



## Supplementary Materials for

### **Identification of the giant impactor Theia in lunar rocks**

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## Materials and Methods

Oxygen isotope analyses have been conducted in dual inlet mode on a MAT253 at the University of Göttingen following a novel protocol for sample O<sub>2</sub> purification (19). Sample oxygen was liberated by means of IR laser fluorination using F<sub>2</sub> gas as a reacting agent. The liberated sample gas was passed through heated NaCl (t = 150°C) and liquid nitrogen cold traps to remove excess F<sub>2</sub> and condensable gases. To remove NF<sub>3</sub> and CF<sub>4</sub>, the sample gas was then passed through a gas chromatograph column, using He as a carrier gas. The purified O<sub>2</sub> was collected on a 5Å molecular sieve held at liquid nitrogen temperatures and was then introduced into the bellow of the mass spectrometer (19). Samples were analyzed for ca. 90 min relative to bottle O<sub>2</sub> that had been calibrated relative to VSMOW by E. Barkan (The Hebrew University, Jerusalem). Note that literature data is not normally reported on true VSMOW scale (19, 32), but literature data presented herein is recalculated accordingly. External reproducibility (1SEM) was < 0.1 ‰ for δ<sup>18</sup>O and δ<sup>17</sup>O and precision of Δ<sup>17</sup>O was ca. 5 - 8 ppm for a single analysis. This is similar to the precision routinely achieved for water samples (33).

The advantage of our method is the purification technique of the sample gas combined with long measurement times in dual inlet mode (19). Especially NF<sub>3</sub> is a problematic gas species. A fragment of this gas (NF<sup>+</sup>) is produced in the source of the mass spectrometer and interferes on the <sup>16</sup>O<sup>17</sup>O<sup>+</sup> signal. Because <sup>17</sup>O is the rare isotope, small amounts of NF<sup>+</sup> can bias <sup>17</sup>O measurements. Traditionally, NF<sub>3</sub> is separated from the O<sub>2</sub> by cryogenic techniques (34). This method is not always perfect and the gas is normally scanned for NF<sub>2</sub><sup>+</sup> fragments prior to measurements. If no NF<sub>2</sub><sup>+</sup> is observed, the measurements are performed. Minute quantities of NF<sub>3</sub> may not be detected by this approach and the operator must make a decision if the gas is clean or not. If more care is taken with Apollo samples over terrestrial samples, this may induce a bias. Hypothetically operators have been slightly more careful when analyzing Apollo samples. This would reduce the average Δ<sup>17</sup>O composition measured for the Moon and explain the marginally reduced Earth-Moon variation of the literature data over our data. By using a GC column perfect separation of NF<sub>3</sub> and other problematic gasses like CF<sub>4</sub> can be guaranteed for this study.

## Supplementary Text

Here we give additional information on the definition of Δ<sup>17</sup>O and why we do not report our data relative to a terrestrial fractionation line (TFL). The O and Ti isotope data of (8-11) is discussed in more detail and additional data for lunar meteorites is presented. We evaluate the possible oxygen isotopic composition of the impactor and of proto-Earth. A chemical mass balance for refractory elements is presented.

### 1. Definition of Δ<sup>17</sup>O.

The triple oxygen isotope composition is reported in δ<sup>18</sup>O vs. δ<sup>17</sup>O space as in (19). We chose a reference line with a slope θ of 0.5305 that passes through the origin, which is defined by VSMOW. Deviations from this reference line are expressed as Δ<sup>17</sup>O = δ<sup>17</sup>O - 0.5305\* δ<sup>18</sup>O. The slope θ is identical to the theoretical high temperature equilibrium

limit (21) and within error identical to the high temperature slope defined by minerals in the San Carlos lherzolite ( $\theta = 0.529 \pm 0.006$  see ref. 19) and the slope of all our mantle minerals ( $\theta = 0.532 \pm 0.006$ ). Our reference line is not defined by the terrestrial rocks and minerals data.

