

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

Corresponding Author: Professor Zaicong Wang

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This manuscript reports new sulfur isotope data from the Chang'e-6 mission. The authors integrate their results with previously reported isotope data from nearside lunar samples, proposing that the lunar mantle has a homogeneous sulfur isotope composition inherited from the Moon-forming impact. Although the data are novel and valuable to the field, I cannot recommend the manuscript for publication without major revisions to improve clarity and strengthen the arguments.

My primary concern with the manuscript is that it assumes the measured sulfur isotope compositions directly represent the depletion inherited from the Moon-forming impact, without adequately testing for or addressing potential secondary processes. Below, I outline specific issues that should be addressed.

Line 31: The sentence presents a strict dichotomy (Moon-forming impact vs. regional degassing). Other processes such as fractional crystallization, partial melting, and small-scale overturn could also affect sulfur isotope signatures and should be considered.

Line 39: This statement should be more nuanced, given the range of $\delta^{34}\text{S}$ values reported later in the manuscript.

Line 133: Ni content is an important tracer for melt petrogenesis and could help evaluate whether sulfur isotopes were altered by impact or partial melting. This discussion is missing and should be included.

Line 158: The observed intra-grain $\delta^{34}\text{S}$ variation is significant enough to warrant a more detailed petrographic description in the results section.

Line 168: Since the conclusion of sulfur isotope homogeneity depends on late-stage crystallization not affecting $\delta^{34}\text{S}$, this claim requires stronger justification.

Line 171: Degassing can still occur, and impact melting and degassing are major processes. Are there no signs of shock or melt in the samples?

Lines 171–177: The intra-grain variations suggest isotopic fractionation occurred at some stage, which directly contradicts the paper's conclusion of a homogeneous S. This issue needs to be addressed.

Line 177: Why use the average $\delta^{34}\text{S}$ instead of the value in the most primitive sample or the lightest, if fractionation is clear? If the authors accept localized degassing in troilite but not mantle-wide degassing, they need to explain why one process occurs while the other does not.

Line 179: It is unclear what the authors are referring to in Fig. 2C. Is there petrographic evidence that the primitive pxn isn't an antecryst?

Line 191: What constitutes "substantial" degassing in this scenario? A ~1.2‰ difference between samples suggest some degree of degassing, yet this is dismissed. Additionally, line 186 acknowledges meteorite-induced sulfur modification. How

is this reconciled with the idea that sulfur remained unaffected?

Line 206: I disagree with the statement that Figure 2a shows a uniform sulfur composition. Equilibrium fractionation at lunar temperatures would not account for a 2‰ range in sulfur isotopes. This variation shows that multiple processes have modified the samples.

Lines 213–219: These sentences are unclear. Zhang et al. (2024) showed that the CE-6 samples originated from mantle reservoirs that experienced substantial differentiation and volatile redistribution, possibly due to impacts. How can these processes not affect sulfur isotopes?

Line 237: Figures 4e and 4f do not exist. This needs to be changed.

Line 237: Why would sulfur isotopes remain homogeneous after large-scale mantle processing when Pb heterogeneity in KREEP basalts suggests that these melts underwent fractionation processes?

Lines 251–256: These sentences need clarification. The manuscript should provide a more in-depth explanation of how sulfur content and other volatile element isotopic compositions can vary significantly, yet the lunar interior can remain homogeneous in sulfur isotopic composition.

Figure 1: Move the Mg# values of pyroxenes closer to their labels to improve readability.

Figure 2: Label the blue region. Why aren't all the Chang'e-6 data grouped together at the top? What are the reported $\delta^{34}\text{S}$ values for the Chang'e-6 non-mare basalts?

Figure 4a: The CE-5 label is missing, please correct this.

Reviewer #2

(Remarks to the Author)

Review of the manuscript "Sulfur isotopes from the lunar farside reveal a global giant impact signature" by Li et al.

This manuscript investigates the sulfur content and $\delta^{34}\text{S}$ isotopic composition of the returned farside soil sample collected by the Chang'e-6 mission. Troilite grains in selected basalt clasts, nonmare clasts (Mg suite and FAN) and breccias from the sample were analyzed. The $\delta^{34}\text{S}$ values of these grains are consistent with those observed in CE5 basalts, Apollo high-Ti and low-Ti basalts, and lunar meteorites, indicating a homogeneous $\delta^{34}\text{S}$ composition in the lunar interior, which is approximately 2‰ heavier than Earth's sulfur isotope signature. In contrast, S concentrations in the lunar mantle are heterogeneous as indicated by S/Dy ratio variations. The S concentration in the CE6 mantle source is estimated to be 120 ppm, the highest among the mantle sources of CE5, Apollo high-Ti and low-Ti basalts and lunar meteorites. The authors proposed that the consistently high and homogeneous $\delta^{34}\text{S}$ composition of the Moon suggests that this characteristic has remained stable since the Moon's formation, a signature from the giant impact.

This work presents a comprehensive investigation into the S isotope composition in the CE6 soil sample and the data are of high quality. While this work is of broad interest, I have several major concerns related to data interpretation that should be considered before its publication. Additionally, there are numerous typos throughout the manuscript and many figure citations are out of order.

Recommendation: Major revision.

Major comments:

First, S is not only a highly volatile but also a siderophile (iron-loving) element. During the lunar core formation, S isotope fractionation between the core and BSM is expected. This fractionation will significantly affect the lunar mantle's $\delta^{34}\text{S}$ composition and its homogeneity, but a discussion on this process is critically lacking in this manuscript.

Second, there are many important processes can alter $\delta^{34}\text{S}$ in the CE6 magma which however are excluded without strong justifications. For example, I do not think magma degassing can be simply ruled out just because one troilite data point near primitive cpx shows roughly consistent $\delta^{34}\text{S}$ value with others. A study on troilites from CE5 samples has shown a decreasing trend of $\delta^{34}\text{S}$ in the magma during crystallization and degassing, with a total of 40% original S loss (Liu et al. 2022, JGR Planets). This amount of S degassing is not negligible. Furthermore, the high S/Dy ratio of CE6 and high S abundance in the CE6 mantle source may indicate that the source region for CE6 magma is sulfide saturated, which will lead to a lighter $\delta^{34}\text{S}$ in the produced melts. The lack of such justifications leaves the argument in the manuscript incomplete and unconvincing.

Finally, for a paper published in Nature Communications, I look forward to a broad discussion on the origin of the Moon. For example, it has been proposed that water in the interiors of the Earth and Moon may have been inherited from carbonaceous chondrites due to their indistinguishable hydrogen isotope compositions (Saal et al. 2015 Science). What are the S isotope

compositions of carbonaceous chondrites and what is the consequence of this hypothesis to the S isotopes in the Moon? In addition, late veneer has been proposed to explain the homogeneous distribution of highly siderophile elements with chondritic proportions in the lunar mantle sources of mare basalts. It would be nice to see discussions on these topics and give a broad view on the formation of the Moon from the perspective of $\delta^{34}\text{S}$ in CE6. The current discussion on the signature from the giant impact is a little shallow and ambiguous - do the authors mean the signature from Theia, or the proto-Earth, or the mixture, or the processes during the impact?

Specific comments:

L59-63: There are many new lunar melt inclusion studies on volatile abundances, such as those by Ni et al. (2019, GCA) and Su and Zhang (2024, GCA). The significant heterogeneity of H_2O in lunar melt inclusions, for example, is interpreted to result from differences in cooling rates, with samples that cooled the fastest best preserving the pre-eruptive volatile contents (Su and Zhang, 2024).

L79-80: Can you give numbers to quantify “relatively limited”? In addition, basalts from CE5 are estimated to have lost ~40% of their original S, which is not the case as justified here.

L93: where does the “90%” information come from? Give relevant references.

L110: I do not see any illustration of basalt clasts, mineral fragments, and impact glasses in Figs. 1d and 1e except troilite.

L115: Fig. S2a instead of Fig. S1a?

L146: Fig. 2b instead of Fig. 2c?

L157: Saal and Hauri (2021, Science Advances) report S concentration and $\delta^{34}\text{S}$ of lunar olivine-hosted melt inclusions. It is generally thought that melt inclusions represent pre-eruptive (undegassed) volatile contents. The $\delta^{34}\text{S}$ data from these melt inclusions should be discussed.

L163: I do not understand why Figs. 2c is listed here.

L217-219: I suggest not extrapolating from S to all volatiles and their isotopes. In addition, this work only reports 1 data point of Mg suite clasts from the CE6 soil sample; whether this data point is representative is unknown.

L226: Figs. 3c, d instead of Figs. 3a, b?

L227: Pyroxene-hosted melt inclusions from lunar very-low-Ti basalts have a S/Dy ratio from 91-296 (Su and Zhang 2024 GCA).

L232: Fig.3b instead of Fig.3c?

L491-492: The Mg# of Cpx [and Opx].

L493: Fig.S1 instead of Fig.S3?

Fig.1: Please also add the An# of plag.

Fig.2a: 1) Are the data from CE5 basalts also from this study? Clarify this in the figure caption. 2) Plot Mg suite and Fe separately and add the $\delta^{34}\text{S}$ values to the figure like others for comparison.

