

Space-weathered rims on lunar ilmenite as an indicator for relative exposure ages of regolith



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Decision letter and referee reports: first round

22nd Mar 24

Dear Professor Li,

Your manuscript titled "Diverse space-weathered rims on ilmenite: constraining the exposure history of Chang'e-5 lunar soils" has now been seen by 3 reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication. In particular for publication in Communications Earth & Environment, we request that you:

(1) Provide a comprehensive overview of the related literature and method details, including the software used and sample thickness information, and (2) strengthen the interpretation of your findings based on statistics and expand your discussion accordingly.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and the completed checklist:

link redacted

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We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

Please do not hesitate to contact us if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Candice Bedford, PhD

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

See attached.

Reviewer #2 (Remarks to the Author):

This manuscript focuses on the chemical and microstructural characterization of ilmenite grains returned by the Chang'e-5 mission through transmission electron microscopy, energy dispersive X-ray spectroscopy, and electron energy loss spectroscopy. Ilmenite grains exhibit different degrees of space weathering and their alteration has been attributed to the influence of the solar wind on the lunar surface. In addition, the different degrees of alteration were used to reconstruct the stratigraphic profiles from the sampling region to elucidate regolith overturn processes.

Additionally, the results shown in the manuscript are novel as no studies report different degrees of space weathering on a singular mineral phase from returned samples. The results from this manuscript will contribute to a better understanding of the space weathering processes on the Moon.

Besides the valuable scientific content of the manuscript, I recommend looking for editing services to improve the readability of the manuscript. Please refer to the attached document "comments to authors" where you will find my questions and suggestions about the manuscript.

Reviewer #3 (Remarks to the Author):

Review of “Diverse space-weathered rims on ilmenite: constraining the exposure history of Chang’e-5 lunar soils” By Guo, Z. et al

Reviewed by K. Burgess

This manuscript presents TEM data of lunar ilmenite grains showing varying degrees of space weathering by solar wind irradiation. The imaging combined with EDS and EELS data provide evidence of how ilmenite rims form, from barely altered surfaces to intensely weathered, vesicle- and npFeO -rich rim more than 100 nm thick. The authors relate these differences in weathering to models of Chang’e-5 regolith emplacement and timing. The data presented support the overall conclusions and are an important contribution to our understanding of this understudied but important lunar phase. The results fit well within current understanding of solar wind irradiation processes and ilmenite rim structure, and clarify how intermediate space weathering stages can be recognized. The most novel part of the research is the attempt to directly link depth within the regolith (core) for each sample to ejecta from specific local craters and their exposure and timing.

However, several key points were not addressed by the paper. Previous work on ilmenite, which is cited, has shown clear importance of crystal orientation for the depth of solar wind irradiation rims (Burgess & Stroud, 2018). This aspect of the grains here is not discussed, even as the difference in rim thickness is used to assume different levels of exposure. In the “ NpFeO -rich rim”, the most heavily altered outer rim is in fact thicker than the outer rim of the “Deeply Damaged rim”, as Fig. 4 also demonstrates, even as planar defects are deeper in the latter. This does not alter the overall conclusions, but the less straightforward aspect of this variation as relates to exposure time needs to be addressed.

Additionally, loss of O and increase of Ti seen in EDS data are used as evidence of different rim development stages, but the Methods section and supplementary table do not provide details of errors, software used, or quantification algorithms. Interpretation of both EDS and EELS data require sample thickness information, which is also not noted. This could strongly affect both the ability to detect He and interpretation of changes in Fe valence, eg in Fig. S5c, where npFeO apparently does not contribute to the signal (Ln 148-150) but could be a small portion of the overall thickness and thus its lack is not meaningful.

It is unclear if EDS composition information or the EELS spectra are from spots or summed from maps and, if spots, what the spot sizes are. Small circles are used in Fig. 5, but it is not clear if that is the actual size. Dwell times for each should also be noted. Ilmenite is usually robust against

damage by the electron beam, but the amorphous rim could be less so, which could affect compositional measurements.

I would really love to see how the adjacent mineral phases respond to space weathering and if similar trends can be related clearly to the ilmenite, potentially with “actual” exposure ages calculated for the silicates using solar flare track dating. I understand that would be a different manuscript, but encourage the authors to publish on that aspect of these samples as well.

Several references are incomplete or not formatted correctly, including Ref. 4, 10, 27, 32. In particular, I cannot assess if Ref. 10 and 27 are the appropriate citations where they are used.

Ln 45-46: While there is ongoing discussion of differentiating between different processes contributing to space weathering, the citations here are pre-2000, and significant work has been done on this topic in the past 25+ years, making these not the best references for the current state of the field or what “remains” an unsolved problem.

Ln 56: I love ilmenite but do not see how it is “more complex” compared to the responses of silicates, which can have basically all of these same features (amorphization, loss of O, npFeO formation, vesicle formation). In fact, its simplicity in having so few elements is part of its power in this context, as any depositional rims are easily recognized.

Ln 81: “vesicles...entrained” in the rim implies a process that is not accurate, as it has an implication of “dragging along with” or picking up during movement.

Ln 90: I think the sentence beginning “All of these microscopic characteristics...” is lacking a verb such as “show”?

Ln 137-138: “the densely damaged rims should be a Ti³⁺-bearing oxide” I am confused by this statement and the use of “However” in the following sentence. Does this mean that the Ti³⁺ component would not initially be incorporated into the ilmenite? And as noted above, small FeO metal grains could be very difficult to see in the EELS data. This seems to say that Ti³⁺ oxide could not form without change in Fe valence, but localized loss of Fe (relative increase of Ti) could have similar effects.

Ln 147: “at” should be “in”

Ln 225: I am not sure what “certain orientation” means here. The grain surface?

Ln 229-232: There are several grammatical errors here that make this set of sentences difficult to understand.

Ln 271: Use of “decisive” is confusing here to me. Do you mean it is the only heating required? I would use “sufficient” instead.