## 2. Why do we not report data relative to a 'terrestrial fractionation line' (TFL)?

The first  $\Delta^{17}\text{O}$  values published were reported as deviations from a TFL defined by rocks, minerals and waters in  $\delta^{18}\text{O}$  vs.  $\delta^{17}\text{O}$  space (5). It was assumed that any mass dependent fractionation process plots on a slope of ca. 0.5. According to quantum mechanics however, various mass dependent processes result in slightly different slopes. A comprehensive outline on this theory is given by Young et al. (21), but the variable slopes were already anticipated by Matsuhisa et al. in 1978 (ref. 35), who suggested using 0.52 as a compromise mean value. Regardless of Matsuhisa's prediction, many laboratories have published TFL slopes (between 0.52 and 0.526) assuming that all samples from the Earth plot on a single line. The observed variability in TFL arrays reflects the expected variability of  $\Delta^{17}\text{O}$  in the Earth's crust (19, 21, 35). With the increasing number of published TFL slopes, the definitions of  $\Delta^{17}\text{O}$  have increased, so that values between laboratories are not comparable at high precision. To make things worse it was recently demonstrated that a rocks and minerals defined TFL does not pass through the origin (i.e. VSMOW) and that  $\Delta^{17}\text{O}$  data for silicates are therefore often not reported on VSMOW scale (32).

We suggest abandoning the concept of using a TFL defined by minerals and rocks as a reference slope and to use the high temperature equilibrium limit (0.5305) with zero intercept as a reference line instead (19). The reasons are: i) 0.5305 is a physically significant figure. ii) High temperature fractionation of mantle minerals and melts will occur close to 0.5305 (see Fig 1). iii) The slope of an arbitrary set of samples (normal TFL) has no meaning. iv) Deviations ( $\Delta^{17}\text{O}$ ) from a rocks and minerals defined TFL that does not pass through the origin will not be on VSMOW scale (32).

Using a different slope will change the absolute numbers of  $\Delta^{17}\text{O}$ , but it will not affect the position of a data point in  $\delta^{18}\text{O}$  vs.  $\delta^{17}\text{O}$  space. Note that  $\Delta^{17}\text{O}$  is simply introduced to better display small deviations of a data point from a chosen reference line. It is a calculated quantity and not a measured value. The measured values are  $\delta^{17}\text{O}$  and  $\delta^{18}\text{O}$ . For this study it is irrelevant which slope is used, because the BSE and BSM have identical  $\delta^{18}\text{O}$ . Both BSE and BSM are points in  $\delta^{18}\text{O}$  vs.  $\delta^{17}\text{O}$  space, and not lines. These points differ in  $\delta^{17}\text{O}$  but not in  $\delta^{18}\text{O}$ . Due to the small variations, our results are not significantly affected if normal  $\delta$  notations are preferred over  $\delta'$  notations.

## 3. Reevaluation of lunar $\Delta^{17}\text{O}$ isotope data.

Previous high precision oxygen isotope studies (8–10) did not resolve a difference between the Earth and the Moon. However, on average lunar samples from these studies plot 9 ppm above the UWG-2 garnet standard (Fig. S1). This average includes lunar meteorites, highland rocks and volcanic glasses. If only basalts ( $n = 58$ ) are considered, the lunar average remains at  $9 \pm 3$  ppm ( $2\sigma$  SEM) above UWG-2 (Fig. S1). We consider UWG-2 to be identical to BSE (Table 1). The slightly lower literature datum ( $\sim 9$  ppm)

over ours (~12 ppm) agrees within uncertainty, but may also be related to a very small analytical artifact (see above).

Most studies focused on lunar basalts (9, 10, *this study*), but Wiechert et al. (8) also analyzed highland rocks. The highland rocks comprise a lower  $\Delta^{17}\text{O}$  (3 ppm above UWG-2) than lunar basalts from the same study (11 ppm above UWG-2). The reason for this discrepancy is unclear and requires further inspection. If real it may point towards a heterogeneous, possibly stratified Moon (36).

