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The proto-Earth as a significant source of lunar material

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A giant impact between the proto-Earth and a Mars-sized impactor named Theia is the favoured scenario for the formation of the Moon[1–3]. Oxygen isotopic compositions have been found to be identical between terrestrial and lunar samples[4], which is inconsistent with numerical models estimating that more than 40% of the Moon-forming disk material was derived from Theia[2,3]. However, it remains uncertain whether more refractory elements, such as titanium, show the same degree of isotope homogeneity as oxygen in the Earth–Moon system. Here we present $^{50}Ti/^{47}Ti$ ratios in lunar samples measured by mass spectrometry. After correcting for secondary effects associated with cosmic-ray exposure at the lunar surface using samarium and gadolinium isotope systematics, we find that the $^{50}Ti/^{47}Ti$ ratio of the Moon is identical to that of the Earth within about four parts per million, which is only 1/150 of the isotopic range documented in meteorites. The isotopic homogeneity of this highly refractory element suggests that lunar material was derived from the proto-Earth mantle, an origin that could be explained by efficient impact ejection, by an exchange of material between the Earth's magma ocean and the protolunar disk, or by fission from a rapidly rotating post-impact Earth.

Apart from the effects of radioactive decay, the isotopic compositions of most terrestrial rocks are related by the laws of mass-dependent fractionation. Meteorites show departures from this rule that can be ascribed to unusual chemical processes, inheritance of nucleosynthetic anomalies, or nuclear transmutations (cosmogenic effects and radioactive decay). In the zoo of elements that show well-documented isotopic anomalies at a bulk planetary scale[5–8], highly refractory titanium, with large nucleosynthetic anomalies on ^{50}Ti, is the most promising to assess the degree of homogeneity in the Earth–Moon system[9]. Taking advantage of our new chemical procedure for titanium separation and developments in multicollector inductively coupled plasma mass spectrometry (MC-ICPMS; see Methods and Supplementary Information for details; ref. 10), we measured the titanium isotopic compositions of 5 terrestrial samples, 37 bulk chondrites, and 24 lunar samples (8 whole rocks, 6 ilmenite separates, 1 pyroxene separate, and 9 soil samples) with substantially higher precision (0.04–0.11 ε-unit for $\varepsilon^{50}Ti$, 2SE, where $\varepsilon^{50}Ti = [(^{50}Ti/^{47}Ti)_{sample}/(^{50}Ti/^{47}Ti)_{rutile} - 1] \times 10^4$) than previous studies[8,9,11,12] (Table 1 and Fig. 1). In agreement with earlier work[8], we found that terrestrial rocks have constant titanium isotopic composition with an $\varepsilon^{50}Ti$ value of $+0.01 \pm 0.01$ (average weighted by uncertainties, $n = 19$), whereas bulk meteorites show a spread in $\varepsilon^{50}Ti$ values of $\sim 6\,\varepsilon$-units.

