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Sulfur isotopes from the lunar farside reveal global volatile loss following the giant impact

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The Moon is strongly depleted in volatile elements and exhibits heavier isotopic signatures (e.g., K, Zn) than the Earth. However, the pronounced nearside–farside dichotomy and uneven distribution of volatiles across lunar interior raise the question of whether such heavier isotopic signatures resulted from a global giant impact or local magmatic processes. Here we report high sulfur contents ($1800 \pm 400 \, \mu g/g$) and $\delta^{34}S$ values ($0.83 \pm 0.16\%$, 2SE, n = 17) in Chang'e-6 basalt from lunar farside, with similar $\delta^{34}S$ values in two nonmare crustal clasts. These values fall within the range reported for nearside mare basalts and basaltic meteorites of different ages and mantle sources, indicating a broadly homogeneous $\delta^{34}S$ composition across lunar interior that is -2‰ heavier than the Earth's mantle. This isotopic signature cannot be explained by core formation or late accretion and is best attributed to global volatile loss during the Moon-forming impact. Subsequent magma ocean evolution and mantle overturn drove heterogeneous volatile budget in lunar mantle.

Current models favor that the Moon formed during an impact between a Mars-size planetary body and the proto-Earth, in which the Moon was derived from a mixture of the impactor and the proto-Earth¹. Key evidences for this include the nearly identical relative abundances of refractory lithophile elements in both the Earth and the Moon, along with similar isotopic anomaly signatures in elements such as O, Ti, Cr and Ca²-5. The pronounced volatile depletion with global enrichments in the heavier isotopes (e.g., S, Cl, K, Rb and Zn)6-10 implies strong evaporation during its formation¹11,12.

Although the lunar interior was previously considered to be devoid of volatiles, the discovery of H, F, Cl, and S in volcanic glasses¹³ affected this assumption. Subsequent analyses of these glasses and melt inclusions in Apollo mare basalts have revealed that volatile concentrations could be approaching those of terrestrial mid-ocean ridge basalts (MORB), albeit with significant heterogeneity^{14–18}. Such

heterogeneity may reflect incomplete accretion during the Moon's formation¹ or subsequent evolutionary processes, including mantle overturn¹9.20 or regional degassing²¹. Since isotopes of volatile-element could be fractionated under these conditions, it thus remains unclear whether isotopic data from current lunar sample collections—primarily lunar nearside Procellarum KREEP Terrane (PKT) region—could provide a comprehensive representation of the entire Moon. In particular, samples from the PKT region demonstrate substantial elemental anomalies from the Feldspathic Highlands Terrane and the farside South Pole-Aitken Terrane, such as K, Th and other incompatible elements²². Barnes et al.²¹ proposed that the enrichment in the heavier Cl isotopes in urKREEP required metal—chloride degassing, inferring that at least one large impact event may have exposed the residual Lunar Magma Ocean (LMO) melt at the lunar surface before complete solidification. If such an event occurred, the volatile elements and

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isotopic compositions in all returned samples from the PKT region may have been altered, thereby not necessarily reflecting the Moon's primordial characteristics²³. Radiogenic isotopes (e.g., μ values reflecting ²³⁸U/²⁰⁴Pb ratio) also point to very heterogeneous mantle domains for different mare basalts²⁴.

Sulfur, a moderately volatile element, could be a revealing tracer of these processes. Given high sulfur contents at sulfide saturation (SCSS) for lunar basalts²⁵, S hardly reaches sulfide saturation until the late stage of the lunar magmatic evolution^{25–27}. Therefore, its isotopes in melts would reflect the primary signature, even after complex LMO overturn and mixing of cumulates²⁸. Moreover, previous sulfur isotope studies have shown that mare basalts formed through effusive eruptions experienced minimal degassing (less than 10%) compared to lunar volcanic glasses produced by fire-fountain eruptions^{8,28,29}. Thus the sulfur isotopic composition of mare basalts should closely reflect their parental magmas and mantle sources^{8,29}. Existing nearside samples, such as Apollo, Chang'e-5 (CE-5) and lunar meteorites, exhibit an overall uniform δ^{34} S value of $0.6 \pm 0.3\%$, sheaver than Earth's sulfur isotope composition³⁰. It raises the question of whether farside samples would confirm this homogeneity.

The Chang'e-6 (CE-6) mission returned the first soil samples from a farside mare basalt unit within the Apollo basin³¹. The low-Ti basalt erupted at about 2.8 Ga and originated from a lunar mantle source region that is highly depleted in incompatible elements—a characteristic distinctly different from nearside PKT rocks³²⁻³⁴. Consequently, the CE-6 samples are critically important for elucidating the distribution of sulfur on the Moon and assessing the homogeneity of lunar sulfur isotopes. Additionally, the CE-6 soil sample contains rock clasts from both the mantle (e.g., basalt) and the crust (e.g., anorthositic norite and anorthosite), providing integrated information on sulfur isotopes in the lunar farside.

In this work, we conducted a detailed petrological investigation of troilites in basalts, nonmare clasts, and breccias from the CE-6 samples, and carried in-situ high-precision sulfur isotope analyses of these troilites²⁸. Given that most of the sulfur in lunar basalts is stored in troilite²⁸, the sulfur isotopes in troilite can be taken to broadly represent or at least be comparable with the bulk-rock sulfur isotopic composition. Our new findings confirm a homogenous sulfur isotope of the whole lunar interior, and from a volatile element perspective, offer further constraints on models for the Moon's formation.