Previous studies generally used shallower reference slopes (termed terrestrial fractionation lines or TFL). This is insignificant for comparing  $\Delta^{17}\text{O}$  if samples comprise broadly similar  $\delta^{18}\text{O}$  (see above and ref. 10).

The slightly elevated  $\Delta^{17}\text{O}$  composition of La Paz Antarctic meteorites (-0.08 ‰; (9)) can be explained by interaction with meteoric water that comprises elevated  $\Delta^{17}\text{O}$  compositions relative to terrestrial rocks (19, 32, 33).

#### 4. The $\Delta^{17}\text{O}$ of lunar meteorites.

Data for lunar meteorites is presented in Fig. S2 and Table S1. These samples were not pretreated prior to measurements. Lunar meteorites plot on a weathering trend. Increasing  $\delta^{18}\text{O}$  is reflecting increasing sample alteration. The  $\Delta^{17}\text{O}$  is decreasing with increasing alteration. The shallow slope of the weathering trend is consistent with a low temperature process.

#### 5. Reevaluation of lunar $\epsilon^{50}\text{Ti}$ isotope data.

In a recent high precision  $^{50}\text{Ti}$  isotope study Zhang and co-authors (11) argue that Earth and Moon are identical within errors. However, the values for Earth and Moon are only within error of another at the 95% confidence level. Reassessing the published Ti isotope data ((11); excluding lunar soil samples due to possible contamination by meteorites) indicates that Earth and Moon differ in  $\epsilon^{50}\text{Ti}$ . With this data treatment the Moon ( $\epsilon^{50}\text{Ti} = -0.05 \pm 0.02$ ; 1 $\sigma$  SEM) falls between Earth ( $\epsilon^{50}\text{Ti} = 0.010 \pm 0.005$ ; 1 $\sigma$  SEM) and EH ( $\epsilon^{50}\text{Ti} = -0.13 \pm 0.02$ ; 1 $\sigma$  SEM) or EL ( $\epsilon^{50}\text{Ti} = -0.29$ ) chondrites (Fig. S3). From the Ti isotope perspective, the Moon could comprise  $44 \pm 9$  % of an EH like impactor or ~20 % of an EL like Theia. Note that the EC  $\epsilon^{50}\text{Ti}$  data is not corrected for possible cosmogenic Ti isotope effects.

#### 6. A chemical mass balance.

To the first order and in regard to refractory lithophile elements, the silicate Moon is considered a chemical mixture between proto-Earth and Theia. If Theia compositionally resembled EC, an EC chemical imprint is expected for the Moon. Unfortunately, the bulk composition of the Moon is not well constrained, because i) mantle samples are not available and ii) the average composition of lunar crust as well as the respective mass fractions are debated. Published lunar compositions (e.g. 36-39) are therefore model-dependent approximations. Nevertheless, estimates on lunar concentrations of Ca, Al, Mg, Si, Cr, and Ti would roughly agree with a mix of 43% EH or 30% EL chondrites (22) with the remaining material sourcing from the proto-Earth (40) (Fig. S4). If the

