors from the AMS analysis and measurements of blank samples immediately before and after the sample extraction. The blanks run before and after the sample of QUE 93069 are consistent

TABLE 1. Chemical composition of QUE 93069 and QUE 94269.

| Sample       | Depth from surface | Wt (mg) | Mg (%) | Al (%) | K (ppm) | Ca (%) | Mn (ppm) | Fe (%) |
|--------------|--------------------|---------|--------|--------|---------|--------|----------|--------|
| QUE 93069,14 | 0–1 mm             | 91.19   | 3.18   | 15.3   | 394     | 11.4   | 448      | 3.37   |
| QUE 93069,14 | 1–4 mm             | 63.61   | 3.15   | 15.0   | 316     | 11.4   | 437      | 3.45   |
| QUE 93069,14 | 5–9 mm             | 86.37   | 3.26   | 15.0   | 330     | 11.4   | 461      | 3.63   |
| QUE 93069,13 | 8–11 mm            | 108.70  | 3.08   | 15.3   | 306     | 11.5   | 443      | 3.40   |
| QUE 94269,13 | 0–2 mm             | 95.09   | 2.50   | 15.0   | –       | 11.7   | 453      | 3.45   |

TABLE 2. Cosmogenic radionuclide concentrations in QUE 93069 and QUE 94269.

| Sample       | Depth from surface | <sup>10</sup> Be (dpm/kg meteorite) | <sup>26</sup> Al (dpm/kg meteorite) | <sup>36</sup> Cl (dpm/kg meteorite) | <sup>14</sup> C (dpm/kg meteorite) |
|--------------|--------------------|-------------------------------------|-------------------------------------|-------------------------------------|------------------------------------|
| QUE 93069,14 | 0–1 mm             | 11.42 ± 0.25                        | 74.0 ± 1.6                          | 12.93 ± 0.21                        | –                                  |
| QUE 93069,14 | 1–4 mm             | 12.02 ± 0.27                        | 74.8 ± 2.3                          | 14.52 ± 0.48                        | –                                  |
| QUE 93069,14 | 5–9 mm             | 12.38 ± 0.26                        | 68.4 ± 1.5                          | 14.80 ± 0.15                        | –                                  |
| QUE 93069,13 | 8–11 mm            | 11.95 ± 0.25                        | 68.8 ± 1.4                          | 14.62 ± 0.25                        | –                                  |
| QUE 93069,45 | –                  | –                                   | –                                   | –                                   | 10.1 ± 0.4                         |
| QUE 94269,13 | 0–2 mm             | 12.04 ± 0.26                        | 74.6 ± 1.4                          | 13.31 ± 0.32                        | –                                  |

and averaged  $(1.32 \pm 0.04) \times 10^6$  <sup>14</sup>C atoms, compared to the amount of  $(3.49 \pm 0.07) \times 10^6$  atoms from the sample.

The chemical compositions and cosmogenic radionuclide concentrations in both QUE 93069 and QUE 94269 are identical and support the hypothesis that these two lunar meteorites are a pair. The remaining discussion assumes that they are the same meteorite. Our chemical analyses indicate that the major target elements in each subsample are uniform to within 1–3% except K, which is enriched in the fusion crust by ~25%. Although the <sup>36</sup>Cl production from K is ~4 times higher than that from Ca, this difference is insignificant because the K concentration is 360 times lower than that of Ca in this meteorite.

The <sup>10</sup>Be and <sup>36</sup>Cl concentrations in the five samples are nearly identical with the exception of <sup>36</sup>Cl, which is low in both thick fusion crusts. Volatilization during formation of the fusion crust is the most likely explanation for the low <sup>36</sup>Cl. The same trend for <sup>36</sup>Cl was observed in the thick fusion crust on the Salem meteorite (Nishiizumi *et al.*, 1990). The <sup>26</sup>Al values in exterior samples are 7–9% higher than in interior samples. If the excess of <sup>26</sup>Al is attributed to SCR bombardment, the gradient of the SCR contribution is ~6 dpm/5 mm. The quite good agreement of the chemical analysis and cosmogenic nuclide measurements of the two meteorites also indicates excellent reproducibility of our experiments because the two meteorites were processed one year apart.