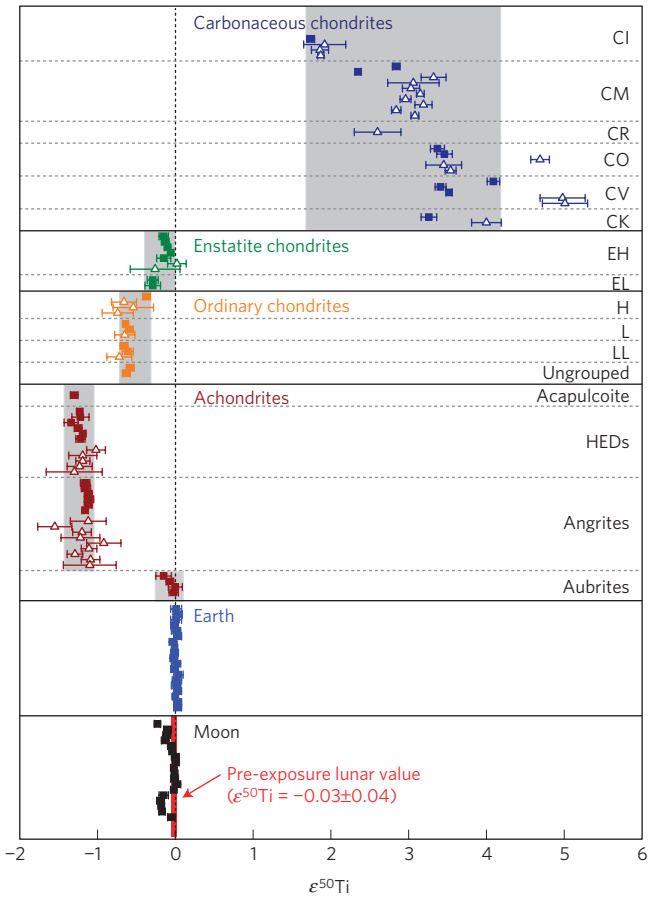

Figure 1 | Titanium nucleosynthetic heterogeneity, $\varepsilon^{50}Ti = [(^{50}Ti/^{47}Ti)_{sample}/(^{50}Ti/^{47}Ti)_{rutile} - 1] \times 10^4$, for carbonaceous, enstatite, ordinary chondrites, and achondrites. The filled and open symbols are from this study and ref. 8, respectively, after internal normalization to $^{49}Ti/^{47}Ti = 0.749766$ (refs 8–10). The uncertainties are 95% confidence intervals (2SE; see Supplementary Information for details). Grey areas cover the ranges of $\varepsilon^{50}Ti$ values for each meteorite group in this study. The red area indicates the pre-exposure lunar $\varepsilon^{50}Ti$ value of -0.03 ± 0.04 from extrapolation of the linear correlation between $\varepsilon^{50}Ti$ and $^{150}Sm/^{152}Sm$ (see Fig. 2a for details).

The majority of lunar samples have titanium isotopic compositions identical to terrestrial samples within the level of

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Table 1 | ε^{50}Ti values for meteorites, terrestrial and lunar samples.

Sample name/no.	Rock/phase	ε^{50}Ti	Sample name/no.	Rock/phase	ε^{50}Ti
Terrestrial samples			**Achondrites (AC)**		
19168	Ilmenite	+0.01 ± 0.33	Ibitira	Eucrite	−1.19 ± 0.04
BCR-2	Basalt	−0.01 ± 0.02	Tatahouine	Diogenite	−1.22 ± 0.06
BIR-1	Basalt	+0.01 ± 0.02	NWA 4801	Angrite	−1.16 ± 0.06
AGV-2	Andesite	+0.03 ± 0.05	NWA 2999	Angrite	−1.13 ± 0.04
Utah Ti after column chemistry	Ti standard	+0.03 ± 0.05	D'Orbigny	Angrite	−1.11 ± 0.06
Carbonaceous chondrites (CC)			NWA 6291	Angrite	−1.12 ± 0.05
Orgueil	CI1	+1.74 ± 0.05	Sahara 99555	Angrite	−1.16 ± 0.04
Murray	CM2	+2.84 ± 0.05	Cumberland Falls	Aubrite	−0.15 ± 0.10
NWA 801	CR2	+2.35 ± 0.04	Bishopville	Aubrite	−0.07 ± 0.05
Ornans	CO	+3.37 ± 0.09	Norton County	Aubrite	0.00 ± 0.09
Lancé	CO3	+3.46 ± 0.10	Peña Blanca Spring	Aubrite	−0.02 ± 0.06
Leoville	CV3	+4.09 ± 0.08	**Lunar samples**		
Allende	CV3	+3.49 ± 0.04	15556—low-Ti basalt	WR	−0.23 ± 0.04
Karoonda	CK4	+3.26 ± 0.10	15016—low-Ti basalt	WR	−0.10 ± 0.05
Ordinary chondrites (OC)			15065,7—low-Ti basalt	Ilmenite	−0.11 ± 0.05
Kesen	H4	−0.37 ± 0.05	14310—low-Ti basalt	Opx	−0.11 ± 0.04
Ausson	L5	−0.64 ± 0.03	72155,37—high-Ti basalt	WR	−0.05 ± 0.05
Isoulane-n-Amahar	L6	−0.59 ± 0.05		Ilmenite	−0.02 ± 0.03
Krymka	LL3.2	−0.66 ± 0.05	70215,66—high-Ti basalt	Ilmenite	+0.01 ± 0.04
Paragould	LL5	−0.61 ± 0.07	75055,40—high-Ti basalt	Ilmenite	−0.02 ± 0.04
NWA 5717_ Lithology A	Ungrouped OC	−0.58 ± 0.05	70017,27—high-Ti basalt	WR	−0.01 ± 0.04
NWA 5717_ Lithology B	Ungrouped OC	−0.63 ± 0.05		Ilmenite	−0.01 ± 0.05
Enstatite chondrites (EC)			71055,45—high-Ti basalt	WR	+0.02 ± 0.05
Sahara 97072	EH3	−0.15 ± 0.06		Ilmenite	−0.02 ± 0.04
Indarch	EH4	−0.13 ± 0.05	75075,30—high-Ti basalt	WR	−0.05 ± 0.05
Adhi Kot	EH4	−0.10 ± 0.04	14321—breccia	WR	−0.04 ± 0.05
Abee	EH4	−0.06 ± 0.04	79155,36—partially glass-coated gabbro	WR	−0.13 ± 0.05
Saint-Sauveur	EH5	−0.15 ± 0.09	60004	Soil	−0.21 ± 0.07
Hvittis	EL6	−0.29 ± 0.07	14163,62	Soil	−0.10 ± 0.06
Jajh deh Kot Lalu	EL6	−0.29 ± 0.10	15001	Soil	−0.15 ± 0.06
Achondrites (AC)			15002	Soil	−0.16 ± 0.05
Acapulco	Acapulcoite	−1.30 ± 0.05	15003	Soil	−0.14 ± 0.05
Kapeota	Howardite	−1.23 ± 0.04	15004	Soil	−0.19 ± 0.05
Millbillillie	Eucrite	−1.22 ± 0.11	15005	Soil	−0.18 ± 0.04
Camel Donga	Eucrite	−1.34 ± 0.09	15006	Soil	−0.17 ± 0.04
Juvinas	Eucrite	−1.25 ± 0.05	76501	Soil	−0.04 ± 0.05