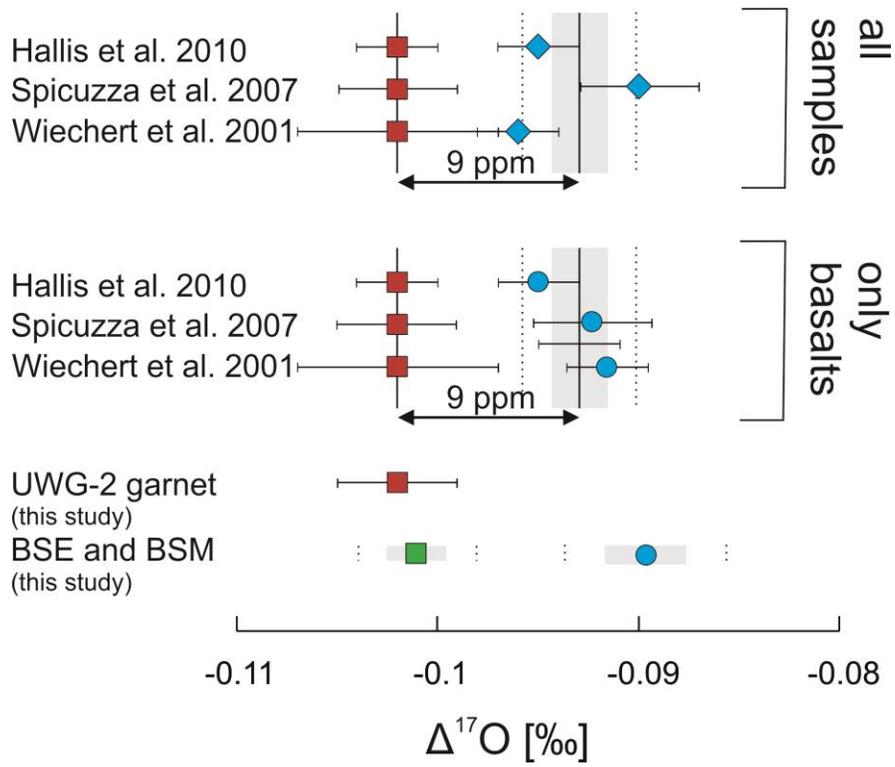
Moon is indeed a mixture of proto-Earth and Theia with EC-like composition, simple mass balance for refractory elements will provide a good starting point for estimating the bulk lunar composition (e.g., using the chemical model of O'Neil (39)).

#### 7. The $\Delta^{17}\text{O}$ composition of Theia.

A simple model for the oxygen isotope composition of potential impactors is provided by Pahlevan and Stevenson (4) (Fig. S5). Because the true distribution of oxygen isotopes in the early inner solar system is unknown, (4) simply assumed a linear distribution with Mars and Earth defining the line. Thus, the model only serves as a first-order approximation.

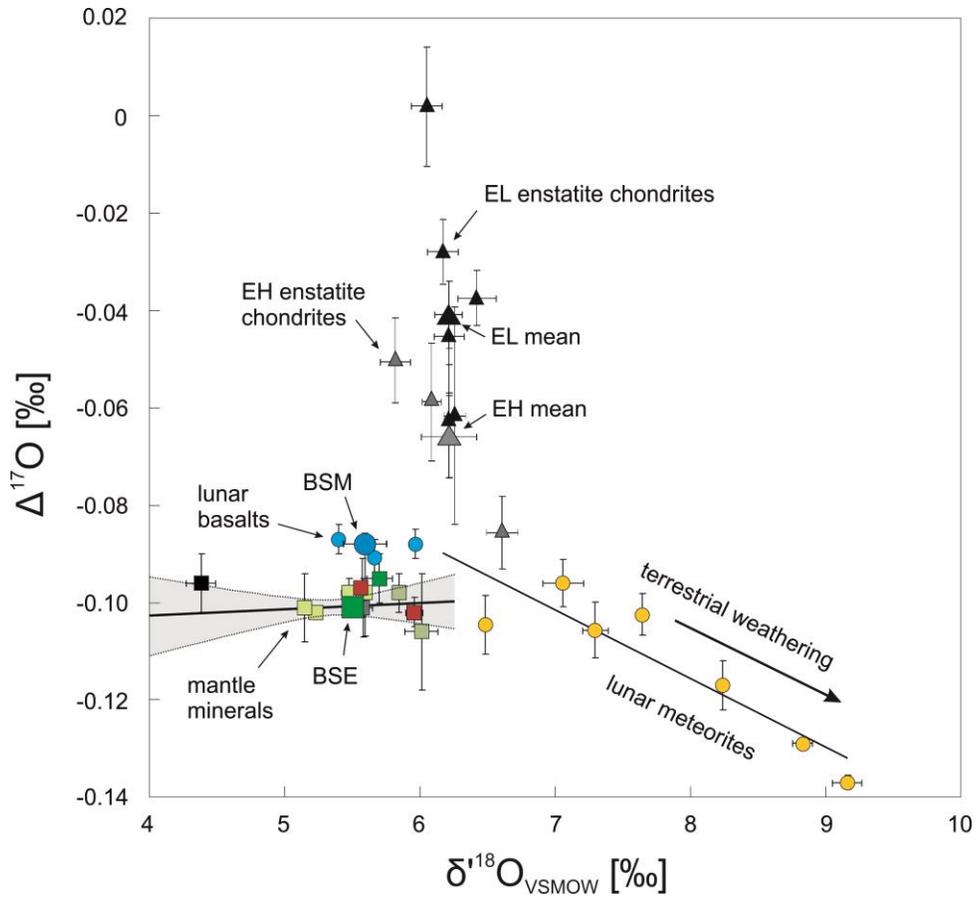
#### 8. The Oxygen isotopic composition of proto-Earth