Fig.3: Wing and Farquhar (2015) also report S and $\delta^{34}\text{S}$ of a series of lunar high-Ti and low-Ti basalts. These data should be added and discussed.

Version 1:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The authors have satisfactorily addressed most of my previous concerns by adding comprehensive discussion on the effects of core-mantle differentiation, late accretion, volatile degassing, sulfide saturation, etc. Considerable effort has gone into the revision of the manuscript. The revised manuscript is clear and well-written.

I only have one comment for the authors:

Fu and Dottin (2025 LPSC) modeled the coevolution of S abundance and $\delta^{34}\text{S}$ of the BSM in the context of giant-impact models. This work indicates that volatile loss from the giant impact event produces an increase of up to ~1‰ in $\delta^{34}\text{S}$, which is insufficient to explain the 2‰ $\delta^{34}\text{S}$ enrichment of the BSM. The work of Fu and Dottin (2025 LPSC) is highly relevant to this work and should be discussed.

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1 [Replies to comments in blue color](#)

2 RE: NCOMMS-25-14618

3 Sulfur isotopes from the lunar farside reveal a global evaporation signature following
4 the giant impact

5 Nature Communications

7 Reviewer #1 (Remarks to the Author):

9 This manuscript reports new sulfur isotope data from the Chang'e-6 mission. The
10 authors integrate their results with previously reported isotope data from nearside
11 lunar samples, proposing that the lunar mantle has a homogeneous sulfur isotope
12 composition inherited from the Moon-forming impact. Although the data are novel
13 and valuable to the field, I cannot recommend the manuscript for publication without
14 major revisions to improve clarity and strengthen the arguments.

16 My primary concern with the manuscript is that it assumes the measured sulfur
17 isotope compositions directly represent the depletion inherited from the Moon-
18 forming impact, without adequately testing for or addressing potential secondary
19 processes. Below, I outline specific issues that should be addressed.

20 [Response:](#)

21 [Thanks for your constructive feedback. Below, according to your comments, we](#)
22 [outline the major revisions we have made in our manuscript.](#)

23 [1. Based on the low Ni concentrations and petrographic features of CE-6 basalts and](#)
24 [associated troilites, we infer minimal meteoritic modification of the basalt clasts. In](#)
25 [contrast, the breccias appear to have experienced minor meteoritic alteration.](#)

26 [2. We evaluated the sulfide saturation state of CE-6 magma by comparing sulfur, Cu,](#)
27 [and Ni contents with those of CE-5 and Apollo mare basalts, and by applying sulfur](#)
28 [solubility models \(SCSS\). The results indicate that CE-6 basalts, like most mare](#)
29 [basalts, evolved under sulfide-undersaturated conditions, both during mantle partial](#)
30 [melting and subsequent magmatic evolution.](#)

31 [3. We modeled sulfur isotope fractionation under different lunar eruptive conditions](#)
32 [and found that the observed \$\delta^{34}\text{S}\$ variation in CE-6 basalts \(\$\sim 1.2\%\$ \) corresponds to](#)
33 [<10% degassing. Notably, a clast containing the highest \$\delta^{34}\text{S}\$ value also exhibits](#)
34 [significant fracturing, suggesting localized degassing or post-crystallization impact](#)
35 [modification. However, this does not alter our overall conclusion that the CE-6](#)
36 [basalts experienced minimal degassing.](#)

37 [Taken together, we conclude that the \$\delta^{34}\text{S}\$ values in CE-6 basalts reliably reflect their](#)
38 [mantle source signatures. A more detailed explanation and discussion of these](#)

possible processes have been added in the revised manuscript and are further addressed in our responses below.

Line 31: The sentence presents a strict dichotomy (Moon-forming impact vs. regional degassing). Other processes such as fractional crystallization, partial melting, and small-scale overturn could also affect sulfur isotope signatures and should be considered.

Response: Thank you for this constructive comment. We agree that the original sentence presented an overly simplified dichotomy. In response, we have revised the sentence on Line 31 to:

“The pronounced nearside–farside dichotomy and uneven distribution of volatiles across the lunar mantle raise the question of whether heavier isotopic signatures result from a global giant impact or local magmatic processes.”

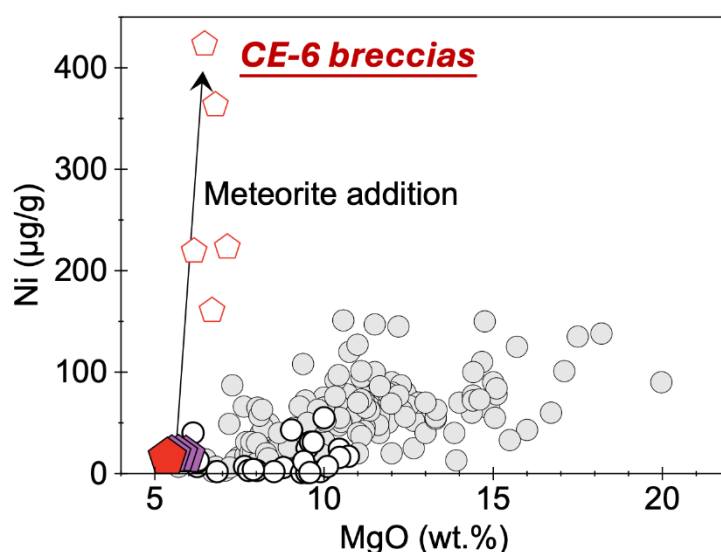
Line 39: This statement should be more nuanced, given the range of $\delta^{34}\text{S}$ values reported later in the manuscript.

Response: Thank you for this helpful suggestion. We have revised the statement.

Line 133: Ni content is an important tracer for melt petrogenesis and could help evaluate whether sulfur isotopes were altered by impact or partial melting. This discussion is missing and should be included.

Response: We appreciate the suggestion and will include this in our discussion. Meteorites typically contain elevated Ni levels; thus, an increase in Ni content within a sample may indicate meteoritic contamination, which can lead to partial melting and accompanying isotopic fractionation through volatile loss. In this study, however, we propose that basalt clasts were minimally affected by meteoritic influence, preserving their primary magmatic signatures, whereas certain breccias may have experienced more substantial meteoritic alteration. The key observations are as follows:

1) Basaltic Clasts. The Ni content in the CE-6 basaltic clasts ($\sim 16.2 \mu\text{g/g}$, Wang et al., 2025, in press) aligns well with that of pristine Apollo basalts and Chang’e-5 basaltic samples (Reply Fig. 1). In addition, the Ni concentration in CE-6 troilite similarly falls within the range reported for Apollo and CE-5 troilite (Fig. S4a). Furthermore, the basaltic clasts analyzed exhibit no clear evidence of meteoritic modification—such as impact melts or shock veins—indicating minimal contamination (Figs. 1a,b and S5).



Reply Figure 1. Ni and MgO content of CE-6 basalt, breccias and Apollo mare basalts. The data of CE-6 basalt and breccia are from Wang et al., (2025). The data of Apollo mare basalts are from Lunar Sample Compendium (<https://wwwcurator.jsc.nasa.gov/lunar/lsc/index.cfm>). CE-5 basalt is from (Z. Wang et al., 2024b)

2) Non-Mare Clasts. Compared with the CE-6 basalts, troilite grains in Mg-suite and FAN samples are generally smaller ($< 10 \mu\text{m}$) and some of them display higher Ni contents (Fig. S4a). Elemental mapping reveals occasionally isolated hotspots of Co, Ni and Cu (Fig. S3). Moreover, Co and Ni exhibit correlated distributions, whereas Cu shows little correlation (Fig. S3), consistent with EPMA measurements (Fig. S4a and b). These anomalous spots likely linked to the lower-temperature exsolution of Fe–Ni–Co sulfides and Fe–Ni–Cu sulfides (Etschmann et al., 2004; Vaughan, 2006). As a result, the elevated Ni content in troilite from the Mg-suite and FAN may arise from signal potential contamination during analysis, due to the large analytical spot size ($1\mu\text{m}$) and the small grain size of the troilite.

3) Breccias. Breccias are common clasts in lunar soil that experienced significant impact-induced modifications (Lucey, 2006). Some breccias exhibit very high Ni concentrations ($>150 \text{ ppm}$, Reply Fig. 1), with troilite Ni levels similar to those previously reported in impact-related troilites ($\sim 1000 \mu\text{g/g}$) (Z. Wang et al., 2024a). In addition, the $\delta^{34}\text{S}$ value of troilites in breccias are overall heavier than that in basalt clasts, potentially reflecting the sulfur volatilization during impact. This observation is consistent with the heavy sulfur signatures documented in lunar soil and impact glasses (B.-W. Wang et al., 2024).

Line 158: The observed intra-grain $\delta^{34}\text{S}$ variation is significant enough to warrant a more detailed petrographic description in the results section.

Response: Thanks for your suggestion. we have expanded our petrographic descriptions in the Results section to provide a more detailed context for these isotopic variations.

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 ($1.30 \pm 0.24\text{‰}$ and $2.32 \pm 0.47\text{‰}$; 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. Consequently, it is possible that this troilite's sulfur isotopic variation was influenced by impact processes.

Line 168: Since the conclusion of sulfur isotope homogeneity depends on late-stage crystallization not affecting $\delta^{34}\text{S}$, this claim requires stronger justification.