Uncertainties for all samples are 95% confidence intervals (2SE). WR: whole rock. A more extensive data table is provided in Supplementary Information.

uncertainties (Table 1). However, several samples show measurable departures from terrestrial composition, with ε^{50}Ti deficits reaching −0.23 ± 0.04 for Apollo 15556 (low-Ti basalt). Leya *et al.*[9] had found homogeneous titanium isotopic composition between the Earth and Moon but their uncertainties were up to 15 times larger than ours, so the negative anomalies that we detected could not have been resolved. Trinquier *et al.*[8] measured only one lunar sample and found terrestrial isotopic composition (within ±0.32 ε-unit using a Neptune MC-ICPMS and ±0.13 using an Axiom MC-ICPMS). However, the variability in ε^{50}Ti values of lunar rocks documented here poses the question of the representativeness of a single sample measurement.

Lunar rocks and soils have been bombarded by cosmic rays for extended periods of time. These cosmic rays generated secondary neutrons, which could have modified the titanium isotopic composition. Some isotopes of samarium and gadolinium have large thermal neutron capture cross-sections and are therefore useful tracers of neutron exposure in the lunar regolith[13–18]. We found that the deficits in the ^{50}Ti/^{47}Ti ratios correlate with the most neutron-sensitive ratios, ^{150}Sm/^{152}Sm and ^{158}Gd/^{160}Gd (Fig. 2), which were measured previously in lunar samples (see Supplementary Table S1; refs 13–18). Weighted linear regressions using the statistics program R give slopes of −66 ± 24 and −35 ± 20, respectively (Fig. 2a,b). Similar correlations were found in four aubrites (a group of achondrites), which are known to have long cosmic-ray exposure histories[19,20]. These correlations demonstrate that the ε^{50}Ti deficits in some lunar samples were produced by neutron capture effects. We modelled the isotopic shifts expected from exposure to cosmic rays (see Methods and Supplementary Information for details) and found that the most important reaction is the capture of fast neutrons on ^{48}Ti to produce ^{49}Ti, which affects the ^{50}Ti/^{47}Ti ratio by way of the internal normalization scheme used for mass fractionation correction (ref. 10). Modelling predicts that the effects on ε^{48}Ti and ε^{46}Ti are two to four times smaller than that on ε^{50}Ti and are therefore not resolved well with our current precision (see Supplementary Fig. S1). The model reproduces the correlations between ε^{50}Ti and ^{150}Sm/^{152}Sm or between ε^{50}Ti and ^{158}Gd/^{160}Gd reasonably well (Fig. 2). The slight mismatch may be due to the fact that neutron capture on titanium is affected by higher energy fast neutrons than on samarium and gadolinium, which are both more sensitive to low-energy thermal neutrons. A similar effect has