The composition of proto-Earth was altered by the impactor. The effect was modeled as a function of Theia's mass and composition (Fig. S6). Ordinary chondrites (LL, L and H) significantly alter the proto-Earth composition, even if the impactor was small. For large impactor's a slight variation in  $\Delta^{17}\text{O}$  between proto-Earth and Theia is sufficient for a significant effect. Impactor to proto-Earth mass ratios ( $m_{\text{Theia}}/m_{\oplus}$ ) were adopted from numerical models.



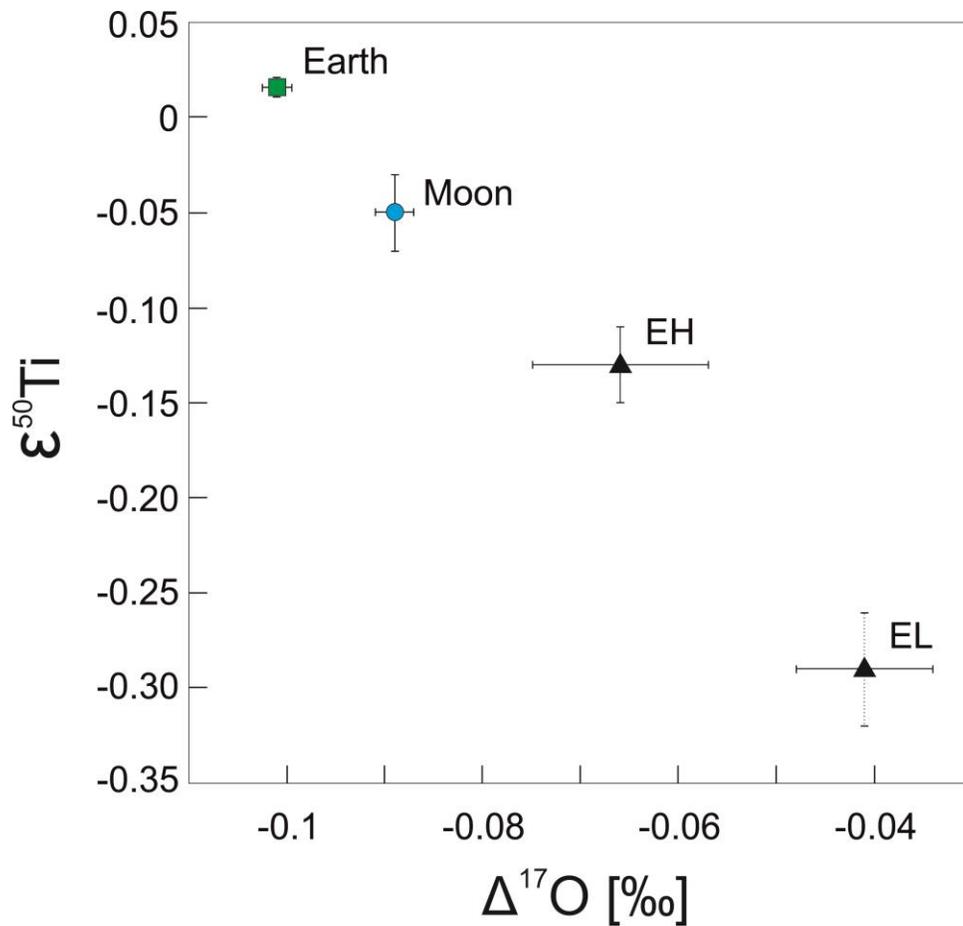
**Fig. S1.**

Literature data was recalculated to VSMOW scale (19, 31) using the published UWG-2 garnet values of the respective studies (8–10). Mean lunar values from all high precision studies (8–10) are