Response: In the revised manuscript, we have expanded our discussion to provide more detailed support for this claim, drawing on two lines of evidence:

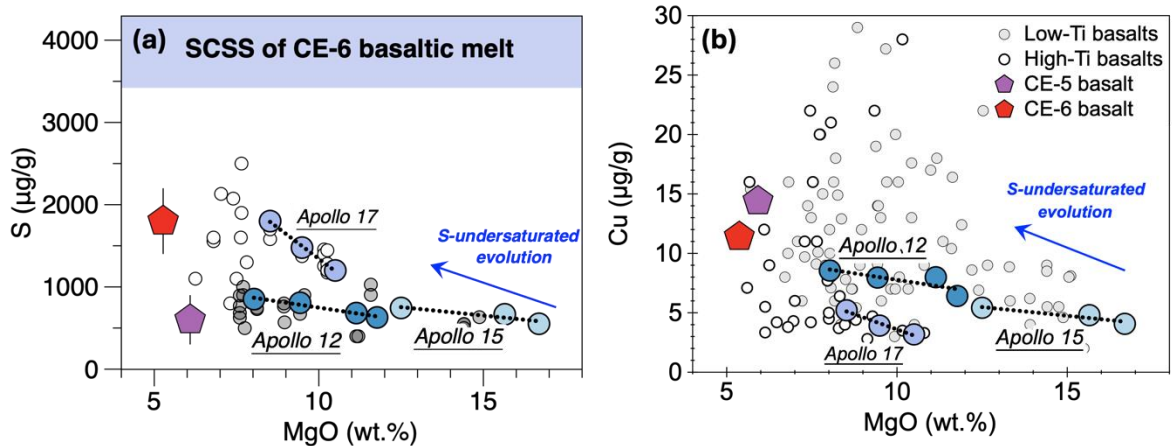
1. Petrographic Observations. As indicated in the main text, the majority of troilites in our samples are intergranular and are primarily associated with late-stage minerals (e.g., ilmenite, plagioclase, fayalite, apatite, and low-Mg# clinopyroxene). This textural context suggests that these troilites crystallized during the final stages of magmatic evolution, yet still preserve uniform sulfur isotope compositions.

2. Evidence for Sulfur-Undersaturated Evolution in CE-6 Basalts.

The sulfur content at sulfide saturation (SCSS) is commonly used to evaluate whether a melt may attain sulfide saturation. This parameter depends on temperature, pressure, and melt composition. Experimental studies have shown that, under lunar conditions, most basaltic magmas have SCSS values exceeding their actual sulfur concentrations, implying a sulfur-undersaturated evolution (e.g., Ding et al., 2018; Ji and Dasgupta, 2025). However, Brenan et al. (2019) proposed that in highly reduced conditions, the presence of Fe-rich and S-poor sulfides could significantly lower the SCSS, potentially leading to early sulfide saturation. Additionally, elevated Ni content in sulfides can also reduce SCSS (Ji and Dasgupta, 2025).

In this study, troilite in CE-6 basalts is FeS ($M/S = 1.0$) and the CE-6 basalt has low Ni (~ 16.2 ppm), making Fe-rich or Ni-rich sulfides unlikely to form. Using the recent sulfur solubility model for lunar silicate melts by Ji and Dasgupta, (2025), we estimate SCSS values of $\sim 3420\text{--}5760$ ppm for CE-6 basalts over relevant temperature ($1250\text{--}1550$ °C) and pressure ($1\text{--}1.5$ GPa) ranges (we used the two potential conditions of CE-5 lunar mantle due to their compositional and mineralogical similarities). This concentration is significantly higher than the observed sulfur content of CE-6 basalts (1800 ± 300 $\mu\text{g/g}$, Reply Fig. 2a), indicating that the CE-6 basalt magmas remained undersaturated in sulfide in the early stage. In addition, the

elevated Cu content in CE-6 basalts also argues against a sulfide-saturated magmatic evolution. Because Cu is both incompatible and chalcophile, the crystallization of sulfides at saturation would markedly reduce the Cu content in the residual melt. The close similarity between the Cu contents of CE-6 and CE-5 basalts (Reply Fig. 2)—where experimental studies have already established a sulfide-undersaturated evolution—further underscores that CE-6 basalts similarly evolved without reaching sulfide saturation. In fact, the incompatible behavior of S and other chalcophile elements supports the sulfur-undersaturated evolution for lunar mare basalts (Gleißner et al., 2022).



Reply Figure 2. S (a) and Cu (b) contents in CE-6, CE-5, and Apollo mare basalts plotted against MgO. The SCSS of the CE-6 basaltic melt (a) was calculated using the silicate sulfur solubility model proposed by Ji and Dasgupta, (2025). S and Cu contents of Apollo 12, 15, and 17 mare basalts (Gleißner et al., 2022) are highlighted, the incompatible behavior of S and Cu in these magmas suggests sulfide-undersaturated evolution of mare basalts. Data source: The S content of CE-5 and Apollo samples are from the reference (Liu et al., 2022; Gargano et al., 2022). The Cu content of CE-6, CE-5 are from the literature (Gleißner et al., 2022; Z. Wang et al., 2024b; Wang et al., 2025) and Lunar Sample Compendium (<https://wwwcurator.jsc.nasa.gov/lunar/lsc/index.cfm>).

Line 171: Degassing can still occur, and impact melting and degassing are major processes. Are there no signs of shock or melt in the samples?

Response: Thanks for your suggestion. As noted in our earlier responses, we do observe clear evidence of shock effects in some of the breccias, which exhibit elevated $\delta^{34}\text{S}$ values consistent with impact-related modifications. In contrast, most basalt clasts and non-mare clasts show neither the elevated Ni contents nor the petrographic evidences (e.g., shock veins, melt pockets) that would indicate meteoritic contamination or post-formation modification.

The certain troilite grain exhibit a larger discrepancy ($1.30 \pm 0.24\%$ and $2.32 \pm 0.47\%$; Fig. S5c), is in contact with pyroxene at the clast's edge, while the clast itself is significantly fractured. Consequently, it is possible that this troilite's sulfur isotopic variation was influenced by impact processes. We thus excluded the $\delta^{34}\text{S}$ values of this grain during calculating the average value of CE-6 basalt.

Lines 171–177: The intra-grain variations suggest isotopic fractionation occurred at some stage, which directly contradicts the paper's conclusion of a homogeneous S. This issue needs to be addressed.

Response: Thank you for your feedback. We will carefully expand the disccision on this aspect in the manuscript. Below, we clarify that sulfur isotopes can fractionate differently under various lunar processes, yet still indicate limited sulfur degassing in most CE-6 basalts.

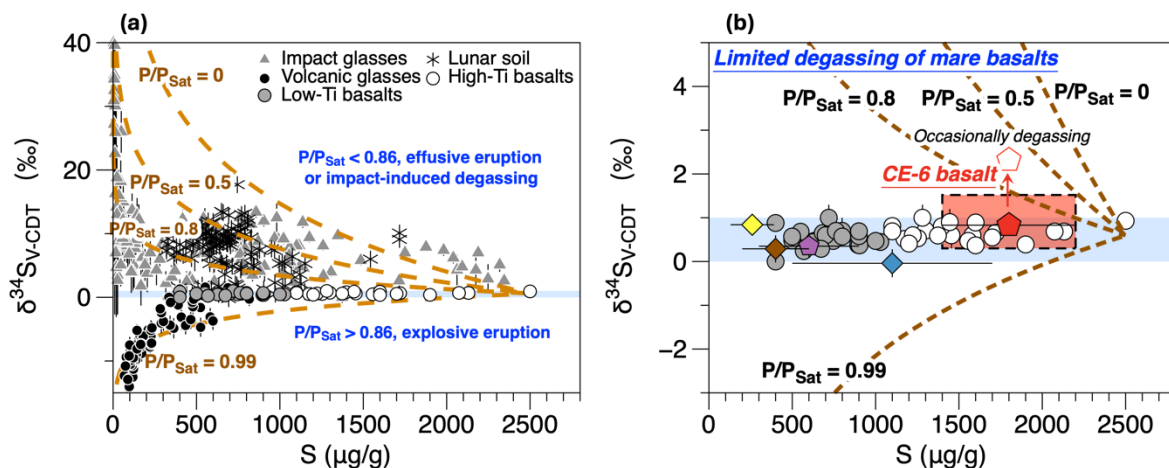
1. Degassing Regimes for Different Eruption Styles

1.1 Explosive Eruptions (e.g., Volcanic Glasses).

Explosive volcanism are characterized by high ascent rate explosive volcanic eruptions via rapid dike propagation driven by CO and H₂, which resulted in widespread pyroclastic deposits such as picritic glasses. Saal and Hauri, (2021) propose a degassing regime with an effective isotope fractionation factor of

$$\alpha'_{kin} = (\alpha_{kin} - 1) \left(1 - \frac{P}{P_{Sat}}\right) + 1$$

where P/P_{Sat} is equal to the effective vapor pressure of a given element relative to the saturation vapor pressure (Saal and Hauri, 2021). Under the condition of explosive eruptions, significant degassing occurs at near-saturation vapor pressures ($P/P_{Sat} \approx 1$), where sulfur-isotope fractionation is dominated by equilibrium processes ($\alpha_{gas-melt} \approx 1.003$), leading to progressively lighter residual melts in terms of $\delta^{34}\text{S}$ (Reply Fig. 3a).