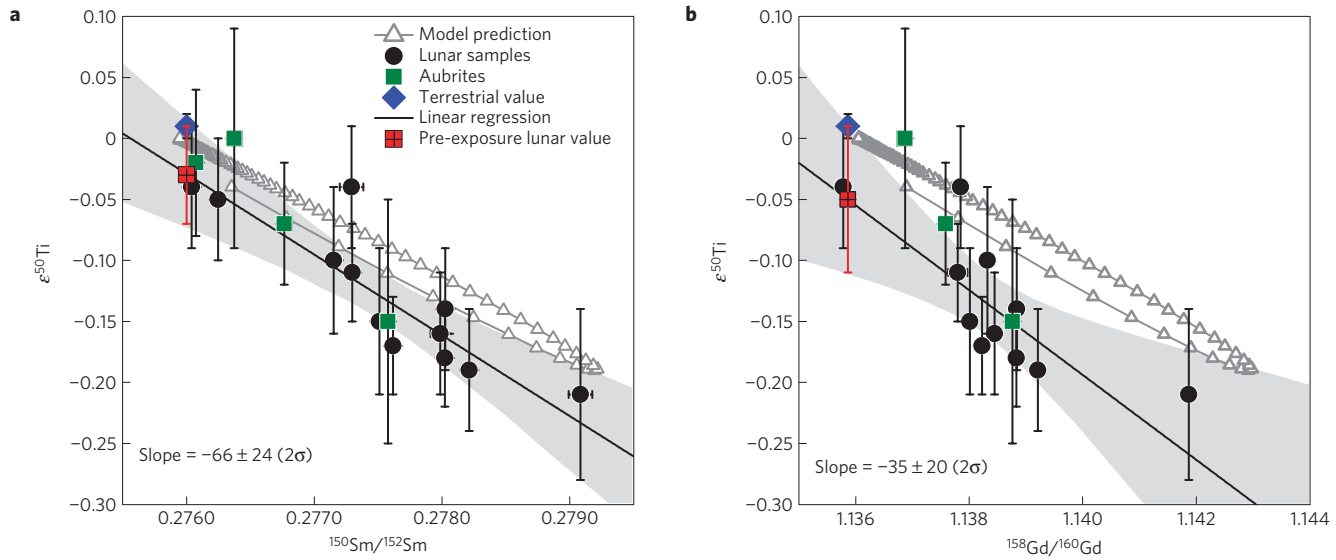

Figure 2 | Correction of cosmogenic Ti isotope effects in lunar samples. ε^{50}Ti versus ^{150}Sm/^{152}Sm (**a**) and ε^{50}Ti versus ^{158}Gd/^{160}Gd (**b**) for terrestrial, lunar, aubrite samples, and model predictions. The measured ^{150}Sm/^{152}Sm and ^{158}Gd/^{160}Gd ratios are from refs 13 to 20 (most of the error bars are smaller than the symbol size). The black solid lines are the best linear fits for lunar samples with 95% confidence envelopes (grey areas). The red square with a cross inside represents the pre-exposure lunar value. The depth in model prediction increases in an anticlockwise loop from the surface (0 m) to 5 m with a step size of 0.04 m.

been documented previously for neutron capture on ^{113}Cd, which is also more strongly affected by fast neutrons than samarium and gadolinium[15].

We extrapolated the linear correlations for the lunar samples shown in Fig. 2a,b to the terrestrial values of ^{150}Sm/^{152}Sm (0.276002 ± 0.000002) and ^{158}Gd/^{160}Gd (1.135861 ± 0.000008; ref. 14), yielding ε^{50}Ti values corrected for cosmic-ray exposure of -0.03 ± 0.04 (Fig. 2a) and -0.05 ± 0.06 (Fig. 2b), respectively, either of which is identical to the weighted average terrestrial ε^{50}Ti value of $+0.01 \pm 0.01$ (see Supplementary Table S1; the Sm-extrapolated ε^{50}Ti value is plotted in Fig. 1 as the pre-exposure lunar value). Thus, the Earth and the Moon have identical ε^{50}Ti values within $\pm 0.04\,\varepsilon$-unit. For comparison, bulk meteorites have ε^{50}Ti values that span $6\,\varepsilon$-units (Fig. 1; refs 8,10).