Ln 355: Typo on “microns” and “to be stuck” is awkward

Fig. 2b,c: white lines are not labeled. Are they meant to show crystal orientation?

Fig. 3b: scale bar doesn't show

Review of: “Diverse space-weathered rims on ilmenite: constraining the exposure history of Chang’e-5 lunar soils” by Guo et al.

Reviewer: B. Cymes

Summary: This manuscript describes TEM, EELS, and EDS analysis of 11 ilmenite-bearing grains from 5 samples of Chang’e-5 lunar soils collected at depth intervals ranging from 0-72 cm. The authors show a diverse range of space weathered ilmenite rims and have developed an exposure scale based on the interpreted degree of space weathering in the samples. This scale and the samples were then applied to describe the exposure history of the Chang’e-5 landing site. The data presented are of high quality and provide a great range of ilmenite space weathering textures, which would be of moderate interest to the community as most studies focus on smaller grains, but the interpretation of the data and its connection to larger-scale phenomena would be stronger if complimentary techniques were included.

General Comments:

1. Statistical limitations on data interpretation

The surface shoveled sample, CE5C0200, had a <10 μm ilmenite-bearing grain (CE5C0200 #1). This is much smaller than the other grains studied, which were > 100 μm . As the degree of space weathering is correlated to the grain size, it is not surprising that CE5C0002 #1 had clear space weathering features.

My concern is on the statistics used in this study. For example, of the other two samples from CE5C0200, #2 and #3, one had a npFe^0 -rich rim (the 2nd most space weathered classification devised here) and the other was not space weathered. For CE5C0200, this means for grains <10 μm , 100% (n=1) were highly space weathered and grains >100 μm , 50% (n=2) were moderately to highly space weathered. While it is appreciated that they analyzed more than one grain from each sample, there is not good agreement between them. The same can be said for CE5Z0806 and CE5Z0204 where 50% of grains (n=2 each) studied were space weathered.

The conclusions that they are using these samples to support, related to large scale mixing and exposure, would be strengthened if they were combined with other measurements, like FMR, reflectance spectroscopy, and bulk chemistry. The nanoscale TEM data alone limit the conclusions which can be drawn. Inclusion of discussion on solar flare tracks as indicators of exposure history is necessary as it has been used as an exposure metric for several decades. Moreover, it was not clear how the core-sample intervals were chosen, but they are ~20 cm, which is coarse. Nor was it clear why the >100 μm grain size chosen for study.

2. Ambiguous TEM interpretations

A few described features are challenging to see in some images and could very well be thickness artifacts. For example, the vesicles in Fig. 3f and 3h look like contrast variations as

there is no clear edge demarcation. The same goes for the disordered areas in Fig. 3f, 3g, and 3h, these could be thickness variations. In contrast, the vesicles in Fig 2a and 2d are very clear.

Certain features, particularly near the surface, are difficult to see because the samples were coated with gold instead of carbon. The high mobility of Au is concerning in general as it could be deposited on the foil sidewall, potentially causing some of the contrast variations. Coating with Au makes HAADF imaging difficult due to the high Z contrast – see Fig. 5.

Specific Comments:

L41-43: Consider removing the sentence about asteroid reflectance spectra and focus on lunar data.

L53: remove “with both simpler and high symmetry crystal structures”

L54: Ref 11, Zinkle 2012 does not mention ilmenite vs. silicates

L77: How does CE5C0200 #1 fall into the exposure classification scheme used in this study? Is it being used as a sort of space weathering standard?

L122: The slight depletion of oxygen observed is not outside the margin of error of the EDS measurement, particularly for oxygen, which can be difficult to quantify accurately due to absorption effects. Could they report how thick the samples were? The slightly damaged rims would be more accurately characterized as undamaged, or not space weathered.

L143: Provide measurement for “tiny” vesicles

L165: Fig. 2b insert says 2.03 Å, this text says 0.203 nm – the units for lattice spacings should be kept consistent throughout the text. Also, these have one too many digits after the decimal.

L180: How large are the “large” vesicles described and shown in 5b and 5c? None of the ones shown in these samples seem to exceed ~10 nm in diameter, which is not really what we would typically call “large”. How do these large vesicles compare with the smaller/tiny/nano-vesicles described and the large vesicles (up to 40 nm) described in CE5C0200 #1? Clarify

L219-222: Revise this sentence for clarity.

L224-225: What orientation is being referred to here and how does its depth range coincide with the irradiation of solar wind on the lunar surface?

L227: Clarify “at this structural damage” Do they mean associated with a specific type of structural damage?

L229-232: The sentence needs to be revised for clarity.

L232-234: What is this interpretation based on? Is this a hypothesis?

L241-244: Revise for clarity. Also, it would be helpful to define what they're referring to when they say, "large vesicles" "smaller bubbles" and "nanovesicles".

L268: Correct to <400 °C? Also, 400 °C is not what would typically be considered "low temperature"

L270-272: Could they elaborate on this more? Pull out what these sources referenced are saying on this and describe their results in context? Also, what do they mean by decisive? Are they claiming that diurnal temperatures drive npFe⁰ and vesicle formation? Because that is not supported.

L348-349: I do not think this study should be characterized as pioneering, given that microscopists have been conducting such studies for several decades.

L357: Could they describe how the 10 µm grain was used in this study? This sentence is not clear.

L365: Provide a citation to dual-beam method or a brief description of the FIB preparation.

L366: How were the EDS data collected, processed, and quantified? This should be included.

L367: Provide more information on the Talos – imaging mode, accelerating voltage, probe diameter, etc.

L376: What was the probe diameter on the Hitachi?

379: Need to provide more information on how EELS data were collected and processed. Dispersion, exposure, type of collection (spot analyses, line scans, spectrum images?), also how was the background subtracted?

L382: They should also include Fe standards

Fig. 1: Missing description of panel a

Fig 2b inset: Too many significant figures.

Fig 4e: The element map in (e) looks binned when compared to the other maps. Could they be processed in the same way for more accurate comparison?

Fig 5c: The He edge is not clearly above the noise in this spectrum. Could they show background-subtracted spectra?