Results and discussion

Petrography

The CE-6 low-Ti basalt clasts exhibit various basaltic textures (porphyritic, vitrophyric, poikilitic and subophitic) and are primarily composed of clinopyroxene, plagioclase, ilmenite with the minor occurrence of cristobalite, troilite, and apatite (Fig. 1a, b). The nonmare lithologies in CE-6 samples are predominantly composed of plagioclase, accompanied by orthopyroxene and minor troilite (Fig. 1c, d), and are classified as noritic anorthosite and norite 35,36. According to the Mg# of pyroxene and An# of plagioclases, the two nonmare clasts are chemically closed to Mg suite and Ferroan Anorthosite (FAN) (Fig. S1, Table S1). Polymict breccias contain clasts and rock fragments exhibiting diverse characteristics and complex origins. As illustrated in Fig. 1e, f, basalt clasts, mineral fragments, and impact glasses are all incorporated within the breccia, indicating extensive sputtering and gardening on the CE-6 landing sites.

We selected 40 large basalt clasts from several hundred lunar soil clasts and performed quantitative mineralogical analyses using TESCAN Integrated Mineral Analyzer (TIMA). The results indicate that the CE-6 basalts exhibit 0.31 \pm 0.07 (2SE) vol% troilite (Table S2, Fig. S2a). Taking into account the varying densities of different mineral phases and the 36.5 wt% sulfur content of troilite (FeS), the sulfur content of CE-6 basalts can be estimated to be $1800\pm400~\mu g/g$. Using the same method, the sulfur content of NWA 14526 and NWA 12008 are

estimated to be $260 \pm 130 \,\mu\text{g/g}$ and $400 \pm 200 \,\mu\text{g/g}$ (Table S1). According to Elardo et al.³⁷, the troilites abundance of NWA 4734 is 0.1 to 0.3 vol % and the S content is thus estimated to be between 600 and 1700 $\,\mu\text{g/g}$.

Most sulfide grains are less than 20 μ m in size, with only a few larger than 50 μ m (Fig. S2c). Although the basalt clasts exhibit various textures, the troilites mainly occur interstitially, typically associated with late-stage crystallizing minerals such as ilmenite, plagioclase, apatite, and clinopyroxene characterized by low Mg# (<10) (Fig. S2b). In non-mare rock clasts (Fig. 1c, d), sulfides occasionally appear within plagioclase grains, with grain sizes generally below 10 μ m. We also found large sulfides occurring in the matrix of breccias (Fig. 1e, f).

Composition of CE-6 troilites

In CE-6 clasts, sulfides largely conform to the formula FeS (troilite, Table S2), which is one of the most common accessory minerals in lunar rocks³⁸. Troilites in the basalt clasts display 0.02 to 0.11 wt% Co, <0.04 wt% Ni, and <0.05 wt% Cu. The Co, Ni and Cu contents are within the range of troilites in Apollo basalts³⁸ and the CE-5 basalt²⁸. The composition of troilites is comparable among basalt clasts, nonmare clasts and breccias, except Ni content. Troilites in noritic anorthosite and norite clasts exhibit variably Ni contents, which mainly result from the heterogenous distribution of Ni in microscales (Figs. 2c and S4 and Table S3). In breccias, most troilites Ni concentrations fall within the basaltic range, although some samples display anomalously elevated Ni contents (Fig. S3).

Sulfur isotopes of CE-6 troilites

Nineteen analyses of sulfur isotopes compositions are conducted on troilites in thirteen basaltic clasts by laser-ablation MC-ICP-MS with spot diameter of 8 µm²⁸ (Table S4, See details in Supplementary methods). To further validate our data, we conducted multiple analyses on sufficiently large troilite grains or different grains from the same clast (Figs. 1 and S5). We found that although most sulfur isotope values from different spots lie within the analytical uncertainty, two analyses from a single grain exhibit a larger discrepancy $(1.30 \pm 0.24\%)$ and $2.32 \pm 0.47\%$; Fig. S5c). Notably, in this basaltic clast, troilite is in contact with pyroxene at the clast's edge, while the clast itself is significantly fractured. As this grain may have been affected by potential degassing, it was excluded from the average value calculation. A similar intra-grain isotopic variation has also been observed in sulfides from CE-5 basalts and lunar meteorites^{28,39}. The $\delta^{34}S_{V-CDT}$ value of troilites in basaltic clasts display a mean value of $0.83 \pm 0.16\%$ (2SE, n = 17). In addition, we found that most clinopyroxenes coexisting with troilite have Mg# values below 10, indicating their crystallization during the late stage of magma evolution (Fig. 2b). Notably, the troilite in contact with the most primitive clinopyroxene exhibits a $\delta^{34}S_{V\text{-}CDT}$ value of $0.77 \pm 0.32\%$ (Fig. 2b). The $\delta^{34}S_{V-CDT}$ values of troilites in NWA 14526 and NWA 12008 are $0.84 \pm 0.17\%$ (2SE, n = 10) and $0.29 \pm 0.31\%$ (2SE, n = 7), respectively.