The isotopic homogeneity between the Earth and the Moon for a highly refractory element such as titanium has important implications for the giant-impact scenario. One possibility to explain this homogeneity is that the majority of the Moon-forming material came from the proto-Earth instead of the impactor. However, this contradicts predictions of state-of-the-art giant impact simulations, which indicate that at most 60% of the Moon-forming material could originate from the proto-Earth[3]. Taking this fraction at face value, this would imply that Theia had the same ε^{50}Ti value as Earth to within $0.10\,\varepsilon$-unit ($0.04\,\varepsilon$-unit/40%). The commonly accepted view of the growth of the Earth is that it proceeded over several tens of million years by collision and accretion of Moon- to Mars-sized planetary embryos[21]. During this stage of chaotic growth, radial mixing of material from different regions of the disk took place and there is a priori no reason to expect that Theia should have the same isotopic composition as the proto-Earth. For the sake of argument, let us assume that Theia had the same isotopic composition as Mars, which may be a stranded planetary embryo[22]. Given that Martian meteorites have $\varepsilon^{50}\text{Ti} = \sim -0.8$ measured using a Neptune MC-ICPMS (ref. 8), the maximum fraction of the material contribution from Theia would be ~8.8% [$(-0.07)\,\varepsilon$-unit/$(-0.8)\,\varepsilon$-unit; −0.07 is the lowest possible lunar value given the inferred value of -0.03 ± 0.04]. Otherwise, one would expect that the Moon has $\varepsilon^{50}\text{Ti} \sim -0.3$ (that is, $-0.8\,\varepsilon$-unit ×40%), which would have been readily resolved with our precision. The same argument can be made with oxygen, although this element would have been more easily exchanged between Earth's mantle and the protolunar disk in the aftermath of the giant impact[23]. A model proposed recently to explain the small mass of Mars is that the terrestrial planets were formed in a narrow annulus of matter truncated at 1 AU by the inward then outward migration of Jupiter[24]. Accordingly, the proto-Earth mantle and Theia may have been made from similar materials and could have shared the same isotopic compositions. An argument against this idea is given by the identical tungsten isotopic compositions of lunar metals and Earth's mantle[25]. Interpretations of the tungsten isotope results are difficult because ^{182}W/^{184}W variations are produced by the decay of the short-lived nuclide ^{182}Hf and there is still significant uncertainty on the Hf/W ratios of the terrestrial and lunar mantles[26]. With this caveat in mind, it is likely that the mantles of Theia and the proto-Earth would have followed different ^{182}Hf-^{182}W paths and their tungsten isotopic compositions at the time of impact were probably different. Although the idea that Theia and the proto-Earth had identical compositions cannot be definitely ruled out[4], this idea seems to be contrived and requires special circumstances for an embryo to have the same titanium, oxygen, and tungsten isotopic compositions as a growing planet.

An alternative scenario is that the mantles of the Earth and the Moon achieved isotopic homogenization in the aftermath of the Moon-forming impact. Pahlevan and Stevenson[23] evaluated this scenario quantitatively for oxygen and concluded that chemical exchange could have taken place between the terrestrial mantle and the protolunar disk. A critical question that is posed by our results is whether such a model could explain the observed homogeneity for a highly refractory element such as titanium. In particular, the timescale for exchange between the magma disk and the vapour atmosphere (τ_{ex}) is sensitive to element volatility as it depends on the vapour pressure of the element considered (P_v):