Supplementary Table 1: Include error in at% measurement. Are these data based on area selections from the maps shown in Fig. 4? Can they show these selections?

Comments to the authors of the manuscript

“Diverse space-weathered rims on ilmenite: constraining the exposure history of Chang’e-5 lunar soils”

Line 21. Replace “source” by “mechanism”

Line 22. Add “the” before “lunar regolith”

Line 23. Can you expand on the pieces of evidence that suggest that ilmenite is highly resistant to solar wind irradiation?

Line 32. Can you explain why only short-term exposure? The deeply damage rims could result from long-term exposure.

Line 38. Add “lunar” before “material”

Line 40. Consider changing “which should strongly alter” to “modifying its reflectance spectrum” or to a similar sentence

Line 41. Add “studies or analysis” after “spectra”

Line 56. Delete “the” before “space weathered”

Line 56. Change “rim” for “rims”

Line 56. Replace “exhibits” for “exhibit”

Line 56-57. You mentioned “ilmenite grains exhibit more complex chemical responses to space weathering than lunar silicate grains”. Add a sentence where you specify what are the structural and chemical differences resulting from space weathering between these two mineral groups.

Line 76. In the manuscript, you never mentioned figures 1b,c,d,e, or f. You could add an introductory sentence where you state that from all the analyzed samples (Fig. 1a,b,c,d,e, and f) CE5C0200 is the sample that is the most space weathered grain.

Line 76-77. Add a sentence expanding on why you think CE5C0200 is the most space-weathered grain

Line 77. Replace “on the lunar surface” by “collected by the Chang’e-5 mission”

Line 79. Add “(Fig. 2)” after “FIB section”

Line 80. Are all npFeO particles elongated? I see npFeO with circular shapes in Fig 1a and Fig 1b. Describe the different shapes present. In addition, where you mentioned “60 nm in length”, is that an average or the maximum length? Again, the npFeO in Fig 1a and b are significantly smaller than that.

Line 81. Did you perform sample tilting (zone axis) or FFT to corroborate that the rim was completely amorphous? Did you do the same for all the samples?

Line 83. Replace “(Fig. 2)” for “(Fig. 2a, b,c,d)”

Line 84. For simplicity, please consider changing “EELS spectra show that the presence of helium is readily recognized (diagnostic peak at about 22 eV) in large vesicles” by “EELS spectra show the presence of helium (diagnostic peak at about 22 eV) in large vesicles” or similar

Line 88. Specify what is the energy of the L2 and L3 peaks

Line 90. Can you explain which characteristics are you referring to?

Line 100. Can you add the Ti EDX map to Fig 1a as you did for the other grains? Or is there an specific reason why you did not included it?

Line 106. Change the colors of the EDX maps on Fig 2. to make it consistent with the EDX maps on Fig.4 and the supplementary material

Line 107. Remove “most space-weathered” from the figure caption

Line 118. Expand on how the rims are structurally consistent with the underlying ilmenite

Line 122. Do you mean Fig 4a instead of Fig 3a? also, add in the text the difference in at% of the O content between the rim and the underlying grain

Line 126. By “this group” do you mean the slightly damage rim category? Reword. In addition, consider replacing “weakest” by a more appropriate word (e.g., lowest)

Line 133. Mention the value of O depletion in at%

Line 133. Consider rewording “the degree of oxygen loss tended to decrease with increasing depth” it is confusing

Line 143. I recommend using “small vesicles” rather than “tiny vesicles”

Line 144. Delete “TEM”

Line 143. What are the sizes and shapes of the vesicles and npFe0?

Line 150. What is the difference between “bubbles” and “vesicles”?

Line 159. Delete “shape”

Line 162 -163. Add the values (at%) of the Ti enrichment and O depletion in the text

Line 164. The sentence “The ubiquitous npFe0 particles in the outer layer are enriched in Fe” is redundant, consider rewording

Line 168. Remove “located beneath the outer layer” it is redundant

Line 182. Remove “located beneath the outer layer” it is redundant

Line 187. Consider changing the word “slender” for “thin” or similar

Line 189. Do you mean “identified” instead of “induced”?

Line 200. Include the O maps in the EDX layered image. Do you have any reason for including Si maps? Consider adding labels to the EDS maps and EELS spectra, maybe like a2, a3 where the BF image is a1. Change the numbering in the manuscript accordingly

Line 200. What are the arrows showing the EELS spectra? In some EELS spectra you include Ti³⁺ and Ti⁴⁺ labels and in others no, be consistent

Line 216. Replace “Given the increasing depth of solar wind action and progressive complexity of microstructure features on ilmenite surfaces, we can classify space-weathered ilmenite, from weakly to strongly weathered rims ... ” with “Based on the thickness and the microstructural complexity of the rims present in the space weathered ilmenite surfaces, we can classify them from the least to the most altered as ...” or similar

Line 219. I think you can change the sentence “By combining previous in-situ ion irradiation simulation experiments and lunar soil observations, we clarifies the specific modifications of different degrees of solar wind irradiation on lunar ilmenite” to “By comparing previous in-situ ion irradiation experiments and analyzes of lunar soils, we could attribute specific characteristics to different degrees of solar wind irradiation on lunar ilmenite” or something similar

Line 223. Expand on what are you referring to as “crystal damage”

Line 225. What do you mean by “certain orientation”?

Line 226. Add a reference about the penetration depth of solar wind particles on lunar surface materials

Line 227. What are you referring to when you mentioned “at this structural damage”? Are you referring to the planar defects? If yes, expand on the formation on those and mention that they have not only been identified in lunar samples but also in solar wind irradiation experiments in oxides by Chaves, L.C., Thompson, M.S., Loeffler, M.J., Dukes, C.A., Szabo, P.S., Horgan, B.H.N. 2023. Evaluating the effects of space weathering on magnetite on airless planetary bodies. *Icarus* 402, 1-14. <https://doi.org/10.1016/j.icarus.2023.115634>

Line 229. What do you mean by “this damage”? are you referring to the planar defects or to the different microstructural features observed (npFe₀, vesicles, planar defects, etc)?