Two nonmare fragments record troilite $\delta^{34}S_{V\text{-}CDT}$ values of $1.14 \pm 0.60\%$ and $0.94 \pm 0.62\%$, respectively (Fig. 2a). In breccias, troilite exhibits $\delta^{34}S_{V\text{-}CDT}$ values ranging from $0.30 \pm 0.40\%$ to $2.19 \pm 0.48\%$ (Fig. 2a). The mean $\delta^{34}S_{V\text{-}CDT}$ value of troilite in breccias, $1.24 \pm 0.31\%$ (2SE, n=14), is slightly higher than that of troilite in CE-6 basalts.

Origin and sulfur isotopes of the troilite in CE-6 samples

The δ^{34} S values of troilite in CE-6 basalt clasts vary from $0.30 \pm 0.51\%$ to $1.52 \pm 0.37\%$. The mean value of $0.83 \pm 0.16\%$ broadly overlaps with those observed in CE-6 nonmare clasts, Apollo basalts, and lunar meteorites (Fig. 2a). In addition, troilite in the CE-6 breccia displays slightly elevated δ^{34} S values, with an average of $1.24 \pm 0.31\%$. In the following sections, we assess the potential influence of meteoritic

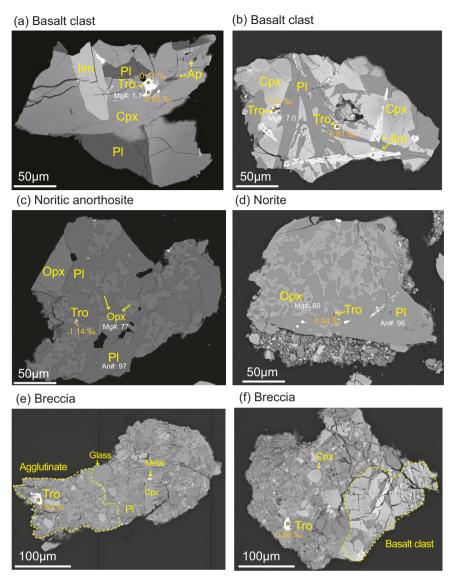


Fig. 1 | Representative lithic clasts in CE-6 soil for sulfur isotope analyses. Large clasts of mare basalts (a, b), noritic anorthosite (c), norite (d) and breccia (e, f) are shown. Orange circles (-8 μm in diameter) mark laser ablation spots for sulfur isotope analyses, with corresponding $\delta^{34}S$ values labeled. The Mg# of

clinopyroxene (Cpx) and orthopyroxene (Opx) closely associated with troilites and An# of plagioclase in anorthositic norite are also indicated. Tro troilite, Pl plagioclase, Cpx clinopyroxene, Opx orthopyroxene, Ap apatite, Ilm ilmenite.

impacts, magmatic differentiation, and degassing processes on these isotopic compositions.

The Ni concentration in troilite from CE-6 basalt clasts falls within the range observed for endogenous troilites in Apollo and CE-5 samples (Fig. S4a), consistent with the low bulk Ni content of the CE-6 basalt (-16.2 $\mu g/g^{36}$). A few troilites in CE-6 breccias exhibit considerable variation in Ni content, reaching 7070–36190 $\mu g/g$ (Figs. 2c and S4a)–levels akin to those observed in troilite from the CE-5 impact melt²8. According to current findings, lunar soil and impact glass represent significant reservoirs of heavy sulfur isotopes on the Moon (δ^{34} S ranging from +4% to +58%), which may be attributed to sulfur evaporation induced by (micro)meteorite impacts or ion sputtering⁴0-⁴². The elevated Ni content and δ^{34} S values observed in some breccia-hosted troilites could reflect meteorite-induced modification on the CE-6 landing site, though the magnitude of fractionation indicates only limited impact-related alteration on CE-6 breccia.

Early magmatic sulfide segregation could enrich remaining melts in light sulfur isotopes⁴³; hence, determining whether CE-6 basaltic magmas reached early sulfide saturation is essential. Most troilites in

CE-6 basalts are associated with late-stage minerals and low-Mg clinopyroxenes (Figs. 2b and S2b), suggesting late-stage crystallization. Moreover, a troilite grain coexisting with a primitive clinopyroxene (Mg# 43.9) exhibits δ^{34} S of 0.77 ± 0.32‰, consistent with late stage-forming troilites (Fig. 2b), indicating limited sulfur isotope variation during magmatic differentiation.

Sulfide saturation is typically evaluated using the SCSS, which is sensitive to temperature, pressure, and melt composition. Experimental studies conducted under lunar conditions suggest that most basaltic magmas have SCSS values (>2600 μ g/g)^{25,27} that exceed the sulfur contents of mare basalt, implying sulfur-undersaturated evolution. While SCSS can be significantly lowered under highly reduced conditions due to the presence of Fe-rich, S-poor, or Ni-rich sulfides^{27,44}, troilite in CE-6 basalt clasts exhibits a stoichiometric FeS composition (metal/sulfur = 1.0), and the bulk Ni concentration of CE-6 basalt is low (-16.2 μ g/g, Fig. S6), rendering these scenarios unlikely. Using the sulfur solubility model of Ji and Dasgupta²⁷, and applying pressure-temperature conditions analogous to those of CE-5 basalts (1250–1550 °C, 1–1.5 GPa) due to compositional and mineralogical