Unraveling the exposure history of this meteorite requires that limits be placed on the size of the ejecta and the depth at which the sample was irradiated on the Moon. Like the production rates of cosmogenic nuclides in meteorites ( $4\pi$  irradiation), which vary with the size and shape of the meteoroid and the depth of the sample (Masarik and Reedy, 1994), the production rates of cosmogenic nuclides on the Moon ( $2\pi$  irradiation) also change with depth (Reedy and Arnold, 1972). The depth profiles of <sup>10</sup>Be, <sup>14</sup>C, <sup>26</sup>Al, and <sup>36</sup>Cl have been measured in the Apollo 15 deep drill core (Jull *et al.*, 1991; Nishiizumi *et al.*, 1984a; Nishiizumi *et al.*, 1984b). These experimental results will be used to derive the exposure conditions on the Moon. The production rates for a  $2\pi$  exposure on the Moon and a  $4\pi$ -exposure geometry during transit to the Earth are relatively well established (Masarik and Reedy, 1994; Reedy and Arnold, 1972; Reedy and Masarik, 1994). The measured activity  $A$  produced

in two different exposure conditions,  $2\pi$  and  $4\pi$ , is dependent on three different time periods (Nishiizumi *et al.*, 1991):

$$A = P_m(1 - e^{-\lambda T_m})e^{-\lambda(T_s + T_a)} + P_s(1 - e^{-\lambda T_s})e^{-\lambda T_a} \quad \text{Eq. (1)}$$

where  $P_m$  is the production rate (saturation activity) on the Moon at a specific depth ( $P_m$  changes with irradiation depth),  $P_s$  is the production rate in the meteoritic body in space ( $P_s$  changes with irradiation depth and with preatmospheric size),  $\lambda$  is the decay constant of the radionuclide,  $T_m$  is the exposure age on the Moon,  $T_s$  is the exposure age in space (the transition time from the Moon to the Earth), and  $T_a$  is the terrestrial age. The ejection time is defined by  $T_s + T_a$ . Because the production rates  $P_m$  and  $P_s$  are not defined by a simple function of depth, the exposure condition for a lunar meteorite must be obtained by a combination of numerical calculations and graphical methods with some assumptions.

### Terrestrial Age

The <sup>14</sup>C activity of 10.1 dpm/kg requires a terrestrial age for QUE 93069 of 5–10 ka providing that the <sup>14</sup>C was produced during a  $4\pi$  irradiation and that the exposure age in space,  $T_s$ , is much longer than several half-lives of <sup>14</sup>C. The assumed <sup>14</sup>C production rate,  $P_s$ , in a small object is 20–35 atom/kg-min (Reedy and Masarik, 1995; Wieler *et al.*, 1996). Alternatively, if most of <sup>14</sup>C was produced on the surface of the Moon (*i.e.*,  $T_s$  is much shorter than a half-life of <sup>14</sup>C), the terrestrial age is ~11 ka based on a <sup>14</sup>C production rate,  $P_m$ , at near surface of the Moon of 40 atom/kg-min. The derived terrestrial age will decrease if we assume the meteorite was exposed at a greater depth on the Moon. Other cosmogenic radionuclide data to be discussed later will lead us to dismiss a model involving exposure at depth. Therefore, the <sup>14</sup>C concentration constrains the terrestrial age,  $T_a$ , of QUE 93069 to be between 5 and 11 ka. Further, evidence for  $4\pi$  solar cosmic-ray produced <sup>26</sup>Al production implies that all <sup>14</sup>C must have been produced by a  $4\pi$  exposure in space. The 5–10 ka age is preferred, based on the constraint imposed by the SCR produced <sup>26</sup>Al. Therefore, at the time of the fall, the radionuclide concentrations were  $(14.9 \pm 0.3)$  dpm <sup>36</sup>Cl/kg (excluding fusion crust),  $(69.1 \pm 1.5)$  dpm <sup>26</sup>Al/kg (interior),  $(75.0 \pm 1.5)$  dpm <sup>26</sup>Al/kg (exterior), and  $(12.0 \pm 0.3)$  dpm <sup>10</sup>Be/kg.

## Exposure Age

The high concentration of solar-wind implanted noble gases and the overwhelming amount of cosmogenic  $^{21}\text{Ne}$  and  $^{38}\text{Ar}$  in QUE 93069 suggest that the meteorite was exposed to cosmic rays in the lunar regolith for an exceedingly long time (Spettel *et al.*, 1995; Swindle *et al.*, 1995; Thalmann and Eugster, 1995). The cosmogenic  $^{38}\text{Ar}$  concentration indicates that the meteorite has an exposure age of  $1100 \pm 400$  Ma, which is the longest of all known lunar meteorites (Thalmann and Eugster, 1995). This exposure age is based on total residence time at shallow depth on the Moon. As a minimum, it is reasonable to assume the meteorite was continuously exposed at shallow depth for at least 10 Ma. In the case when this continuous exposure occurred until ejection from the Moon, the term  $(1 - e^{-\lambda T_m})$  for all four radionuclides in Eq. (1) is equal to 1.0. Equation (1) can now be rewritten as:

$$A_0 = P_m e^{-\lambda T_s} + P_s (1 - e^{-\lambda T_s}) \quad \text{Eq. (2)}$$

where  $A_0$  is the radionuclide concentration at the time of fall.