elevated with respect to UWG-2 garnet, which is identical to BSE. Error bars and grey envelopes are  $1\sigma$  SEM.



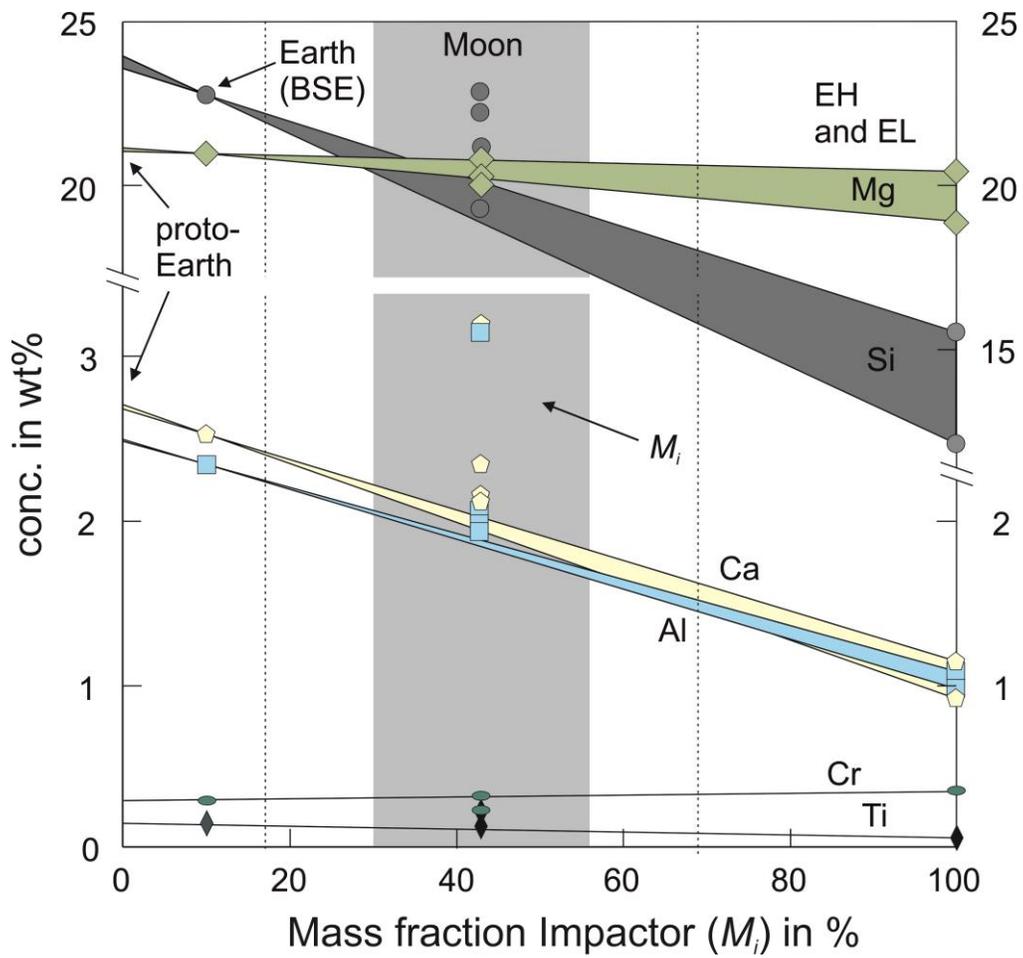
**Fig. S2**

Terrestrial (squares) and lunar (circles) samples as well as enstatite chondrites (triangles) in  $\delta^{18}\text{O}$  vs.  $\Delta^{17}\text{O}$  space. Lunar meteorites (yellow circles) are all finds and have been weathered by meteoric waters in desert environments. They form a weathering array with a shallow slope of 0.5164, which is consistent with a low temperature process. However, this slope is probably not related to one single process because the individual meteorites stem from various desert environment's and interacted with variable water compositions.