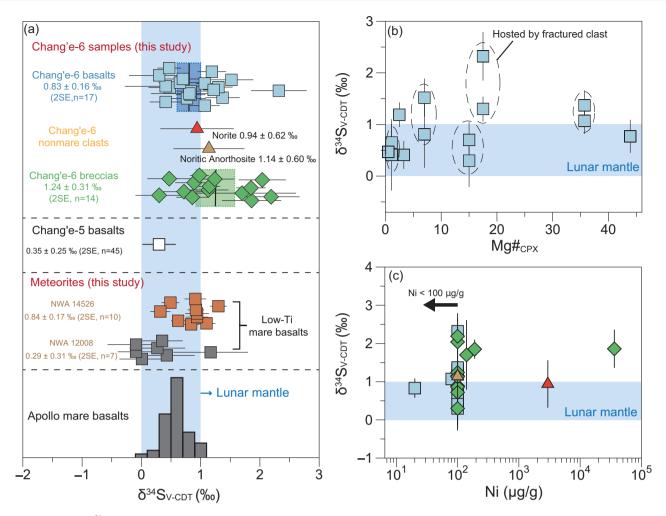


Fig. 2 | Sulfur isotopes (δ^{34} S) of troilites in the basalt clasts, breccias and nonmare clasts in CE-6 soil and lunar meteorites analyzed in this study. Bulk rock δ^{34} S values of Apollo basalts^{8,28,29} and CE-5 basalts^{28,39} are shown for comparison (**a**). δ^{34} S values and Mg# of associated clinopyroxene are plotted in (**b**). Data

enclosed by the dashed black circle in (**b**) represent troilites from the same clast or grain. **c** shows Ni content versus δ^{34} S for the same troilite grain. Error bars represent ± 2 standard errors (2SE) of δ^{34} S measurements.

similarities, we estimate SCSS values for CE-6 magmas in the range of 3420– $5760\,\mu g/g$. This range far exceeds the measured sulfur content $(1800\pm400\,\mu g/g)$, suggesting a sulfur-undersaturated evolution at the early stage of magmatism (Fig. 3a). Furthermore, copper, being both incompatible and chalcophile, is sensitive to sulfide segregation. Early sulfide saturation would markedly lower Cu concentrations in the residual melt⁴⁵. The relatively high Cu contents observed in CE-6 and CE-5 basalts (Fig. 3b) further support the absence of early sulfide segregation during magmatic evolution, as well shown in most mare basalts⁴⁵. In fact, the incompatible behavior of S and other chalcophile elements supports the sulfur-undersaturated evolution for the lunar mare basalts⁴⁵. Such a conclusion is also supported by the petrological evidence that most troilites formed at late stages of magmatic evolution (Figs. 1a, b and S5).

The influence of degassing on sulfur isotopes during lunar magmatism is largely governed by the eruption style (fire-fountain eruptions or effusive eruption)^{29,46}. According to the model by Saal and Hauri⁴⁶, fire-fountain eruptions, such as those that produced volcanic glasses, occur under conditions where the ratio of the effective vapor pressure of sulfur to its saturation pressure (P/P_{Sat}) approaches 1. Degassing under such near-saturation conditions (with a fractionation factor $\alpha_{gas-melt}$ = 1.003) could progressively enriched the residual melt in lighter sulfur isotopes (δ^{34} S value ranging from +1.3% to –14.0%)⁴⁶ (Fig. 4a). In contrast, CE-6 basalts are interpreted

as typical mare basalts formed by effusive eruptions. Any degassing in these lavas would have occurred near the lunar surface under low $P/P_{\rm Sat}$ conditions (e.g., <0.86)^{8,29}, where kinetic isotope fractionation dominates-similar to that observed during impact-related sulfur volatilization (Fig. 4a). Notably, even modest sulfur loss (~20%) under vacuum conditions can induce substantial isotopic enrichment in the residual melt (e.g., a + 7% δ^{34} S shift)^{8,29} (Fig. 4b). In-situ δ^{34} S values of troilite in most CE-6 basalt clasts fall within a relatively narrow range (~1.2%, noting the external uncertainty of ± 0.50 %), suggesting that the sulfur loss during magma emplacement was minimal, likely less than 10%8. Moreover, olivine-hosted melt inclusions of Apollo mare basalts-typically considered to be unaffected by degassing-exhibit δ^{34} S values ranging from $-0.30 \pm 0.91\%$ to $1.6 \pm 1.81\%^{46}$. These values closely match those of troilite in our study, further supporting the interpretation that the CE-6 basalts experienced limited sulfur degassing. This observation aligns with findings from Apollo mare basalts, whose Zn isotope systematics also indicate limited degassing²⁹. One plausible explanation is that thicker mare lava flows developed quenched surface crusts that suppressed volatile escape, thereby minimizing isotopic fractionation despite the Moon's low ambient pressure²⁹.