For QUE 93069, the  $4\pi$ -production rate,  $P_s$ , which is based on the measured chemical composition and its small preatmospheric size, is  $\sim 10$ – $25$  atom/kg-min for  $^{36}\text{Cl}$ ,  $\sim 50$ – $150$  atom/kg-min for  $^{26}\text{Al}$ , and  $17$ – $23$  atom/kg-min for  $^{10}\text{Be}$ . The  $2\pi$ -production rate on the Moon,  $P_m$ , is a function of depth and chemical composition (Reedy and Arnold, 1972; Reedy and Masarik, 1994). In previous studies, the  $2\pi$ -production rates of cosmogenic nuclides were calculated using the observed production profiles in Apollo 15 drill core, target elemental compositions, and the Reedy-Arnold theoretical production model (Nishiizumi *et al.*, 1984a; Nishiizumi *et al.*, 1984b; Reedy and Arnold, 1972). In this analysis, we will adopt the observed activities of  $^{26}\text{Al}$  and  $^{10}\text{Be}$  in lunar core 60010,3234 (23 g/cm $^2$ ), 60009,3271 (51 g/cm $^2$ ), and 60009,3273 (75 g/cm $^2$ ) because the chemical composition of 60009/10 (average 2.9% Mg, 15.6% Al, 900 ppm K, 11.6% Ca, 450 ppm Mn, and 3.4% Fe) is similar to that of QUE 93069. These activities are 13.3, 12.9, and 11.3 dpm  $^{10}\text{Be}$ /kg and 74, 70, and 66 dpm  $^{26}\text{Al}$ /kg at 23, 51, and 75 g/cm $^2$ , respectively. The averaged measured  $^{36}\text{Cl}$  concentration (14.9 dpm/kg sample at fall) in QUE 93069 is corrected for chemical composition to 15.6 dpm  $^{36}\text{Cl}$ /kg (Fe + 8Ca + 32K). The normalized  $^{36}\text{Cl}$  activity in QUE 93069 is compared then to measurements of  $^{36}\text{Cl}$  in 18 lunar cores with shielding between 20 and 80 g/cm $^2$ . The observed activities (using  $\pm 2\sigma$ ) in QUE 93069 are fitted then to those activities in lunar cores with 45–80 g/cm $^2$  for  $^{10}\text{Be}$ , 15–85 g/cm $^2$  for  $^{26}\text{Al}$ , and 20–40 and 65–85 g/cm $^2$  for  $^{36}\text{Cl}$ . The depth that can best account for all the radionuclide activities is 65–80 g/cm $^2$ . If this analysis is correct (ejection from 65–80 g/cm $^2$ ), then QUE 93069 subsequently experienced a negligible  $4\pi$  exposure. The assumption of a slightly deeper ejection depth (lower production rates) and some  $4\pi$  exposure, such as 0.1 Ma, produces a poor fit for the  $^{36}\text{Cl}$  and  $^{10}\text{Be}$  data.

The  $^{26}\text{Al}$  within 4 mm of the surface is  $\sim 9\%$  higher than that of the interior samples. If this excess  $^{26}\text{Al}$  is SCR produced, it is necessarily produced during the  $4\pi$  irradiation because the fusion crust on both sides (QUE 93069 and 94269) contains SCR-produced  $^{26}\text{Al}$ . The production rate of SCR-produced  $^{26}\text{Al}$  is strongly dependent on depth and the object's size in space. Since QUE 93069 and 94269 seem physically to fit together and contain SCR-produced  $^{26}\text{Al}$  in fusion crust on opposite sides, it is reasonable to assume that QUE 93069 was a small object in space, similar to the Salem meteorite. Salem contains the highest observed (SCR + GCR produced)

$^{26}\text{Al}$  ( $\geq 150$  dpm  $^{26}\text{Al}$ /kg) among chondrites (Evans *et al.*, 1987; Nishiizumi *et al.*, 1990). For the composition of QUE 93069, this Salem  $^{26}\text{Al}$  surface activity corresponds to  $\sim 300$  dpm/kg, and the excess of  $^{26}\text{Al}$  in the surface of QUE 93069 ( $6 \pm 2$  dpm/kg) requires an exposure of  $\geq 0.02$  Ma  $4\pi$  exposure. Although  $^{36}\text{Cl}$  is produced during the same  $4\pi$ -exposure period, the  $^{36}\text{Cl}$  excess is only 4% over the expected  $2\pi$  production on the lunar surface. If our adopted  $2\pi$ -production rate is correct, the  $^{36}\text{Cl}$  data require the  $4\pi$ -exposure duration to be  $\leq 0.05$  Ma. This limit is less than the estimate given in Nishiizumi *et al.* (1995), which was based on different  $2\pi$ -production rates and without the  $^{14}\text{C}$  result.