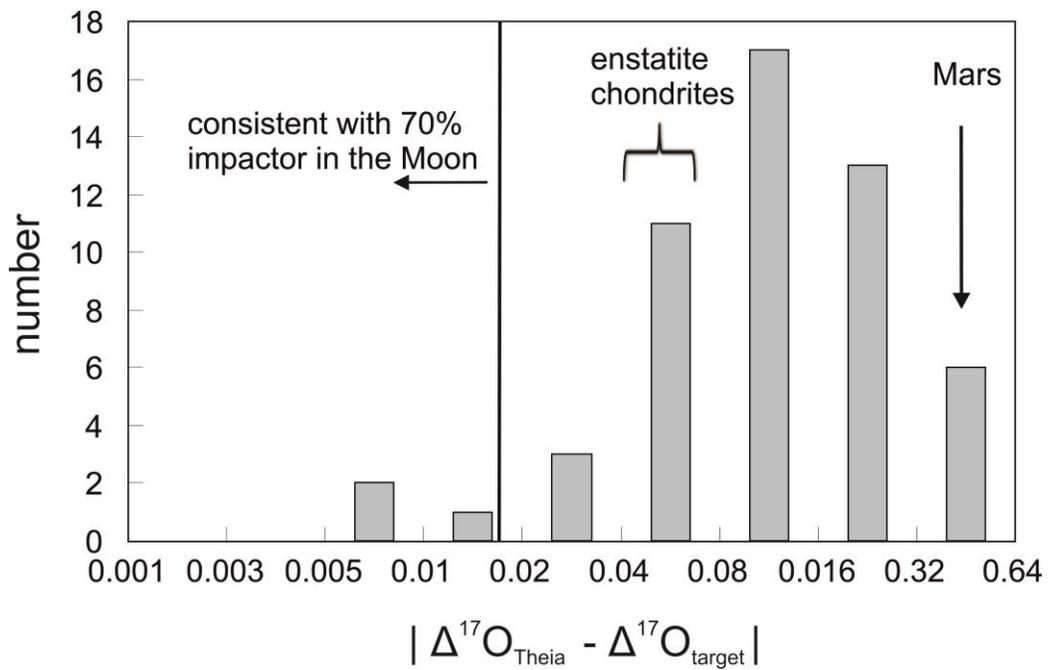
**Fig. S3**

Plot of  $\Delta^{17}\text{O}$  (*this study*) vs.  $\epsilon^{50}\text{Ti}$  (*11*). We have omitted the lunar soil samples from the Ti isotope data of (*11*). Error bars are 1σ standard errors of the mean (1σ SEM). The  $\epsilon^{50}\text{Ti}$  for EL is based on only two datapoints (dotted error bar).