$$\tau_{ex} = \frac{C\sigma}{P_v}\sqrt{\frac{2\pi RT}{m}} \quad (1)$$

where C is the element concentration in the magma disk, R is the gas constant, σ is the surface mass density of the disk (5×10^7 kg m^{-2}; ref. 23), T is the temperature (3,000 K; ref. 23) and m is the molar mass of gas species. We calculated the timescales for the moderately volatile elements chromium, silicon and magnesium, using relevant thermodynamic data, and obtained values on the order of weeks (see Supplementary Information for details), consistent with the calculated timescale of ref. 23. For tungsten, titanium and calcium, we obtained timescales of ~0.2, ~1 and ~30 years, respectively. These timescales are one or two orders of magnitude higher than those for moderately volatile elements. Whether titanium isotope anomalies were homogenized between the Earth and the Moon depends critically on the lifetime of the magma disk. N-body simulations predicted that once disk material is condensed, Moon-formation can take place in about a month to a year[27]. However, the timescale for cooling of the disk may have been longer, up to 100–1,000 years[23,28]. Thus, even for highly refractory elements, the rate-limiting step may not have been magma–atmosphere exchange but rather large-scale turbulent transport across the disk. The hypothesis of turbulent mixing needs quantitative examination on dynamical aspects, especially regarding exchange of mass and angular momentum during the mixing process. Turbulent mixing requires that the proto-Earth sheds mass at least at its equator; however, the angular momentum of the proto-Earth would be too small to sustain this process[29].

Other scenarios are worth exploring, such as formation of the Moon by fission from a rapidly rotating Earth following a giant impact[30] or by the collision of an icy embryo formed in the outskirts of the protoplanetary disk onto the proto-Earth[3]. A long-standing difficulty with the fission model is that the angular momentum of the Earth–Moon system concentrated into the proto-Earth would leave the Earth spinning insufficiently rapidly to allow fission[29]. However, it has recently been suggested that this angular momentum could have been dissipated through resonances among the Moon, the Earth's core and the Sun. Further work remains to be done to assess this possibility. Alternatively, an icy impactor that formed beyond the snow line would have delivered water, but would have delivered minimal amounts of rock-forming elements such as titanium to the Earth–Moon system. The water–steam disk created by such an impact may have promoted oxygen isotope equilibration between Earth's mantle and the protolunar disk. The feasibility of such a scenario remains to be evaluated from a dynamical point of view. In all instances, the isotopic homogeneity of refractory elements in the Earth–Moon system is a fundamental new constraint to lunar-formation theories.

Methods

Experimental methods. Approximately 1–10 mg of powdered terrestrial/lunar sample or ~100 mg of powdered bulk meteorite were dissolved. After dissolution, titanium was separated through a two-stage procedure using Eichrom TODGA and Bio-Rad AG1-X8 resins. On TODGA, titanium was collected with molybdenum and minor niobium, tantalum and tungsten in 10 ml of 12 M HNO_3 + 1 wt% H_2O_2. On AG1-X8, titanium was separated from molybdenum, niobium, tantalum and tungsten with 10 ml of 9 M HCl + 0.01 M HF. The sample solution was evaporated and redissolved in 0.3 M HNO_3 + 0.0015 M HF. All the measurements were done by means of an Aridus desolvating nebulizer on a Thermo Neptune MC-ICPMS at the University of Chicago. The uncertainties are based on repeat analyses of a sample by standard bracketing (95% confidence intervals of the average of $n = 14$–18 repeat analyses; that is, 2 standard errors). Measurements of geostandards and terrestrial samples yielded normal titanium isotopic compositions within uncertainties, demonstrating that the measurements were accurate. Full methods can be found in Supplementary Information and ref. 10.

Model of cosmic-ray effect. The production rates were calculated by folding the flux densities of the projectiles with the excitation functions of the relevant nuclear reactions. We approximated the flux densities in the lunar surface by those of an L-chondrite with a radius of 5 m. Note that this approximation is reliable for thermal and epithermal neutrons, as demonstrated by comparing measured and modelled shifts in samarium and gadolinium isotopic compositions. The thermal neutron capture rates on samarium and gadolinium were calculated using the cross-sections from the JEFF-3.0/A database. For the cosmogenic production of titanium isotopes, we considered thermal neutron capture effects on titanium (burnout and production within titanium isotopes), and thermal neutron capture on ^{45}Sc, ^{46}Ca and ^{48}Ca to produce ^{46}Ti, ^{47}Ti and ^{49}Ti, respectively. Also, we considered the spallogenic production of titanium isotopes from iron and nickel, and the production and burnout of titanium isotopes by $(n, 2n)$ reactions (for example, the production of ^{47}Ti from ^{48}Ti by medium energy neutrons).

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Author contributions

J.Z., N.D. and A.M.D. planned the project, J.Z. measured the Ti isotopic compositions of all samples, I.L. provided model predictions of cosmogenic effects. A.F., J.Z., N.D. and A.M.D. calculated the timescale for evaporative exchange. All authors contributed to discussion, interpretation of the results and writing of the manuscript.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturegeoscience. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to J.Z.