We conclude that the paired meteorites QUE 93069 and QUE 94269 had a 5–10 ka terrestrial age, had a short  $4\pi$  exposure of 0.02–0.05 Ma, and were ejected from 65–80 g/cm $^2$  below the surface of Moon.

According to the Antarctic Meteorite Newsletter (1994), the thin section of QUE 93069 is very similar to that of the MacAlpine Hills 88104/88105 pair (MAC 88105). Our chemical analysis also indicates a very similar chemical composition except that Mg is slightly higher in QUE 93069. These two lunar meteorites were collected 40–45 km apart in Antarctica. However, the cosmogenic radionuclide concentrations in QUE 93069 are very different from those in MAC 88105 (Eugster *et al.*, 1991; Jull and Donahue, 1991; Nishiizumi *et al.*, 1991; Vogt *et al.*, 1991). We found 3.42 dpm/kg of  $^{36}\text{Cl}$ , 17.5 dpm/kg of  $^{26}\text{Al}$ , and 2.26 dpm/kg of  $^{10}\text{Be}$  in MAC 88105 (Nishiizumi *et al.*, 1991). MacAlpine Hills 88105 was most likely ejected from a depth of 360–400 g/cm $^2$  on the Moon 0.26–0.29 Ma ago. This meteorite was exposed to cosmic rays as a small body in space for  $\sim 0.04$ – $0.05$  Ma. The terrestrial age of the meteorite is 0.21–0.25 Ma. The short terrestrial age and short ejection age of QUE 93069 indicate that QUE 93069 represents material from a different ejection from the Moon than MAC 88105.

The exposure histories of 15 lunar meteorites, of which 11 are independent cases, have been studied. Figure 1 shows ejection and terrestrial ages of all the lunar meteorites studied thus far except QUE 94281. Since ejection ages of some meteorites overlap, we cannot rule out the same ejection events for these meteorites. Mea-

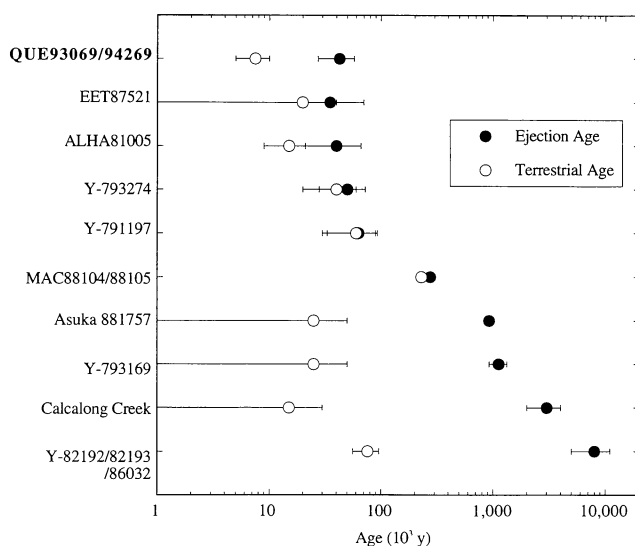


FIG 1. Ejection ages and terrestrial ages of lunar meteorites. The open circles indicate terrestrial ages, and the closed circles indicate ejection times from the Moon. The distance between the two circles indicates the transition time from the Moon to the Earth.

surements of  $^{41}\text{Ca}$ ,  $^{53}\text{Mn}$ , and  $^{14}\text{C}$  in these lunar meteorites will help establish exposure chronologies. At least half of the lunar meteorites have very short transition times, reaching the Earth within 0.1 Ma of their ejection. In contrast, SNC meteorites have much longer transition times (Bogard *et al.*, 1984; Nishiizumi *et al.*, 1986), near three Ma or longer (with the notable exception of EET 79001).

Arnold simulated the dynamics of meteorites having a lunar origin using a Monte Carlo model (Arnold, 1965). According to this analysis, of those objects ejected from the Moon that escape the Earth's gravity field, roughly half reach the Earth with transit times of <1 Ma. The nature of the model did not allow an estimate of the fraction with  $4\pi$  exposure <0.1 Ma, but a rough fraction might be 15–20% (Arnold, pers. comm.). One explanation is that some, and perhaps most, of the objects that did not escape from the Earth-Moon system upon ejection were captured quickly by the Earth. However, at the present time, the case rests on only ten samples, which are not enough to prove or refute the model calculations.

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