One troilite grain records a higher δ^{34} S value of $2.32 \pm 0.47\%$, which may reflect intragrain heterogeneity as CE-5 basalt and some lunar meteorites²⁸. This could result from localized degassing through

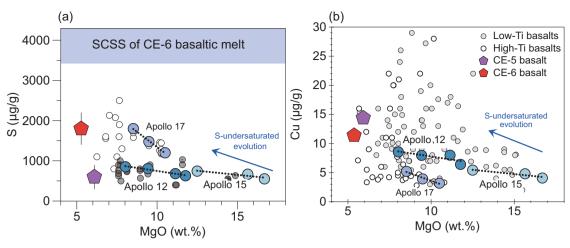
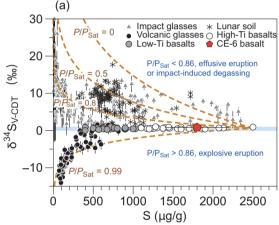
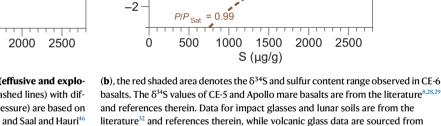


Fig. 3 | Sulfide-undersaturated evolution of lunar mare basalts. S (a) and Cu (b) contents in CE-6, CE-5, and Apollo mare basalts plotted against MgO. The sulfur contents at sulfide saturation (SCSS) of the CE-6 basaltic melt (a) was calculated using the silicate sulfur solubility model proposed by Ji and Dasgupta²⁷. S and Cu contents in Apollo 12, 15, and 17 mare basalts⁴⁵ are highlighted, and the

incompatible behavior of S (a) and Cu (b) in these magmas suggests sulfideundersaturated evolution of most lunar mare basalts. Data source: The S content of CE-5 and Apollo samples are from the refs. 29,39. The Cu content of CE-6, CE-5 are from the literature 36,45,72 and Lunar Sample Compendium (https://www.curator.jsc. nasa.gov/lunar/lsc/index.cfm).

Limited degassing of mare basalts





the ref. 46.

 δ^{34} SV-CDT

(b)

NWA 14526

2

0

Fig. 4 | Sulfur isotope degassing under different eruption (effusive and explosive) and impact conditions. The modeled curves (brown dashed lines) with different P/P_{Sat} (effective vapor pressure relative to saturation pressure) are based on the lunar sulfur isotope degassing model from Gargano et al.29 and Saal and Hauri46 using an initial δ^{34} S value of 0.6% and an initial sulfur content of 2500 $\mu g/g$ (a). In

open fractures or post-crystallization impact processing, as evidenced by the heavily fractured texture of the host clast (Fig. S5c). Such localized processes can generate isolated δ³⁴S enrichments without affecting the broader conclusion that CE-6 basalts experienced overall limited sulfur degassing.

Sulfur isotopic composition of the farside mantle and crust

Lunar basalts offer crucial insights into the Moon's mantle composition. The CE-6 low-Ti basalts, which erupted at ~2.8 Ga, originated from a highly depleted mantle region^{32,33}. As there is no evidence for early sulfide saturation or significant magmatic degassing, their sulfur content (1800 \pm 400 μ g/g) and δ^{34} S value (0.83 \pm 0.16%) likely reflect the composition of the parental melt. Given the high SCSS (>2600 µg/g) of magma under lunar mantle conditions, the mantle source of CE-6 basalts-like other lunar mare basalts^{25,27}-was likely sulfurundersaturated during partial melting. Additionally, sulfur isotope fractionation between sulfide and silicate melt is minimal at high temperatures (-1300 °C; $\alpha_{\text{melt-sulfide}} \approx 0.999$)⁴³, suggesting that sulfur isotope fractionation during partial melting was negligible. Therefore,

the δ^{34} S of the melt is considered a reliable proxy for the mantle source composition.

1500

 $S(\mu g/g)$

2000

2500

In addition to basalt clasts, the CE-6 landing site contains significant nonmare materials, primarily derived from the Chaffee S and other craters within the Apollo basin^{47,48}. Our analyses revealed δ^{34} S values of $1.14 \pm 0.60\%$ and $0.94 \pm 0.62\%$ in troilite from noritic anorthosite and norite clasts, respectively. Petrographic observations suggest minimal alteration driven by impacts (Fig. 1b, c). Some sulfides exhibit elevated Ni contents, possibly attributable to the lowertemperature exsolution of Fe-Ni-Co sulfides and Fe-Ni-Cu sulfides^{49,50} (Figs. S3c, S4). Consequently, these clasts likely preserve the sulfur isotopic composition of the farside crust of variable depth.

Mg-suite rocks are thought to form through partial melting of early magma-ocean cumulates at the base of the crust⁵¹ during the Moon's early evolution⁵². Recent studies suggest that some Mg-suitelike (norite) clasts in the CE-6 soil crystallized at 4.25 Ga ago and likely represents a product of the South Pole-Aitken (SPA) impact melt sheet³⁵. Notably, clasts with similar lithologic and geochemical characteristics have also been identified in Luna 20 samples from the

Crisium Basin, where they have been interpreted as Mg-suite rocks formed in the lower lunar crust⁵³. Although the origin of the Mg-suite (or nortic) clasts in the CE-6 samples is beyond the scope of this study, the high Mg# (-77; Fig. S1) in pyroxene strongly suggests that they are generated from a deep, secondary crust. Taken together, basalt and Mg-suite clasts from the CE-6 landing site span a range of depths, source regions, and ages in the lunar farside mantle and crust. Despite these differences, their sulfur isotopic composition overlaps with each other (Fig. 2a), reinforcing the conclusion that the Moon's interior from the farside overall exhibits sulfur-isotope homogeneity, although they underwent diverse magmatic processes.