Line 229. What is “earliest appeared shallow”? consider rewording

Line 231. By “intensity” are you meaning an increase in the energy of the solar wind particles or are referring to the exposure time? Consider rewording

Line 232. Correct “more deeper”

Line 235. What are you referring to when you mentioned “nano-vesicles”? to the “tiny vesicles” or “planar defects”? They have not been defined previously in the manuscript.

Line 241. Correct the typo “vesilces”

Line 263. Are npFe₀ present on the crater or not at all? If I am not mistaken, there are some scattered dark circular spots that look like npFe₀

Line 341. Change to “degrees of space weathering”

Line 355. Correct the typo "micros"

Line 355. Were the lunar soils placed on top of a carbon substrate or on top of an SEM stub with a carbon substrate? Consider changing the word “stuck”

Line 367. What type of EDS detector?

Line 371. For simplicity, change “Electron energy loss spectroscopy (EELS) is a useful method for determining the nanoscale vesicle components and valence states of transitional elements in extraterrestrial samples, which is utilized in this study to characterize ...” to “Electron energy loss spectroscopy (EELS) was used to characterize ...” or similar

Line 384. Did you use any software to run the model? If yes, briefly mention the method/software used

Ms. Ref. No.: COMMSENV-24-0071-T

Title: Diverse space-weathered rims on ilmenite: constraining the exposure history of Chang'e-5 lunar soils

Changed title to "[Diverse space-weathered rims on ilmenite: Implications for the exposure history of Chang'e-5 lunar soils](#)".

Authors: Zhuang Guo, Mingwei Zhang, Bojun Jia, Yang Li, Wenzhe Fa, Yong Pang, Chenxi Zhu, Yuanyun Wen, Xiongyao Li, Jianzhong Liu, Ziyuan Ouyang

Communications Earth & Environment

Reviewer #1 (B. Cymes, Remarks to the Author):

Summary: This manuscript describes TEM, EELS, and EDS analysis of 11 ilmenite-bearing grains from 5 samples of Chang'e-5 lunar soils collected at depth intervals ranging from 0–72 cm. The authors show a diverse range of space weathered ilmenite rims and have developed an exposure scale based on the interpreted degree of space weathering in the samples. This scale and the samples were then applied to describe the exposure history of the Chang'e-5 landing site. The data presented are of high quality and provide a great range of ilmenite space weathering textures, which would be of moderate interest to the community as most studies focus on smaller grains, but the interpretation of the data and its connection to larger-scale phenomena would be stronger if complementary techniques were included.

[Reply: Thanks for your approval of our research and your highly professional comments. We have provided more details about the methodology to make the manuscript more reliable and to emphasize solid arguments based on convincing results. We hope that our response can address your concerns.](#)

General Comments:

1. Statistical limitations on data interpretation

The surface shoveled sample, CE5C0200, had a $<10\ \mu\text{m}$ ilmenite-bearing grain (CE5C0200 #1). This is much smaller than the other grains studied, which were $>100\ \mu\text{m}$. As the degree of space weathering is correlated to the grain size, it is not surprising that

CE5CO002 #1 had clear space weathering features.

My concern is on the statistics used in this study. For example, of the other two samples from CE5C0200, #2 and #3, one had a npFe₀-rich rim (the 2nd most space weathered classification devised here) and the other was not space weathered. For CE5C0200, this means for grains <10 μm, 100% (n=1) were highly space weathered and grains >100 μm, 50% (n=2) were moderately to highly space weathered. While it is appreciated that they analyzed more than one grain from each sample, there is not good agreement between them. The same can be said for CE5Z0806 and CE5Z0204 where 50% of grains (n=2 each) studied were space weathered.

The conclusions that they are using these samples to support, related to large scale mixing and exposure, would be strengthened if they were combined with other measurements, like FMR, reflectance spectroscopy, and bulk chemistry. The nanoscale TEM data alone limit the conclusions which can be drawn. Inclusion of discussion on solar flare tracks as indicators of exposure history is necessary as it has been used as an exposure metric for several decades. Moreover, it was not clear how the core-sample intervals were chosen, but they are ~20 cm, which is coarse. Nor was it clear why the >100 μm grain size chosen for study.

Reply: Thank you for your comments. As you said, grain size is an indicator of the maturity of the lunar soil, and finer grain sizes usually represent a more mature lunar soil. Here, Ilmenite (CE5C0200 #1) from the finest fraction of shoveled Chang'e-5 lunar soils (<10 μm) is selected as the most space-weathered ilmenite on the lunar surface for studying (Fig. 1a). The complex structural and chemical microscopic features exhibited by this grain are a summary of the response of ilmenite to intense solar wind irradiation.

Yes, what we observed is that the different grains in the same drilled samples show varying space-weathered characteristics. However, it is also not surprising for us because the lunar regolith has undergone numerous overturns and ejecting, and the differences in exposure between particles should be large as expected. In the discussion section, we address the possible reasons for the occurrence of diverse space weathering features in these grains, which are closely related to the evolution of lunar regolith in different depths (Lines 350 – 369). For example, ilmenite grains from most surface layers (CE5C0200 and CE5Z1002)

simultaneously include weakly (CE5C0200 #3), intermediate (CE5Z1002 #2), and highly weathered ilmenite grains (CE5Z1002 #1, CE5C0200 #1, and #2). We suggest that this should be attributed to the fact that the lunar soil at that depth has likely overturned more than 1,000 times since the formation of the Xu Guangqi crater and that the surface lunar soil has a complex origin, ranging from Xu Guangqi ejecta, bedrock clasts excavated from fresh impact craters (C2–C12), and possible trace amounts of distal ejecta from large impact craters. All of these contribute to the varied exposure history of Chang'e-5 shoveled soils.