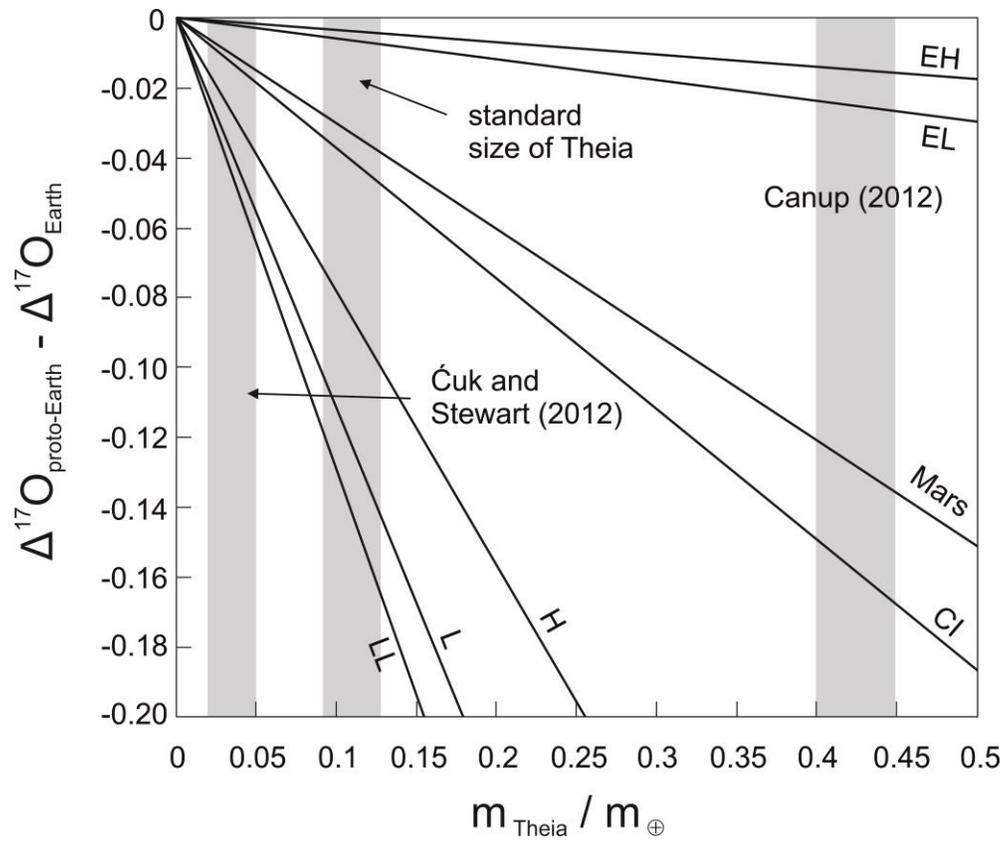
**Fig. S4**

Mass balance between the BSE (40), and enstatite chondrites (22). Estimates for bulk lunar composition (36-39) broadly agree if 43% of an EH impactor is mixed with material from the proto-Earth.



**Fig. S5**

Modeled oxygen isotopic compositions of potential impactors, modified after (4). Impactors with oxygen isotopic compositions similar to enstatite chondrites are not unusual.



**Fig. S6**

Computed proto-Earth  $\Delta^{17}\text{O}$  composition, as a function of  $m_{\text{Theia}}/m_{\oplus}$  for variable impactor compositions.

**Table S1.**

Triple oxygen isotope analysis of lunar meteorites. Definitions are identical to those in Table 1.

<b>Sample</b>	<b>Locality</b>	$\delta^{18}\text{O}$	$\delta^{17}\text{O}$	$\delta'^{18}\text{O}$	<b>SD</b>	$\delta'^{17}\text{O}$	<b>SD</b>	$\Delta^{17}\text{O}$	<b>SD</b>	<b>SEM</b>	<b>N</b>
Kalahari 008	Botswana	9.20	4.74	9.16	0.15	4.72	0.08	-0.137	0.002	0.001	2
Kalahari 009	Botswana	8.87	4.57	8.83	0.11	4.56	0.06	-0.129	0.001	0.001	2
Dho908	Oman	7.09	3.66	7.06	0.21	3.65	0.11	-0.096	0.007	0.005	2
Dho910	Oman	6.50	3.34	6.48	0.03	3.33	0.01	-0.105	0.008	0.006	2
Dhofar 911	Oman	7.33	3.77	7.30	0.16	3.77	0.08	-0.106	0.010	0.006	3
Dho1084	Oman	8.28	4.26	8.24	0.02	4.25	0.02	-0.117	0.016		1
Dho 1085	Oman	7.68	3.96	7.65	0.06	3.96	0.03	-0.102	0.007	0.004	3

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