Reply Figure 3. Sulfur isotope degassing under different eruption (effusive and explosive) and impact conditions. The modeled curves (brown dashed lines) with

different P/P_{Sat} are based on the lunar sulfur isotope degassing model from Gargano et al. (2022) and Saal and Hauri (2021) using an initial $\delta^{34}\text{S}$ value of 0.6‰ from Wang et al. (2024) and an initial sulfur content of 2500 $\mu\text{g/g}$, representing the upper limit for lunar basalt. In panel (b), the red shaded area denotes the $\delta^{34}\text{S}$ and sulfur content range observed in CE-6 basalts. Data for impact glasses and lunar soils are from Wang et al. (2024) and references therein, while volcanic glass data are sourced from Saal and Hauri (2021).

1.2 Effusive Eruptions (e.g., Mare Basalts) and Impact-Induced Evaporation (e.g., Impact Glasses and Breccias).

In contrast, CE-6 basalts are interpreted as classic mare basalts emplaced through effusive eruptions. If degassing occurred, it would mainly happen at the lunar surface under low P/P_{Sat} (Wing and Farquhar, 2015; Gargano et al., 2022), where kinetic fractionation prevails—similar to the mechanisms observed in impact-related sulfur volatilization. Even a small amount of sulfur loss (~20%) can markedly increase $\delta^{34}\text{S}$ in the residual melt (e.g., a +7‰ shift under vacuum) (Gargano et al., 2022). Indeed, the pronounced sulfur-isotope fractionation observed in lunar impact glasses and soil highlights the potential role of impact processes in driving volatile loss (Reply Fig. 3a).

2. Evidence for Limited Degassing in CE-6 Basalts

Despite the potential for kinetic fractionation, in situ troilite $\delta^{34}\text{S}$ values in most CE-6 basaltic clasts range from $0.30 \pm 0.51\text{‰}$ to $1.52 \pm 0.37\text{‰}$ (given the external uncertainty in this study is 0.50 ‰ for $\delta^{34}\text{S}$). This relatively narrow range implies minimal overall sulfur loss (<10%, Reply Fig. 3b), consistent with findings from Apollo mare basalts and their S, Cl, and Zn isotope data (Wing and Farquhar, 2015; Gargano et al., 2020), which also point to low degrees of degassing. One likely explanation is that thicker mare lava flows formed quenched crusts, limiting volatile escape and thus reducing isotopic fractionation despite the lunar surface's low ambient pressure.

3. Variations in Individual Grains and Breccias

One troilite grain displays a higher $\delta^{34}\text{S}$ value ($2.32 \pm 0.47\text{‰}$), potentially resulting from localized degassing via open fractures or post-crystallization impacts, as suggested by the clast's highly fractured texture (Fig.S5c). Such localized processes can produce isolated, elevated $\delta^{34}\text{S}$ values without undermining the broader conclusion of low overall degassing. Breccias, with a relatively higher mean $\delta^{34}\text{S}$ ($1.24 \pm 0.31\text{‰}$, 2SD, $n = 14$), likely record degassing caused by meteoritic impacts, though the magnitude of fractionation indicates only limited impact-related alteration. Furthermore, the lowest $\delta^{34}\text{S}$ value in the breccias ($0.30 \pm 0.40\text{‰}$) overlaps with the range observed in CE-6 basaltic troilite, suggesting a consistent initial sulfur-isotope composition across the CE-6 suite.

In summary, while a troilite of a certain basalt clast and several breccias exhibit elevated $\delta^{34}\text{S}$ due to localized degassing or impacts, most CE-6 basaltic troilites

show limited $\delta^{34}\text{S}$ variation, indicating minimal sulfur loss. These observations support our interpretation of a broadly homogeneous sulfur-isotope composition in CE-6 basalts.

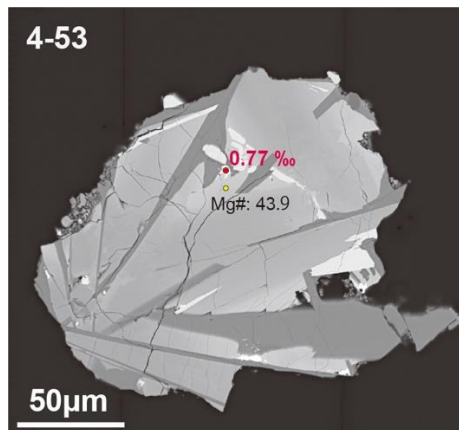
Line 177: Why use the average $\delta^{34}\text{S}$ instead of the value in the most primitive sample or the lightest, if fractionation is clear? If the authors accept localized degassing in troilite but not mantle-wide degassing, they need to explain why one process occurs while the other does not.

Response: Thank you for raising this important point. We choose to report the average $\delta^{34}\text{S}$ for CE-6 basalts rather than relying solely on the most primitive or lightest troilite value for the following reasons: (1) Our in situ sulfur isotope analyses have an uncertainty of approximately $\pm 0.50\text{‰}$. The most primitive $\delta^{34}\text{S}$ value measured is 0.77‰ , and the lightest is 0.30‰ . Initially, we calculated a mean of $0.93 \pm 0.22\text{‰}$ (2SE) across 19 analyses. However, the highest $\delta^{34}\text{S}$ point (2.32‰) may have been influenced by magmatic or impact-induced degassing; thus, excluding that outlier yields a mean $\delta^{34}\text{S}$ of $0.83 \pm 0.16\text{‰}$ (2SE) from 17 points. Given our measurement precision and the inference of limited degassing in CE-6 basalts, these values remain broadly consistent within uncertainty. (2) Averaging multiple data points provides a statistically more robust representation of the bulk sulfur isotopic composition than a single-spot analysis. Furthermore, an average value enables more meaningful comparisons with prior whole-rock $\delta^{34}\text{S}$ measurements for mare basalts.

Previous research on Apollo mare basalts indicates that the degassing of mare basalts is generally minimal (Wing and Farquhar, 2015; Gargano et al., 2022; Z. Wang et al., 2024a). If degassing does occur, it is likely confined to surface lava flow conditions, where rapid cooling forms a crust that restricts substantial volatile loss. Consequently, degassing effects tend to be localized (e.g., along fractures or flow surfaces) rather than pervasive, leaving most of the basalt and its mantle source unaffected. This scenario explains why individual troilite grains may exhibit localized isotopic variation (e.g., $\delta^{34}\text{S}$ as high as 2.32‰), yet the average composition remains largely unchanged.

Line 179: It is unclear what the authors are referring to in Fig. 2C. Is there petrographic evidence that the primitive pxn isn't an antecryst?

Response: Thank you for your comment. As shown in the figure below (Reply Fig. 4), the basaltic clast displays a subophitic texture, where pyroxene is intergrown with plagioclase. This textural relationship suggests that the pyroxene crystallized in place rather than being an antecryst. There is no petrographic indication—such as abrupt compositional zoning or discordant grain boundaries.



Reply Figure 4. The SEM picture of the primitive clinopyroxene in a basalt clast.

Line 191: What constitutes "substantial" degassing in this scenario? A $\sim 1.2\text{‰}$ difference between samples suggest some degree of degassing, yet this is dismissed. Additionally, line 186 acknowledges meteorite-induced sulfur modification. How is this reconciled with the idea that sulfur remained unaffected?

Response: The $\sim 1.2\text{‰}$ difference between basaltic samples can be attributed to minimal degassing under effusive conditions (e.g., less than 10% for mare basalts) (Wing and Farquhar, 2015), coupled with the $\pm 0.50\text{‰}$ precision of our in situ sulfur isotope analyses. In other words, this range does not necessarily imply a "substantial" degree of degassing, but rather a level consistent with slow effusive eruptions where any significant volatile loss is largely restricted by rapid surface quenching (Gargano et al., 2022).

When we refer to "meteorite-induced sulfur modification," we are specifically describing breccias that show clear textural or chemical evidence (e.g., elevated Ni, shock textures) of meteoritic contribution.

Line 206: I disagree with the statement that Figure 2a shows a uniform sulfur composition. Equilibrium fractionation at lunar temperatures would not account for a 2‰ range in sulfur isotopes. This variation shows that multiple processes have modified the samples.

Response: Thank you for pointing this out. The breccia samples were clearly affected by impact processes. Therefore, the isotopic homogeneity discussed here refers specifically to the basalt clasts, as well as the Mg-suite and FAN clasts—samples that are more representative of the lunar interior. As discussed before, the $\delta^{34}\text{S}$ values of basalt clasts, mainly ranging from $0.30 \pm 0.51\text{‰}$ to $1.52 \pm 0.37\text{‰}$, can be explained by minor degrees of degassing and the analytical uncertainty ($\sim 0.5\text{‰}$). It overlaps with the Mg-suite and FAN clasts. Thus we can say that the Moon's interior from the farside overall exhibits sulfur-isotope homogeneity.