Here, I would like to emphasize that although the exposure age of particles in a layer varies widely, the presence of extremely space-weathered grains tends to indicate that this layer of lunar soil has undergone a long exposure, and therefore we believe that the deeper CE5Z0204 lunar soil containing highly weathered ilmenite should have undergone a relatively long exposure history, and we believe this to be a solid argument in our manuscript. However, as you suggest, our manuscript investigates only a very small number of lunar soils, and the fact that no heavily weathered particles were observed does not mean that this layer is not exposed. Therefore, we rewrote the sentence as “It is worth noting that our study involves only a very small number of lunar soil grains, and the exposure history of the lunar soil layer at each depth is likely very heterogeneous, which is consistent with our observations. General observations of less weathered ilmenite grains in CE5Z0403 and CE5Z0806 can only provide evidence that regolith at depths of 26 cm and 45 cm at the Chang'e-5 landing site is highly likely to be exposed for a short period” in lines 373 – 377 of the revised manuscript.

Your suggestion is good, analysis techniques for bulk soils could provide key parameters about the formation and evolutionary history of lunar soils, and that's the most reliable way to relate to the large-scale features on the Moon. However, these methods are difficult to apply to drilled samples; on the one hand, the amount of drilled samples is very small (the drilled samples from all the depths of Chang'e-5 added up to just over 200 grams, while the shoveled samples were over 1,500 grams), and whole-rock chemistry or FMR are difficult to be obtained; on the other hand, the reflectance spectroscopy (OMAT) can provide critical information about the lunar soil maturity, but this is still targeted at the most

superficial layers of the lunar soil. Our starting point for this study is also to get more evolution information on the lunar soil in the vertical profile with a small amount of sample, which is crucial for our understanding of the formation and evolution of the lunar regolith. We are also well aware that this study cannot come to an accurate conclusion, but this is a pioneering attempt, and we have confirmed that the Chang'e-5 drilled samples collected an ancient layer of lunar soil that predates the formation of Xu Guangqi crater, and this can also be applied to the study of asteroid samples as well!

We added “The reflectance spectral results of the Chang'e-5 subsurface soil also show fresher spectral features compared to the surface soils (Wu et al., 2024), which supports the possibility that the intermediate layer of the Chang'e-5 landing site has only experienced weak exposure” in lines 377 – 381 of the revised manuscript.

We have not observed solar flare tracks in these ilmenite grains, which are more common in plagioclase and olivine minerals (Keller et al., 2021). Because we have only applied for samples from these layers from CNSA, we can only perform these studies. Nevertheless, we have obtained key information about the vertical profile of the lunar soil at the Chang'e-5 landing site, such as the thickness constraints of the Xu Guangqi ejecta material and the presence of ancient exposed lunar soil beneath the Xu Guangqi ejecta. Large particles were chosen because ilmenite is usually found in basaltic rock clastic and rarely in the form of single particles, and large particles are more suitable for FIB slicing.

Ambiguous TEM interpretations

A few described features are challenging to see in some images and could very well be thickness artifacts. For example, the vesicles in Fig. 3f and 3h look like contrast variations as there is no clear edge demarcation. The same goes for the disordered areas in Fig. 3f, 3g, and 3h, these could be thickness variations. In contrast, the vesicles in Fig 2a and 2d are very clear.

Certain features, particularly near the surface, are difficult to see because the samples were coated with gold instead of carbon. The high mobility of Au is concerning in general as it could be deposited on the foil sidewall, potentially causing some of the contrast variations. Coating with Au makes HAADF imaging difficult due to the high Z contrast – see Fig. 5.

Reply: Yes, the vesicles in the Figs. 3f and 3h are not clear, and we believe that these vesicles are smaller in size and therefore more difficult to see compared to the vesicles in Fig. 3. It's worth mentioning that these bubbles are very visible when viewed in real-time, and these vesicles are clearer to see in Figs. 3c – 3e. As we know that the contrast of bright-field images is affected by many factors, it is indeed necessary to identify vesicles by combining the HAADF images in Fig. 5 with the detectable helium within these vesicles. We have added an index of the corresponding images to the representation of the vesicles in the text. We don't believe that there are significant thickness differences in the nano-scaled regions within the FIB section, and these disordered areas were identified by the discontinuities in lattice fringes.

Thanks for your reminder. The reason we gold-plated it was to avoid overlooking the carbon-containing components that might be present in other lunar soils. In our experiments, the effect of Pt may be greater compared to Au. We try to avoid using one imaging mode to determine the microstructural features inside these rims, we usually combine the EDX maps, bright field, and dark field images to make a comprehensive judgment. Thank you again for your suggestions, we will try to avoid these factors as much as possible in our subsequent studies.

Specific Comments:

L41–43: Consider removing the sentence about asteroid reflectance spectra and focus on lunar data.

Reply: Thanks for the suggestion, but space weathering is important for the study of asteroids, especially since S-type asteroids have a similar space weathering model to the Moon since solar wind-dominated early alteration was initially detected in asteroid spectra, but can only be verified by studying lunar samples, which have a larger number of samples. Therefore, we consider the spectra of asteroids to be important indicators of solar wind irradiation processes, and we believe that this part of the introduction is necessary.

L53: remove “with both simpler and high symmetry crystal structures”.

Reply: We removed this sentence from the revised manuscript.

L54: Ref 11, Zinkle 2012 does not mention ilmenite vs. silicates.

Reply: We removed the herein Zinkle's reference from the revised manuscript.

L77: How does CE5C0200 #1 fall into the exposure classification scheme used in this study?
Is it being used as a sort of space weathering standard?

Reply: Thank you for your comment. The purpose of this grain introduction is to clarify the complex and multiple space weathering responses of ilmenite under solar wind irradiation, which is undoubtedly the most weathered degree and falls into the deeply damaged rim group. We added the “and the above-described CE5C0200 #1 grain fall into the deeply damaged rim group.” in lines 118 – 119 of the revised manuscript.

L122: The slight depletion of oxygen observed is not outside the margin of error of the EDS measurement, particularly for oxygen, which can be difficult to quantify accurately due to absorption effects. Could they report how thick the samples were? The slightly damaged rims would be more accurately characterized as undamaged, or not space weathered.