Large impact events on the Moon, such as those that formed the SPA and Apollo basins, are believed to have penetrated the lunar crust and excavated mantle materials⁵⁴⁻⁵⁷. Whether these impacts significantly modified the mantle's chemical composition remains uncertain. Since the CE-6 basalt erupted within the region of Apollo and SPA basins at ~2.8 Ga-later than both the SPA (~4.25-4.33 Ga)35,58 and Apollo (~3.9-4.2 Ga) basins-it might have originated from a mantle region potentially influenced by these impacts. The nonmare clast either directly represents rocks crystallized from impact melt produced during the SPA-forming event35, or may result from magmatic activity originating from the deep mantle⁵³, which may have escaped significant modification by the massive impact. The consistent δ³⁴S of the basalt, noritic anorthosite and norite clasts in the CE-6 samples implies that any impact-induced fractionation of sulfur isotopes during the SPA and Apollo impact events was minimal on the studied clasts in the present work. Nevertheless, only a limited number of crustal samples have been analyzed, and further investigations are required to fully assess the extent to which large impacts modified the Moon's farside mantle and crust compositions.

A global evaporation signature inherited from the giant impact

Existing lunar samples exhibit considerable variation in sulfur contents, with high-Ti basalts typically hosting higher sulfur levels than low-Ti basalts (Fig. 5a-c). The S/Dv ratios of mare basalts further demonstrate significant heterogeneity in the mantle sulfur budget (Fig. 5d), consistent with data from olivine-hosted melt inclusions (S/ Dy: 50-200)¹⁶⁻¹⁸. Moreover, the lower S/Dy ratio compared with the MORB value (~212)⁵⁹ indicated that most Apollo basalts stem from a volatile-depleted lunar mantle source. Numerous studies have linked differences in radiogenic isotopes (e.g., variable Sr-Nd isotopes and μ values) among mare basalts and KREEP basalts to the incorporation of varying proportions of KREEP (or KREEP-like) components during mantle overturn²⁴. The fact that mare basalts with distinct μ values and S/Dy ratio (Fig. 5d) suggest that sulfides were incorporated to different extents along with KREEP components, producing an uneven distribution of sulfur in the lunar mantle. This process likely happened during mantle overturn in the early stage of lunar evolution^{19,20}. Nevertheless, given the relatively low sulfur content and high sulfide solubility for lunar magmas, the sulfides tend to have not crystallized until the very late stages of the LMO²⁶ and mare magmatism²⁴. Thus, these mantle reservoirs maintain consistent sulfur isotopic compositions (Fig. 5e, f). KREEP-bearing cumulates (e.g., NWA 6950) also exhibit δ^{34} S values similar to those of other basalts, supporting the homogeneous δ³⁴S values among diverse basalt types and heterogenous lunar mantle.

Prior to the CE-6 mission, all sample-return landings were conducted in the nearside PKT, raising concerns that the observed isotopic signatures and volatile depletions may reflect local anomalies²³. Uneven accretion during the giant impact¹ or large-scale impacts triggering urKREEP degassing^{21,23} could produce such signatures. In this study, the CE-6 basalts returned from lunar farside display an elevated S/Dy ratio (184 ± 42) compared to many Apollo basalts (Fig. 5d), a result that may reflect either relative enrichment of S in the CE-6 mantle source or depletion in rare-earth elements^{32,33}. Assuming a

bulk silicate Earth (BSE) of Dy content $(0.67\,\mu g/g)^{60}$ in the CE-6 mantle source, we estimate a sulfur concentration of $120\pm30\,\mu g/g$, which is slightly higher but still within the upper range proposed for low-Ti basalt sources $(79-120\,\mu g/g)^{17}$. It further supports the idea of sulfur heterogeneity within the lunar mantle. Nevertheless, the $\delta^{34}S$ values of CE-6 basalts and nonmare clasts remain similar to those from nearside Apollo samples and meteorites (Figs. 2a and 6a), excluding a major contribution of localized phenomena or near- and farside asymmetry. It suggests that the lunar interior acquired a sulfur isotopic composition heavier than that of the BSE early in its history and has remained isotopically homogeneous through subsequent processes such as magmatic differentiation and eruption of mare basalts (Fig. 6b).

Because sulfur is both volatile and siderophile, its isotopic composition in the bulk silicate Moon (BSM) could be influenced by coremantle differentiation, late accretion, and volatile evaporation. Experimental studies suggested that metal–silicate fractionation may favor lighter sulfur isotopes in the silicate phase 46,61 . This would result the LMO enriched in light sulfur isotopes, which is inconsistent with our result (Fig. S7a). In contrast, first-principles calculations by Wang et al. 62 proposed that such fractionation under lunar conditions would be minimal (<0.05‰) (Fig. S7b). Thus, the core formation hardly accounts for the 22 % heavier 834 S observed on the BSM relative to the BSE.