Lines 213–219: These sentences are unclear. Zhang et al. (2024) showed that the

CE-6 samples originated from mantle reservoirs that experienced substantial differentiation and volatile redistribution, possibly due to impacts. How can these processes not affect sulfur isotopes?

Response: Recent work suggests that the Mg-suite in CE-6 soils may have formed from impact melt associated with the South Pole–Aitken (SPA) collision (Su et al., 2025), a process that could involve significant melting and volatile loss. However, our sulfur isotope results indicate that troilite in these Mg-suite clasts has $\delta^{34}\text{S}$ values consistent with both CE-6 basalts and nearside Apollo samples. Within the limits of our current data, this suggests that any sulfur-isotope modification tied to the SPA impact was either minimal or did not persist within these Mg-suite lithologies. More work is still required on this topic.

Line 237: Figures 4e and 4f do not exist. This needs to be changed.

Response: revised.

Line 237: Why would sulfur isotopes remain homogeneous after large-scale mantle processing when Pb heterogeneity in KREEP basalts suggests that these melts underwent fractionation processes?

Response: Thank you for your comment. Here, the heterogeneity of Pb isotopes primarily reflects the variability among different lunar basalt mantle source regions, which is mainly caused by the varying degrees of KREEP (K, REE and P-rich, represent the latest melt of lunar magma ocean) component incorporation into these sources. On the other hand, existing studies indicate that whether in the lunar magma ocean or lunar basaltic magmas, sulfides typically reach saturation only in the late stages of magma evolution (Ding et al., 2018; Ji and Dasgupta, 2025), provided that Fe-rich, S-poor, or Ni-rich sulfides do not form—types of sulfides that are rarely reported in lunar basalts. Consequently, the differentiation process of lunar magmas has minimal impact on sulfur isotope compositions, allowing them to remain relatively homogeneous even after large-scale mantle evolution.

Lines 251–256: These sentences need clarification. The manuscript should provide a more in-depth explanation of how sulfur content and other volatile element isotopic compositions can vary significantly, yet the lunar interior can remain homogeneous in sulfur isotopic composition.

Response:

We thank the reviewer for pointing out the need for clarification. The variation in sulfur content across different lunar lithologies likely reflects the effects of magma ocean crystallization, mantle overturn, and subsequent partial melting, which can redistributed sulfur. Crucially, the absence of early, large-scale degassing or sulfide segregation allowed the $\delta^{34}\text{S}$ signature of the bulk silicate Moon to remain largely preserved (Z. Wang et al., 2024a).

Moreover, previous studies have shown that mare basalts unaffected by meteoritic contamination exhibit relatively narrow ranges in other volatile isotope systems, such

as $\delta^{66}\text{Zn}$ ($1.4 \pm 0.5\%$; Kato et al., 2015), $\delta^{41}\text{K}$ ($-0.08 \pm 0.09\%$; Tian et al., 2020), and $\delta^{87}\text{Rb}$ ($0.05 \pm 0.12\%$; Pringle and Moynier, 2017). These observations collectively suggest that while the abundance of volatile elements may vary due to secondary processes, their isotopic compositions—established during the Moon's early differentiation—remain relatively uniform across the lunar interior. More work still need to be done in lunar farside samples to test the model.

Figure 1: Move the Mg# values of pyroxenes closer to their labels to improve readability.

[Response:](#) revised.

Figure 2: Label the blue region. Why aren't all the Chang'e-6 data grouped together at the top?

What are the reported $\delta^{34}\text{S}$ values for the Chang'e-6 non-mare basalts?

[Response:](#) revised.

Figure 4a: The CE-5 label is missing, please correct this.

[Response:](#) We didn't show CE-5 magmatism in Fig. 4a, because it is included in the Low-Ti mare basalts in the nearside of the Moon.

Reviewer #2 (Remarks to the Author):

Review of the manuscript "Sulfur isotopes from the lunar farside reveal a global giant impact signature" by Li et al.

This manuscript investigates the sulfur content and $\delta^{34}\text{S}$ isotopic composition of the returned farside soil sample collected by the Chang'e-6 mission. Troilite grains in selected basalt clasts, nonmare clasts (Mg suite and FAN) and breccias from the sample were analyzed. The $\delta^{34}\text{S}$ values of these grains are consistent with those observed in CE5 basalts, Apollo high-Ti and low-Ti basalts, and lunar meteorites, indicating a homogeneous $\delta^{34}\text{S}$ composition in the lunar interior, which is approximately 2‰ heavier than Earth's sulfur isotope signature. In contrast, S concentrations in the lunar mantle are heterogeneous as indicated by S/Dy ratio variations. The S concentration in the CE6 mantle source is estimated to be 120 ppm, the highest among the mantle sources of CE5, Apollo high-Ti and low-Ti basalts and lunar meteorites. The authors proposed that the consistently high and homogeneous $\delta^{34}\text{S}$ composition of the Moon suggests that this characteristic has remained stable since the Moon's formation, a signature from the giant impact.

This work presents a comprehensive investigation into the S isotope composition in the CE6 soil sample and the data are of high quality. While this work is of broad interest, I have several major concerns related to data interpretation that should be considered before its publication. Additionally, there are numerous typos throughout the manuscript and many figure citations are out of order.

Recommendation: Major revision.

Response:

Thanks for your constructive feedback. According to your comments, we outline the major revisions we have made in our manuscript:

1. We incorporated a detailed discussion on the potential effects of core–mantle differentiation on the BSM sulfur isotopic composition. Existing experimental and theoretical studies suggest either a direction of fractionation opposite to our observations or a very small magnitude of isotope fractionation ($<0.05\text{‰}$), both of which are insufficient to explain the $\sim 2\text{‰}$ $\delta^{34}\text{S}$ offset between the Moon and Earth.

2. We assessed the effects of degassing, sulfide saturation, and partial melting processes on sulfur isotope compositions, as discussed in the response above.

3. We evaluated the role of late accretion in modifying the lunar sulfur isotope composition. Given the limited mass of late-accreted material and the mismatch between chondritic $\delta^{34}\text{S}$ values and those of the BSM, we conclude that late accretion cannot account for the observed Earth–Moon sulfur isotope fractionation.

Finally, we expanded our discussion to include sulfur loss during the giant impact, using Rayleigh distillation modeling to estimate the degree of sulfur volatilization ($\sim 20\text{--}75\%$) required to produce the observed $\delta^{34}\text{S}$ difference between Earth and the Moon. This scenario provides the most plausible explanation for the heavy sulfur isotope signature of the BSM.

Major comments:

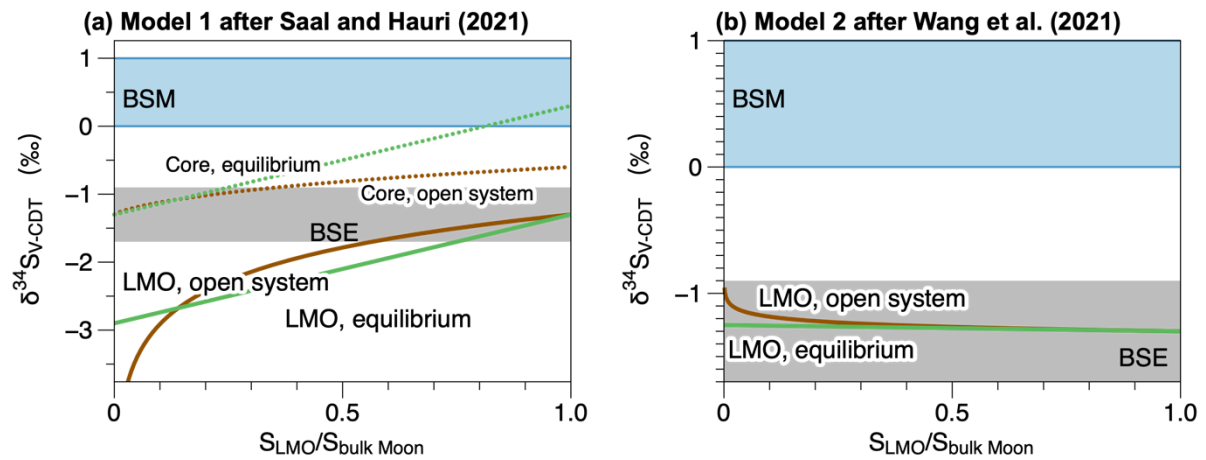
First, S is not only a highly volatile but also a siderophile (iron-loving) element. During the lunar core formation, S isotope fractionation between the core and BSM is expected. This fractionation will significantly affect the lunar mantle's $\delta^{34}\text{S}$ composition and its homogeneity, but a discussion on this process is critically lacking in this manuscript.

Response: Thank you for highlighting this important point. We agree that sulfur isotope fractionation during lunar core formation is a critical and still debated issue (Labidi et al., 2013; Wang et al., 2021), and we have now added a discussion on this topic in the revised manuscript.