Reply: Thank you for your suggestion. As you mentioned, the EDS measurement is not going to get accurate O content. However, the HAADF image of this rim shows a little bright contrast, indicating a subtle difference in chemical composition from the substrate. In addition, the oxygen deficiency by preferential sputtering induced by solar wind irradiation has been confirmed by many previous studies (Christoffersen et al., 2011; Zhang et al., 2010; Zhu et al., 2014). Combined with the quantitative results, we believe that sputtering loss of oxygen should have occurred. We realize that the undamaged rim you suggested may be more accurate as there was no structural damage observed. We changed the “slightly damaged rim” to the “undamaged rim” in the whole manuscript.

We added “(Orim, 50.9 at% vs. Osubstance, 54.4 at%)” in line 143 of the revised manuscript.

L143: Provide measurement for “tiny” vesicles.

Reply: We provide the TEM – EDX maps, HRTEM, and EELS results of “tiny” vesicles in the Figs. 3f, 3c, and 5a.

L165: Fig. 2b insert says 2.03 Å, this text says 0.203 nm – the units for lattice spacings should be kept consistent throughout the text. Also, these have one too many digits after the decimal.

Reply: Thank you for your comment, we changed “0.203 nm” to “2.03 Å” in line 190 of the revised manuscript.

L180: How large are the “large” vesicles described and shown in 5b and 5c? None of the

ones shown in these samples seem to exceed ~10 nm in diameter, which is not really what we would typically call “large”. How do these large vesicles compare with the smaller/tiny/nano-vesicles described and the large vesicles (up to 40 nm) described in CE5C0200 #1? Clarify

Reply: Thank you for your comment, we are sorry for our confusing expression. As shown in Fig. 5. The 5b and 5c rims contained some larger vesicles (>5nm in diameter) than the “tiny” vesicles observed in the 5a, and helium was detected in these relatively large vesicles. We changed “large vesicles” to “vesicles (>5nm in diameter)” in line 195 of the revised manuscript.

L219–222: Revise this sentence for clarity.

Reply: We are sorry for our confusing expression. We changed “By combining previous in-situ ion irradiation simulation experiments and lunar soil observations, we clarify the specific modifications of different degrees of solar wind irradiation on lunar ilmenite.” to “By comparing previous in-situ ion irradiation experiments and analyses of lunar soils, we could attribute specific characteristics in these Chang’e-5 ilmenite grains to different degrees of solar wind irradiation on the lunar surface” in lines 254 – 257 of the revised manuscript.

L224–225: What orientation is being referred to here and how does its depth range coincide with the irradiation of solar wind on the lunar surface?

Reply: Thank you for your comment. What we want to express is that the structure damage within the one ilmenite rim is approximately parallel, as shown in Fig. 3i. Its depth is consistent with the ion-irradiation simulation experiments under lunar conditions. We changed “The structural damage observed in the Chang’e-5 ilmenite rims show a certain orientation and its depth range (<200 nm) coincides exactly with energetic particle irradiation of solar wind on the lunar surface” to “The structural damage is concentrated on the surface of Chang’e-5 ilmenite, and its depth range (100–200 nm) coincides exactly with the results of energetic particle irradiation under simulated lunar conditions” in lines 260 – 262 of the revised manuscript.

L227: Clarify “at this structural damage” Do they mean associated with a specific type of structural damage?

Reply: Thank you for your comment. we changed “at this structural damage in the space-

weathered ilmenite rims” to “in the planar defects of Apollo ilmenite space-weathered rims, thus the solar wind ion implantation is the main cause of such planar defects” in lines 263 – 265 of the revised manuscript.

L229–232: The sentence needs to be revised for clarity.

Reply: We are sorry for our confusing expression. We changed “The earliest appeared shallow localized nanoscale structural disorder shown as mottled contrast in bright-field images, and with increasing solar wind irradiation intensity, the disorder progresses amorphous and deeper.” to “In Chang’e-5 ilmenite grains, the earliest structural damage appears as localized nano-scale structural disorder in the most superficial layer, and with the accumulation of solar wind irradiation, the superficial layer is gradually amorphous, and the structural disorders and planar defects appear at a deeper depth” in lines 265 – 268 of the revised manuscript.

L232–234: What is this interpretation based on? Is this a hypothesis?

Reply: Yes, we deleted this hypothesis and rewrote this sentence as “and the structural disorders and planar defects begin to appear at a deeper depth” in line 268 of the revised manuscript.

L241–244: Revise for clarity. Also, it would be helpful to define what they’re referring to when they say, “large vesicles” “smaller bubbles” and “nanovesicles”.

Reply: Thank you for your comment. We have addressed this question in our previous Reply.

L268: Correct to <400 °C? Also, 400 °C is not what would typically be considered “low temperature”.

Reply: Thank you for your comment. What we are trying to express is the need for lower temperatures to avoid the loss of solar wind gases. However, the <400 °C and the low temperatures you mentioned do seem to contradict each other. We changed “under low-temperature conditions (<400°C to avoid release)” to “at a temperature condition of <400°C (to avoid the solar wind gases release)” in lines 281 – 282 of the revised manuscript.

L270–272: Could they elaborate on this more? Pull out what these sources referenced are saying on this and describe their results in context? Also, what do they mean by decisive? Are they claiming that diurnal temperatures drive npFe0 and vesicle formation? Because

that is not supported.

Reply: Thank you for your comment, we changed it to “Previous studies have proposed that a long-term thermal process provided by diurnal solar heating (100–120°C) may promote the formation of npFe⁰ particle and large vesicle within the 40 nm thickness range on the ilmenite surface by radiation-enhanced diffusion” in lines 305-308 of the revised manuscript.

L348–349: I do not think this study should be characterized as pioneering, given that microscopists have been conducting such studies for several decades.