Late accretion could also be a contributing factor. The low contents of highly siderophile elements in mare basalts suggest that late-accreted material added only -0.02 wt.% of the Moon's mass into its mantle after core formation⁶³, with sulfur contributions estimated at <10%⁴⁵. Among potential sources, only CM and CO chondrites show $\delta^{34}S$ values marginally overlapping with BSM⁶⁴ (Fig. S8). However, assuming a BSE-like initial $\delta^{34}S$ and a 10% chondritic input still fails to reproduce the observed Earth–Moon offset. More importantly, late accretion cannot explain the $\delta^{34}S$ of the Earth, which experienced a larger fraction of materials from the late accretion⁶².

Therefore, the depletion of sulfur content and enrichment in heavy sulfur isotopes in the lunar interior relative to the BSE are best explained by the volatile evaporation process, consistent with patterns observed for other moderately volatile elements such as K and Zn^{7,9}. Additionally, based on the sulfur isotopic composition of lunar impact glasses³², we estimate isotopic fractionation factors ($\alpha_{gas-melt}$) in the range of ~0.995–0.998 for δ^{34} S during impact-induced events (Fig. S9a). Applying these fractionation factors, we estimate that 20-75% of lunar sulfur would have been lost to produce the observed δ³⁴S value of the BSM (Fig. S9b). Importantly, the globally homogeneous sulfur isotopic composition of the BSM rules out the possibility that the observed $\delta^{34}S$ signature reflects localized impact events²³. Instead, it supports a global evaporation imprint inherited from the giant impact, either in the protolunar disk^{7,9} or during tidally assisted magma ocean degassing^{65,66}. This main conclusion is broadly consistent with a recent modeling work⁶⁷, which also highlights the role of the giant impact in driving volatile loss and sulfur isotope fractionation.

Despite the homogeneity in sulfur isotopic composition across the lunar interior, the variation of sulfur contents and Dy/S ratios of different mantle domains^{16,17} suggests that complex post-formation processes, such as mantle overturn, have significantly modified the distribution of sulfur, Nd–Pb isotopic signatures, and possibly other volatile elements within the lunar mantle. Future analyses of the isotopic composition of other volatile elements (e.g., Cl, K, Rb, Cu and Zn) in lunar farside samples will be essential for further testing this model.

Methods

CE-6 samples and petrological analyses

The scooped CE-6 soil (CE6CO200YJFM003, 500 mg) was obtained from the China National Space Administration Agency. Hundreds of lithic fragments with the size exceeding 50 µm were manually picked out from the CE-6 soil. Then, they were subsequently mounted in resin

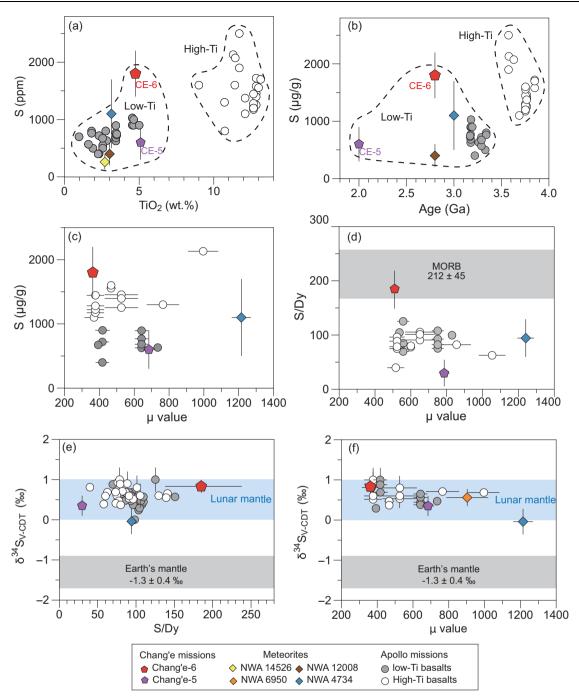


Fig. 5 | Comparison of lunar farside and nearside basalts and other basaltic meteorites. Mare basalts show variable S content with high-Ti basalt higher than low-Ti basalts (\mathbf{a} - \mathbf{c}). The S/Dy of both low-Ti and high-Ti basalts are variable and lower than that of mid-ocean ridge basalts (212 ± 45)⁵⁹ (\mathbf{d}). The δ^{34} S values further show a global homogeneity (0–1‰), although mantle domains are very

heterogenous (**e**, **f**). Data source: The δ^{34} S values of meteorites (NWA 4734 and NWA 6950) are from the ref. 28. Ages of CE-6, CE-5, meteorites and Apollo basalts are from the refs. 24,32,33,73. The μ values (238 U/ 204 Pb ratio) of CE-6, CE-5, meteorites and Apollo basalts are from the literature 33 and references therein.

sections for detailed petrological and geochemical analyses (Fig. 1). These clasts include basalt clasts, breccias and nonmare clasts (e.g., Mg-suite clasts). Two lunar low-Ti basaltic meteorites (NWA 14526 and NWA 12008) are also analyzed for comparison.

The lithic fragments were analyzed by TESCAN Integrated Mineral Analyzer system (TIMA3 X GHM) at China University of Geosciences, Wuhan. Detailed methods for TIMA analyses can be found in Wang et al.²⁸. The combination of TIMA measurements with BSE images and X-ray spectra can identify individual grains and locate grain boundaries. The number of pixels of each grain phase was then converted

into the relative surface area as a modal percentage. Based on this, the system automatically compared the measured BSE and EDS data of each phase with a built-in classification scheme, distinguished mineral phases of basaltic clasts, and then computed mineral volumetric fractions (Fig. S2).