Based on previous studies, we consider that core–mantle differentiation can't explain the observed $\sim 2\text{‰}$ $\delta^{34}\text{S}$ difference between the Moon and Earth. The rationales are as follows:

(1) According to the modeling by Saal and Hauri (2021), assuming an initial $\delta^{34}\text{S}$ composition of the Moon equivalent to the Earth's mantle value (they adopted a chondritic initial value, though this does not significantly affect the conclusion), a metal–silicate melt sulfur partition coefficient of ~ 100 , and sulfur isotope fractionation factors ($\alpha_{\text{core-LMO}}$) of 1.0016 for batch equilibrium and 1.0007 for an open system, the

resulting LMO becomes progressively enriched in lighter sulfur isotopes during core segregation (Reply Fig.5a). This prediction is inconsistent with our observed heavier $\delta^{34}\text{S}$ values in the lunar mantle.



Reply Figure 5. Core-LMO sulfur partitioning model equilibrium or open system. The $\delta^{34}\text{S}$ values (‰) of the LMO and lunar core are plotted against the ratio of the sulfur content of LMO during core segregation to that of the bulk Moon. Model 1 (a) is after Saal and Hauri (2021), an equilibrium sulfur isotope fractionation factor $\alpha_{\text{core-mantle}}$ of 1.0016 and 1.0007 for batch equilibrium and open system, respectively. The different is that we start with a BSE $\delta^{34}\text{S}$ value of 1.3 ± 0.4 ‰. Model 1 (a) is after Wang et al. (2021), using the the sulfur isotope fractionation factor $\alpha_{\text{core-mantle}}$ of 0.99995, our calculatong demonstrated that the fractionation bewteen the core and silicate mantle is much smaller than the ~ 2 ‰ offset we observe.

(2) Wang et al. (2021), based on first-principles calculations, suggested that under reducing conditions—relevant to lunar and terrestrial core formation—the $\delta^{34}\text{S}$ fractionation between the core and silicate mantle is very small (<0.05 ‰). Their calculated fractionation factor ($\alpha_{\text{core-LMO}} \approx 0.99995$) would result in a $\delta^{34}\text{S}$ difference between the BSM and BSE of less than 0.1 ‰ (Reply Fig.5b), much smaller than the ~ 2 ‰ offset we observe.

Together, both models suggest other process is required to explain the present-day $\delta^{34}\text{S}$ difference between BSE and BSM.

Second, there are many important processes can alter $\delta^{34}\text{S}$ in the CE6 magma which however are excluded without strong justifications. For example, I do not think magma degassing can be simply ruled out just because one troilite data point near primitive cpx shows roughly consistent $\delta^{34}\text{S}$ value with others. A study on troilites from CE5 samples has shown a decreasing trend of $\delta^{34}\text{S}$ in the magma during crystallization and degassing, with a total of 40% original S loss (Liu et al. 2022, JGR Planets). This amount of S degassing is not negligible. Furthermore, the high S/Dy ratio of CE6 and high S abundance in the CE6 mantle source may indicate that the source region for CE6 magma is sulfide saturated, which will lead to a lighter $\delta^{34}\text{S}$ in

the produced melts. The lack of such justifications leaves the argument in the manuscript incomplete and unconvincing.

Response: In the revised manuscript, we have expanded our discussion to provide more detailed support for this claim:

1. Evidence for Limited Degassing in CE-6 Basalts

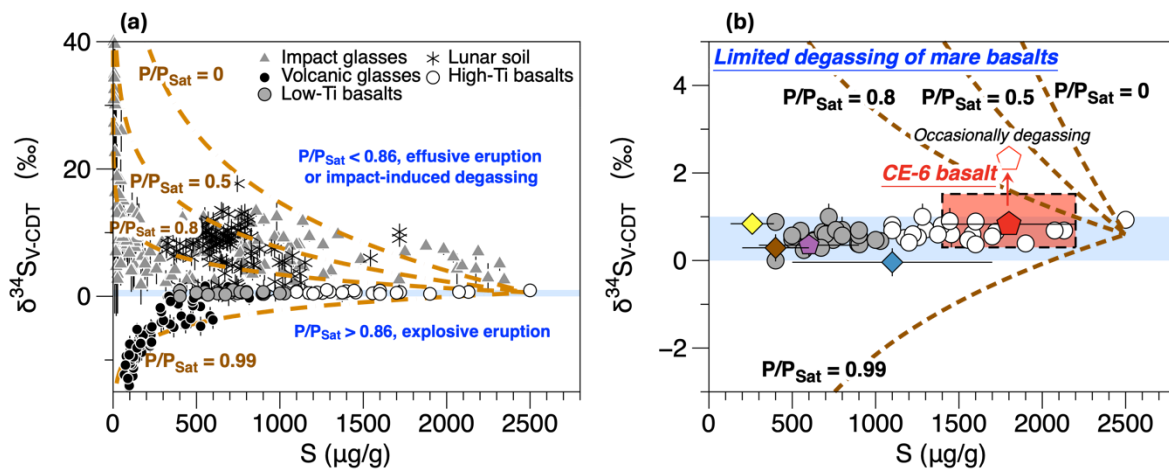
1.1 Degassing Regimes for Different Eruption Styles

1.1.1 Explosive Eruptions (e.g., Volcanic Glasses).

Explosive volcanism are characterized by high ascent rate explosive volcanic eruptions via rapid dike propagation driven by CO and H₂, which resulted in widespread pyroclastic deposits such as picritic glasses. Saal and Hauri, (2021) propose a degassing regime with an effective isotope fractionation factor of

$$\alpha'_{kin} = (\alpha_{kin} - 1) \left(1 - \frac{P}{P_{Sat}}\right) + 1$$

where P/P_{Sat} is equal to the effective vapor pressure of a given element relative to the saturation vapor pressure (Saal and Hauri, 2021). Under the condition of explosive eruptions, degassing occurs at near-saturation vapor pressures ($P/P_{Sat} \approx 1$), where sulfur-isotope fractionation is dominated by equilibrium processes ($\alpha_{gas-melt} \approx 1.003$), leading to progressively lighter residual melts in terms of $\delta^{34}S$ (Reply Fig. 3a).



Reply Figure 3. Sulfur isotope degassing under different eruption (effusive and explosive) and impact conditions. The modeled curves (brown dashed lines) with different P/P_{Sat} are based on the lunar sulfur isotope degassing model from Gargano et al. (2022) and Saal and Hauri (2021) using an initial $\delta^{34}S$ value of 0.6‰ from Wang et al. (2024) and an initial sulfur content of 2500 $\mu g/g$, representing the upper limit for lunar basalt. In panel (b), the red shaded area denotes the $\delta^{34}S$ and sulfur content range observed in CE-6 basalts. Data for impact glasses and lunar soils are

from Wang et al. (2024) and references therein, while volcanic glass data are sourced from Saal and Hauri (2021).

1.1.2 Effusive Eruptions (e.g., Mare Basalts) and Impact-Induced Evaporation (e.g., Impact Glasses and Breccias).

In contrast, CE-6 basalts are interpreted as classic mare basalts emplaced through effusive eruptions. If degassing occurred, it would mainly happen at the lunar surface under low P/P_{Sat} (Wing and Farquhar, 2015; Gargano et al., 2022), where kinetic fractionation prevails—similar to the mechanisms observed in impact-related sulfur volatilization. Even a small amount of sulfur loss (~20%) can markedly increase $\delta^{34}\text{S}$ in the residual melt (e.g., a +7‰ shift under vacuum) (Gargano et al., 2022). Indeed, the pronounced sulfur-isotope fractionation observed in lunar impact glasses and soil highlights the potential role of impact processes in driving volatile loss (Reply Fig. 3a).

1.2 Limited Degassing in CE-6 Basalts

Despite the potential for kinetic fractionation, in situ troilite $\delta^{34}\text{S}$ values in most CE-6 basaltic clasts range from $0.30 \pm 0.51\text{‰}$ to $1.52 \pm 0.37\text{‰}$ (given the external uncertainty in this study is 0.50 ‰ for $\delta^{34}\text{S}$). This relatively narrow range implies minimal overall sulfur loss (<10%, Reply Fig. 3b), consistent with findings from Apollo mare basalts and their S, Cl, and Zn isotope data (Wing and Farquhar, 2015; Gargano et al., 2020), which also point to low degrees of degassing. One likely explanation is that thicker mare lava flows formed quenched crusts, limiting volatile escape and thus reducing isotopic fractionation despite the lunar surface's low ambient pressure.

One troilite grain displays a higher $\delta^{34}\text{S}$ value ($2.32 \pm 0.47\text{‰}$), potentially resulting from localized degassing via open fractures or post-crystallization impacts, as suggested by the clast's highly fractured texture (Fig.S5c). Such localized processes can produce isolated, elevated $\delta^{34}\text{S}$ values without undermining the broader conclusion of low overall degassing. Breccias, with a relatively higher mean $\delta^{34}\text{S}$ ($1.24 \pm 0.31\text{‰}$, 2SD, $n = 14$), likely record degassing caused by meteoritic impacts, though the magnitude of fractionation indicates only limited impact-related alteration. Furthermore, the lowest $\delta^{34}\text{S}$ value in the breccias ($0.30 \pm 0.40\text{‰}$) overlaps with the range observed in CE-6 basaltic troilite, suggesting a consistent initial sulfur-isotope composition across the CE-6 suite.