Reply: Thank you for your suggestion. As you said, microscopy studies have been developed for several decades, and there are many nano – scaled minerals as well as interesting phenomena that have been revealed. However, our pioneering means the use of microscopic analysis to indicate the exposure history of lunar soil. Macroscale lunar soil evolution is largely based on remote sensing and modeling, with little support from sample data. Although we can understand the exposure history of the lunar soil through the measurement of noble gases or iron content, which is difficult to achieve with a small number of drilled lunar samples. Here, we use the characterizations of single-grain weathered features to enable the determination of relative exposure ages on lunar regolith profiles, which we believe is pioneering as it does not require a large number of samples. This does provide additional useful information on the exposure history of lunar soil, despite the high heterogeneity of exposure between particles.

We changed “pioneering attempt to systematically understand the lunar regolith evolution using microscopic analysis” to “Many indices have been proposed in the last decades to estimate the exposure time of lunar soils, such as agglutinate content, Is/FeO parameter, noble gas contents, and the solar flare track density, and our study is an alternative approach to identifying soil exposures for cases where only a small amount of samples are returned, such as deep lunar regolith and asteroids” in lines 392 – 396 of the revised manuscript.

L357: Could they describe how the 10 µm grain was used in this study? This sentence is not clear.

Reply: Thank you for your comment. CE5C0200 #1 grain is about 10 µm in diameter, which

was selected from this sample and represented the most space-weathered ilmenite, as shown in Fig. 2.

We changed “Also, a shoveled fine ilmenite-bearing grain (<10 μm) adhered to the silicon wafer from CE5C0200 shoveled lunar soil.” to “The finest-grained shoveled Chang’e-5 soil were dispersed onto a gold-plated silicon wafer using $\geq 99.8\%$ high-purity ethanol, then the sample was covered with a gold film for SEM observations, and the CE5C0200 #1 grain ($\sim 10 \mu\text{m}$) was selected in this sample for studying the most space-weathered ilmenite” in lines 404 – 408 of the revised manuscript.

L365: Provide a citation to dual-beam method or a brief description of the FIB preparation.

Reply: The method of double-beam preparation of FIB samples has been very well established and described in many articles, we cite the Burgess and Stroud et al., (2018) paper here. We added “and the FIB method is a common technique for the in-situ preparation of transmission electron microscope (TEM) foils (Burgess et al., 2018)” in lines 416 – 417 of the revised manuscript.

L366: How were the EDS data collected, processed, and quantified? This should be included.

Reply: Thank you for your comment. EDS data was collected using an EDS detector on the STEM instrument, and the quantification of TEM – EDS data was calculated by an interactive TEM quantitative method in Bruker Esprit software, equipped with the Talos F200X instrument.

We changed it to “Chemical analysis of FIB sections, including X-ray elemental mapping and EDS spectral acquisition, was performed using the SDD-EDS detector on the Talos F200X instrument. The quantification of TEM-EDS data was calculated by an interactive TEM quantitative method in Bruker Esprit software, and data in the supplementary Table 1 are based on area selections from the quantitative TEM-EDX maps” in lines 424 – 429 of the revised manuscript.

L367: Provide more information on the Talos - imaging mode, accelerating voltage, probe diameter, etc.

Reply: Thank you for your comment. we added “FIB sections were characterized with a FEI Talos F200X field–emission scanning transmission electron microscope (FE–STEM) operating at 200 kV, at the Suzhou Institute of Nano–tech and Nano–bionics, CAS. STEM images were collected in high–angle annular dark field (HAADF) mode, with a 0.16 nm probe.” In lines 423 – 424 of the revised manuscript.

L376: What was the probe diameter on the Hitachi?

Reply: Thank you for your comment. we added “with a 0.078 nm probe of STEM mode,” in line 437 of the revised manuscript.

379: Need to provide more information on how EELS data were collected and processed. Dispersion, exposure, type of collection (spot analyses, line scans, spectrum images?), also how was the background subtracted?

L382: They should also include Fe standards.

Reply: Thank you for your suggestion. We changed it to “The energy dispersion for EELS measurements was 0.1 eV/channel with an energy resolution was ~0.5 eV, measured from the full width at half the maximum height of the zero-loss peak. EELS integration times were 8.8 s for Fe point analyses, 26.4 s for Fe analyses at each point in the line-scan, 1 s for He point analyses, 10 s for line-scans of Ti at each point, and 17.6 s for Ti analyses at each point in the line-scan. The synthetic Ti_2O_3 , rutile (TiO_2), fayalite, terrestrial hematite, and FeNi metal from meteorite (GRV 051874) were prepared as Ti^{3+} , Ti^{4+} , Fe^{2+} , Fe^{3+} , and Fe^0 reference standards for the comparing valence transition in the space-weathered ilmenite rims (Supplementary Fig. 6). Peak alignment to compensate for energy drift during EELS SI acquisition and the background subtracted by power law were carried out using Gatan Digital Micrograph software” in lines 439 – 450 of the revised manuscript.

We also present the EELS spectra for the Fe standards in the Supplementary Fig. 6.

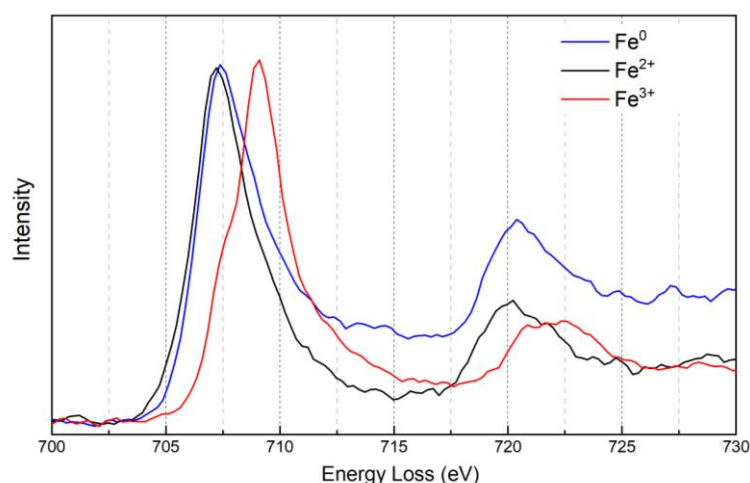


Fig. 1: Missing description of panel a.