Major elements of minerals

Major elemental compositions of different minerals were determined using a JEOL JXA-8230 electron microprobe analyzer at the State Key Laboratory of Geological Processes and Mineral Resources, China

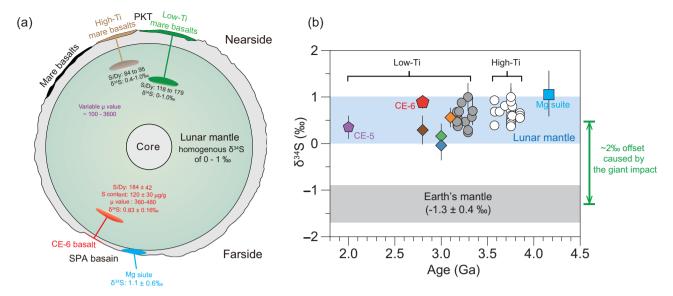


Fig. 6 | Distribution of S/Dy, μ value and δ^{34} S in the lunar mantle of different regions. The lunar mantle demonstrates significant variation on S contents and μ values (a), but their sulfur isotopes remain consistent after volatile loss induced

by the giant impact, regardless of ages of basalts and Mg-suite rocks (**b**). Symbols follow the same legend as in Fig. 5; the green diamond represents NWA 10597.

University of Geosciences, Wuhan (GPMR-CUG). The focused beam was typically at 3 µm diameter for silicate and 1 µm (spot analysis mode) for sulfides. For silicate phases, the standards used for ZAF matrix correction were jadeite (Na), olivine (Si), diopside (Ca, Mg), almandine (Fe, Al), sanidine (K), rutile (Ti), rhodonite (Mn), and chromium oxide (Cr). For sulfides, the standards were hematite (Fe), pyrite (S), pentlandite (Ni), cobalt (Co), chalcopyrite (Cu) and chromite (Cr), with an alloy standard for Fe–Ni metals. Some grains were mapped for expected major and minor elements (e.g., S, Fe, Ni, Co, and Cu) (Fig. S3).

Laser ablation MC-ICP-MS for sulfur isotopes

In this study, we performed in-situ sulfur isotopes analysis in troilites with an ESI NWR FemtoUC femtosecond laser ablation system (New Wave Research, Fremont, CA, USA) coupled to a Neptune Plus MC-ICP-MS instrument (Thermo Fisher Scientific, Germany) at the GPMR. Given ultrafast energy deposition, the femtosecond laser ablation system can significantly reduce the matrix effect and is a convenient and robust approach for in-situ sulfur isotopic analyses of extraterrestrial samples which have achieved similar analytical uncertainty with Nano SIMS²⁸. A frequency of 10 Hz with 5 s⁻¹ ablation for a total of 80 pulses was used, allowing high spatial-resolution analyses of 8-µm spot sizes in this study^{28,68}. Detailed analytical strategies and data quality of this method has also been discussed in Wang et al.²⁸.

The pyrite reference PPP-1⁶⁹ with $\delta^{34}S_{V\text{-}CDT} = 5.3 \pm 0.2\%$ was used as an external standard. The pyrrhotite reference YP136 and troilites from two iron meteorites (Muonionalusta and NWA 859) were analyzed as unknown samples to monitor data quality. The YP136 show $\delta^{34}S_{V\text{-}CDT} = 1.46 \pm 0.48\%$ (n = 10, 2 SD) within the range of reference values $(1.5 \pm 0.3\%)^{70}$ (Table S5 and Fig. S10). The $\delta^{34}S_{V\text{-}CDT}$ of troilites in Muonionalusta and NWA 859 are $0.12 \pm 0.48\%$ and $0.45 \pm 0.50\%$, respectively, overlapping with the values reported before²⁸. The results are also consistent with the $\delta^{34}S_{V\text{-}CDT}$ of iron meteorites measured using a ThermoFinnigan MAT 253 mass spectrometer⁷¹. The reproducibility of the reference materials suggests that the external uncertainty in this study is 0.50% (2sd) for $\delta^{34}S_{V\text{-}CDT}$, which is comparable to sulfur isotopes obtained by nano-SIMS³⁹.

Data availability

All data generated and analyzed during this study are included in the Supplementary Information files. The sulfur isotope and mineralogical

data generated in this study have been deposited in the Figshare database under accession code https://doi.org/10.6084/m9.figshare. 29136590.

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Author contributions

Z.C.W. and Y.H.L. designed the research and applied for the CE-6 samples. F.M. and J.S. helped in refining the ideas. Y.H.L., Z.C.W., W.Z., Z.C.H., J.Q.Z., and X.C. measured the sulfur isotopes of troilites, Y.H.L., Z.C.W., K.Q.Z., Z.B.S., Q.H., J.Q.Z., T.Y.L., F.B.P., X.C., L.X., X.W., and K.C. prepared samples and obtained EMPA and TIMA data. Y.S.L. and Z.C.H. provided access to the experimental platforms. Y.H.L. and Z.C.W. wrote the manuscript draft, and all other authors participated in the data interpretation and editing of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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