2. Sulfur-Undersaturated during partial melting of mantle source

As discussed in our previous response, the CE-6 basalts evolved under sulfur-undersaturated conditions and limited degassing. Therefore, their measured sulfur content ($1800 \pm 400 \mu\text{g/g}$) likely reflects the upper range of sulfur concentration in melts derived from partial melting of their mantle source. This value is significantly lower than the sulfide saturation sulfur concentration (SCSS) expected for basaltic melts under lunar mantle conditions (>2600 ppm) (Ding et al., 2018).

Assuming a melt fraction of 3–5%—similar to that inferred for CE-5 basalts—the sulfur content of the mantle source would be approximately 60–130 ppm. This

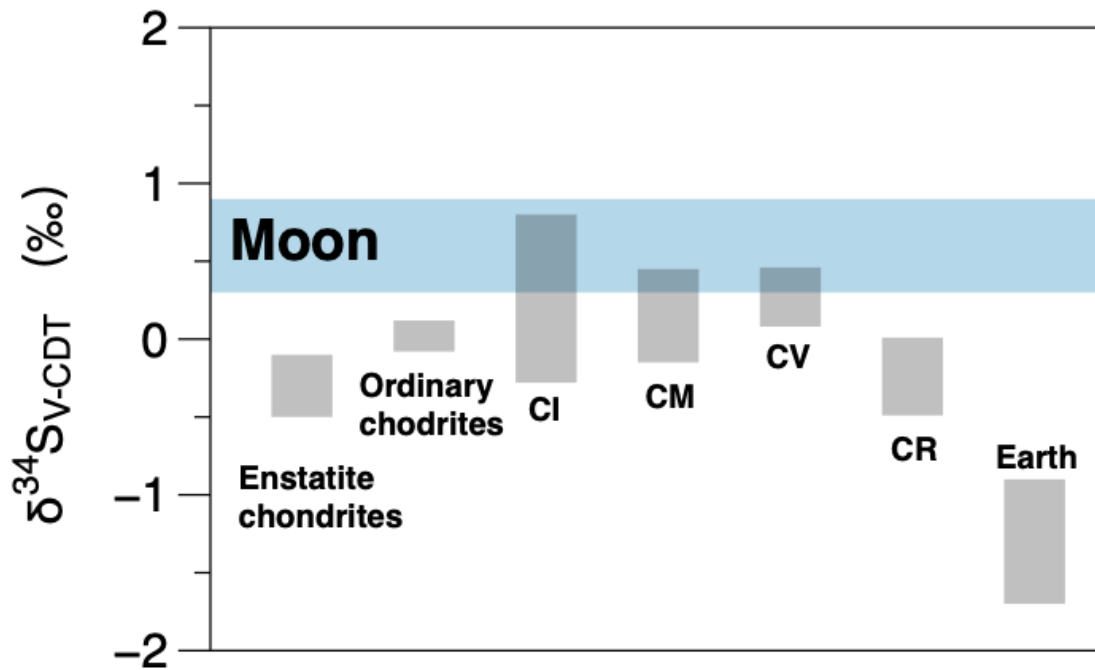
estimate is consistent with values inferred from S/Dy systematics ($120 \pm 30 \mu\text{g/g}$) and falls within the range proposed for the mantle sources of Apollo basalts. Moreover, Ding et al. (2018) demonstrated that sulfides are likely exhausted in the source regions of most lunar basalts during their formation. Additionally, at high temperatures ($\sim 1300^\circ\text{C}$), sulfur isotope fractionation between melt and sulfide is minimal ($\alpha_{\text{melt-tro}} \approx 0.999$) (Marini et al., 2011), indicating that partial melting of lunar mantle source would have negligible impact on the sulfur isotopic composition of the resulting melt (Saal and Hauri, 2021).

Finally, for a paper published in Nature Communications, I look forward to a broad discussion on the origin of the Moon. For example, it has been proposed that water in the interiors of the Earth and Moon may have been inherited from carbonaceous chondrites due to their indistinguishable hydrogen isotope compositions (Saal et al. 2015 Science). What are the S isotope compositions of carbonaceous chondrites and what is the consequence of this hypothesis to the S isotopes in the Moon? In addition, late veneer has been proposed to explain the homogeneous distribution of highly siderophile elements with chondritic proportions in the lunar mantle sources of mare basalts. It would be nice to see discussions on these topics and give a broad view on the formation of the Moon from the perspective of $\delta^{34}\text{S}$ in CE6. The current discussion on the signature from the giant impact is a little shallow and ambiguous - do the authors mean the signature from Theia, or the proto-Earth, or the mixture, or the processes during the impact?

Response: Thank you for this excellent and thought-provoking comment. We have expanded the discussion in the revised manuscript to address the broader implications of sulfur isotopes for the origin of the Moon, as suggested.

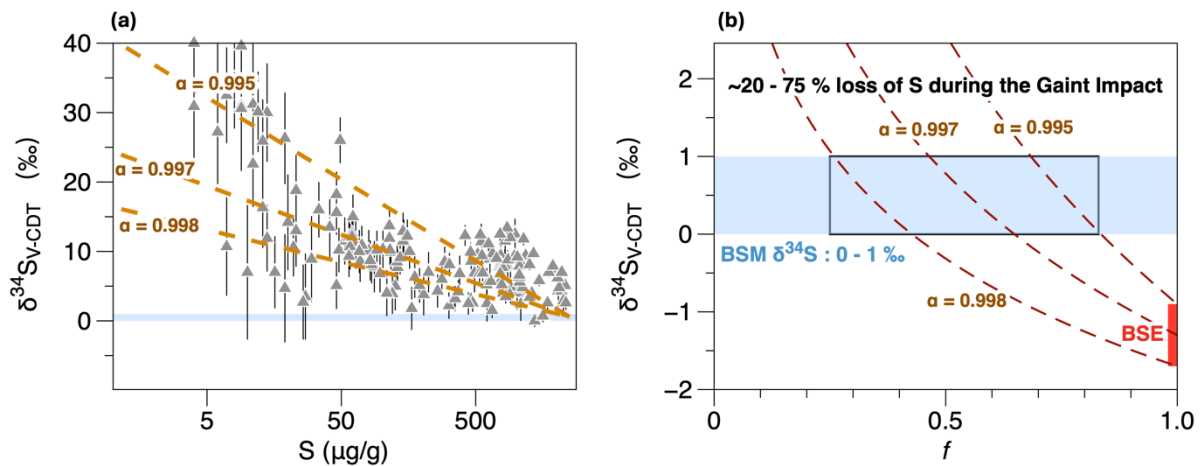
The similarity isotopic anomalies between the Earth and the Moon in O, Cr, Ca and Ti strongly supports the idea that both bodies share a common primordial source (Young et al., 2016; Schiller et al., 2018; Mougél et al., 2018; Zhang et al., 2012). Therefore, the $\sim 2\%$ $\delta^{34}\text{S}$ offset observed between the BSE and the BSM is more likely to reflect secondary processes that occurred during or after the Moon-forming impact. These processes include core–mantle differentiation, late accretion, and volatile loss during the impact. We have already discussed the limited role of core–mantle differentiation in a previous response, and here we focus on the possible contributions from late accretion and volatile loss.

Day and Walker (2015) reported that the highly siderophile element (HSE) of mare basalts suggest that the amount of material added during late accretion represents only a small fraction ($\sim 0.02 \text{ wt.}\%$) of the Moon's mass. Further study estimated that the sulfur contributed by this late-accreted material is likely less than 10% (Gleißner et al., 2022). As shown in the Reply Fig. 6, only CM and CO carbonaceous chondrites show $\delta^{34}\text{S}$ values that slightly overlap with BSM. However, assuming a BSE-like initial $\delta^{34}\text{S}$ and a 10% contribution from these chondritic sources, it is difficult to account for the full $\sim 2\%$ $\delta^{34}\text{S}$ difference between the Earth and Moon. More importantly, late accretion cannot explain the $\delta^{34}\text{S}$ of the Earth, which experienced more extensive late accretion (Day and Walker, 2015). Therefore, we conclude that late accretion likely played only a minor role in establishing the lunar sulfur isotopic signature.



Reply Figure 6. The $\delta^{34}\text{S}$ value of different chondrites. Only CM and CO carbonaceous chondrites show $\delta^{34}\text{S}$ values that slightly overlap with BSM. Data source: data of enstatite chondrites are from (Gao and Thiemens, 1993a, b). Data of enstatite chondrites are from (Defouilloy et al., 2016). Data of CI, CM, CO and CR are from (Alexander et al., 2022).

Instead, we propose that the ^{34}S enrichment in the Moon was primarily caused by volatile loss during the giant impact as indicated by other volatile elements (e.g., Zn, K and Rb). Based on the $\delta^{34}\text{S}$ of lunar impact glasses (Cui et al., 2024), we can yield isotopic fractionation factors ($\alpha_{\text{gas-melt}}$) in the range of 0.995–0.998 for S isotopes fractionation during impact events (Reply Fig. 7a). Applying these fractionation factors, we estimate that 20–75% of lunar sulfur would have been lost during the giant impact to produce the observed $\delta^{34}\text{S}$ value of the BSM. Assuming a present-day mantle sulfur content of ~67–80 ppm (Ding et al., 2018; Gleißner et al., 2022; Su and Zhang, 2024), this implies that the Moon originally contained approximately 100–300 ppm sulfur prior to volatile loss. This result supports the hypothesis of incomplete loss of highly volatile elements during an the giant impact (Pahlevan, 2014; Nakajima and Stevenson, 2018). As Nakajima and Stevenson (2018) proposed that heavier species in the upper layers of the Moon-forming disk could limite the diffusion of volatiles, leading to inefficient depletion of volatiles from the disk.



Reply Figure 7. Sulfur degassing during surface impact processes and the giant impact event. According to the Rayleigh distillation equation: $\delta^{34}\text{S} = (1000 + \delta^{34}\text{S}_0) \times f^{(\alpha-1)} - 1000$, where $\delta^{34}\text{S}_0$ represents the initial sulfur isotopic composition and f is the remaining fraction of sulfur after evaporation. In this calculation, we use an initial $\delta^{34}\text{S}_0$ of $-1.3 \pm 0.4\text{‰}$ (representative of the BSE) and an initial sulfur content of $2500 \mu\text{g/g}$. Based on the observed relationship between sulfur content and sulfur isotopic composition in lunar impact glasses, the sulfur isotope fractionation factor (α) during impact-induced volatilization can be constrained to a range of 0.995–0.998. Using this range of α values, we estimate that achieving the present $\delta^{34}\text{S}$ value of the BSM would require approximately 20–75% sulfur loss during the giant impact event.

Specific comments:

L59-63: There are many new lunar melt inclusion studies on volatile abundances, such as those by Ni et al. (2019, GCA) and Su and Zhang (2024, GCA). The significant heterogeneity of H_2O in lunar melt inclusions, for example, is interpreted to result from differences in cooling rates, with samples that cooled the fastest best preserving the pre-eruptive volatile contents (Su and Zhang, 2024).

Response: Thank you for this helpful comment. The studies by Ni et al. (2019) and Su and Zhang (2024) provide important insights in our work. We have now cited and widely discussed these works in the revised manuscript. Additionally, a study from Ni et al. (2017) that H_2O concentrations in lunar melt inclusions in olivine can significantly lose in minutes during homogenization experiments, while F, Cl and S in the inclusions are well preserved.

640

641

642 L79-80: Can you give numbers to quantify “relatively limited”? In addition, basalts
643 from CE5 are estimated to have lost ~40% of their original S, which is not the case
644 as justified here.

645

646 **Response:** According to the degassing model for mare basalts shown in Reply Fig.
647 3b, a $\delta^{34}\text{S}$ shift of $\sim 1.2\text{‰}$ requires only a very limited extent of degassing, consistent
648 with the $<10\%$ degassing inferred from bulk-rock sulfur isotope studies of mare
649 basalts (Wing and Farquhar, 2015). The sulfur isotopic variability observed in CE-5
650 basalts may reflect analytical uncertainty associated with in situ measurements
651 ($\sim 0.6\text{‰}$ for NanoSIMS), as well as potential effects of intra-grain diffusion and
localized degassing within troilite (Wang et al., 2024a).

652

653 L93: where does the “90%” information come from? Give relevant references.

654

655 **Response:** 90% of the sulfur budget in CE-5 basalts was estimated by Wang et al.,
656 (2024a), based on sulfur concentrations in troilite, apatite and late-stage mesostasis.
657 However, for CE-6 basalts, a higher proportion of sulfur is likely hosted in sulfides.
658 This is because CE-6 basalts are notably depleted in incompatible elements (e.g.,
659 REEs), implying minimal apatite and late-stage mesostasis phases. Consequently,
most of the sulfur in CE-6 basalts is expected to reside in sulfide minerals.

660

661 L110: I do not see any illustration of basalt clasts, mineral fragments, and impact
662 glasses in Figs. 1d and 1e except troilite.

663

Response: revised.

664

665 L115: Fig. S2a instead of Fig. S1a?

666

Response: revised.

667

668 L146: Fig. 2b instead of Fig. 2c?

669

Response: revised.

670

671 L157: Saal and Hauri (2021, Science Advances) report S concentration and $\delta^{34}\text{S}$ of
672 lunar olivine-hosted melt inclusions. It is generally thought that melt inclusions
673 represent pre-eruptive (undegassed) volatile contents. The $\delta^{34}\text{S}$ data from these
674 melt inclusions should be discussed.

675

676 **Response:** Thanks for your excellent suggestion! Olivine-hosted melt inclusions from
677 Saal and Hauri (2021) exhibit $\delta^{34}\text{S}$ values ranging from $-0.30 \pm 0.91\text{‰}$ to $1.6 \pm$
 1.81‰ . These values closely match the troilites in our study ($0.30 \pm 0.51\text{‰}$ to $1.52 \pm$

678 0.37‰), further supporting the interpretation that the CE-6 basalts experienced
679 limited sulfur degassing.

680
681 L163: I do not understand why Figs. 2c is listed here.

682 Response: revised.

683
684 L217-219: I suggest not extrapolating from S to all volatiles and their isotopes. In
685 addition, this work only reports 1 data point of Mg suite clasts from the CE6 soil
686 sample; whether this data point is representative is unknown.

687 Response: We agree that it is not appropriate to broadly extrapolate from sulfur to
688 all volatiles and their isotopes. Accordingly, our revised discussion now focuses on
689 sulfur abundance and isotopic composition.

690 Regarding the Mg-suite clast, we acknowledge that only one data point was
691 obtained, as the troilite grains within this clast are small and pose analytical
692 challenges for sulfur isotope measurements. Nevertheless, recent studies suggest
693 that this clast may represent a direct product of the SPA impact melt sheet (Su et
694 al., 2025), making it a valuable data point for preliminary discussion.

695 We have clarified this context in the revised manuscript and emphasize that further
696 isotopic studies on additional Mg-suite clasts and other farside lithologies are
697 essential for broader interpretations.

698
699 L226: Figs. 3c, d instead of Figs. 3a, b?

700 Response: revised.

701
702 L227: Pyroxene-hosted melt inclusions from lunar very-low-Ti basalts have a S/Dy
703 ratio from 91-296 (Su and Zhang 2024 GCA).

704 Response: Here, we use the range of 50–200, because in the original study they
705 recommended S/Dy values (50–200) based on melt inclusions with high MgO
706 contents (6–16 wt%).

707
708 L232: Fig.3b instead of Fig.3c?

709 Response: revised.

710
711 L491-492: The Mg# of Cpx [and Opx].

712 Response: revised.

713

714 L493: Fig.S1 instead of Fig.S3?

715 [Response:](#) revised.

716

717 Fig.1: Please also add the An# of plag.

718 [Response:](#) revised.

719

720 Fig.2a: 1) Are the data from CE5 basalts also from this study? Clarify this in the
721 figure caption. 2) Plot Mg suite and Fan separately and add the $\delta^{34}\text{S}$ values to the
722 figure like others for comparison.

723 [Response:](#) revised. CE-5 data are from(Z. Wang et al., 2024a) and Liu et al., (2022).

724

725 Fig.3: Wing and Farquhar (2015) also report S and $\delta^{34}\text{S}$ of a series of lunar high-Ti
726 and low-Ti basalts. These data should be added and discussed.

727 [Response:](#) Thanks for your suggestion. S and $\delta^{34}\text{S}$ data have already been
728 included in the figures and widely discussed in the manuscript.

729

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The authors have satisfactorily addressed most of my previous concerns by adding comprehensive discussion on the effects of core-mantle differentiation, late accretion, volatile degassing, sulfide saturation, etc. Considerable effort has gone into the revision of the manuscript. The revised manuscript is clear and well-written.

Response: We sincerely thank the reviewer for their thoughtful comments and constructive suggestions. Your feedback has significantly helped us improve the clarity and completeness of the manuscript. We truly appreciate your time and kind support.

I only have one comment for the authors:

Fu and Dottin (2025 LPSC) modeled the coevolution of S abundance and $\delta^{34}\text{S}$ of the BSM in the context of giant-impact models. This work indicates that volatile loss from the giant impact event produces an increase of up to $\sim 1\text{‰}$ in $\delta^{34}\text{S}$, which is insufficient to explain the 2‰ $\delta^{34}\text{S}$ enrichment of the BSM. The work of Fu and Dottin (2025 LPSC) is highly relevant to this work and should be discussed.

Response: We thank the reviewer for this valuable suggestion. Fu and Dottin (2025 LPSC) proposed, based on their modeling, that the giant impact could lead to an enrichment of up to $\sim 1\text{‰}$ in the $\delta^{34}\text{S}$ value of the bulk silicate Moon (BSM). This result is broadly consistent with our main conclusion—that the giant impact induced large-scale volatile loss and isotopic fractionation on a global scale. The difference lies in the magnitude of the enrichment: while their model predicts $\sim 1\text{‰}$, our geochemical data show a $\sim 2\text{‰}$ $\delta^{34}\text{S}$ offset between the BSM and the bulk silicate Earth (BSE). As illustrated in Figure 4a of our manuscript, sulfur isotope fractionation during volatile loss can be substantial, and a 1‰ variation may easily result from differences in input parameters or modeling assumptions. However, as Fu and Dottin's work is currently only available as a conference abstract and does not include detailed calculations or derivations, it is difficult for us to fully evaluate the robustness of their result. We have added a brief discussion of their findings in the revised manuscript.