Reply: Thank you for your careful review. We added “a. The secondary electron (SE) image of the ilmenite-bearing clast from the finest shoveled Chang'e-5 lunar soil” in line 122 – 123 of the revised manuscript.

Fig 4e: The element map in (e) looks binned when compared to the other maps. Could they be processed in the same way for more accurate comparison?

Reply: We are sorry that we can't go any further process with this. The chemical heterogeneity of most surfaces is of interest, and the chemical characteristics of most surfaces are already clearly distinguishable.

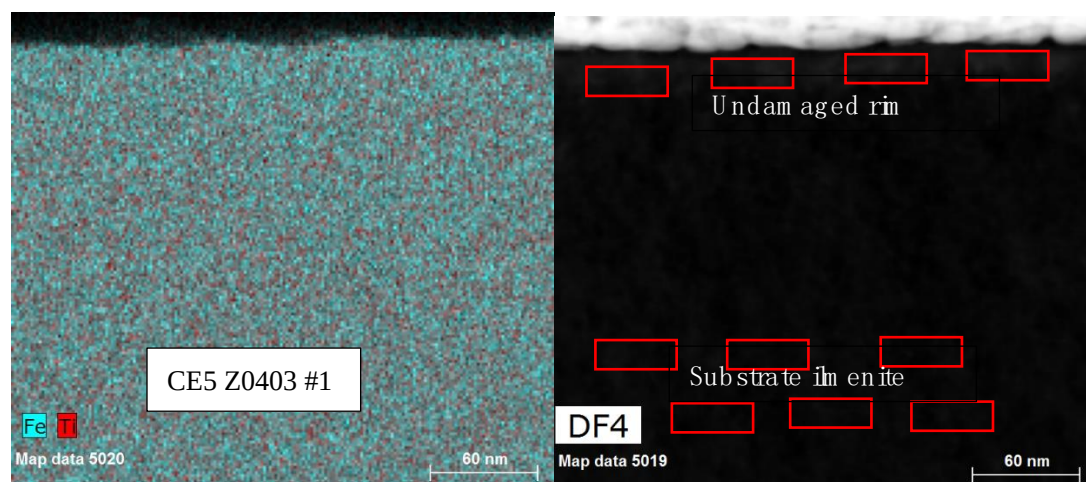
Fig 5c: The He edge is not clearly above the noise in this spectrum. Could they show background-subtracted spectra?

Reply: We have shown all raw data in the provided repository link. The He spectra data presented in the manuscript are raw and have not been subjected to background-subtracted. The quality of the EELS spectra is controlled by many factors such as the thickness of the sample and the elemental content. The detectable He signal is very sharp compared to the spectra from the He – free substrate mineral.

Supplementary Table 1: Include error in at% measurement. Are these data based on area selections from the maps shown in Fig. 4? Can they show these selections?

Reply: Thank you for your comment. You are right, all these data are based on area selections from the maps shown in Fig. 4. Quantitative data is obtained by selecting the

rectangular frame as shown below. We also give the error for these data in Supplementary Table 1.



We added “The quantification of TEM-EDS data was calculated by an interactive TEM quantitative method in Bruker Esprit software, and data in the supplementary Table 1 are based on area selections from the quantitative TEM-EDX maps” in lines 426-429 of the revised manuscript.

Reviewer #2 (Remarks to the Author):

This manuscript focuses on the chemical and microstructural characterization of ilmenite grains returned by the Chang’e-5 mission through transmission electron microscopy, energy dispersive X-ray spectroscopy, and electron energy loss spectroscopy. Ilmenite grains exhibit different degrees of space weathering and their alteration has been attributed to the influence of the solar wind on the lunar surface. In addition, the different degrees of alteration were used to reconstruct the stratigraphic profiles from the sampling region to elucidate regolith overturn processes.

Additionally, the results shown in the manuscript are novel as no studies report different degrees of space weathering on a singular mineral phase from returned samples. The results from this manuscript will contribute to a better understanding of the space weathering processes on the Moon.

Besides the valuable scientific content of the manuscript, I recommend looking for editing services to improve the readability of the manuscript. Please refer to the attached document “comments to authors” where you will find my questions and suggestions about

the manuscript.

Reply: Thanks for your approval of our research and your detailed comments. We have thoroughly revised the manuscript to make it more readable. We hope our response addresses your questions.

Specific Comments:

Line 21. Replace “source” by “mechanism”.

Line 22. Add “the” before “lunar regolith”.

Reply: We revised it in the revised manuscript.

Line 23. Can you expand on the pieces of evidence that suggest that ilmenite is highly resistant to solar wind irradiation?

Reply: As the abstract has a word count requirement, we have expanded on this point in the introduction section.

We added “Ilmenite is more resistant to amorphization by ion irradiation than silicates owing to its close-packed crystal structure, which has been demonstrated in previous studies” in lines 58 – 60 of the revised manuscript.

Line 32. Can you explain why only short-term exposure? The deeply damage rims could result from long-term exposure.

Reply: Thank you for your comment. we have detailed explained this in the discussion section. Given the fast saturation rate of the solar wind (less than a few thousand years), we therefore conclude that the varying degrees of space-weathered microstructures by of ilmenite by solar wind should be useful only within the short – term exposure (a few thousand years).

We realize that this expression may confuse the reader, so we changed it to “the relative exposure ages of regolith on airless bodies” in line 35 – 36 of the revised manuscript.

Line 38. Add “lunar” before “material”.

Reply: We revised it in the revised manuscript.

Line 40. Consider changing “which should strongly alter” to “modifying its reflectance

spectrum” or to a similar sentence.

Reply: Thank you for your comment. we changed it to “thus modifying their reflectance spectrum” in line 43 of the revised manuscript.

Line 41. Add “studies or analysis” after “spectra”.

Line 56. Delete “the” before “space weathered”.

Line 56. Change “rim” for “rims”.

Line 56. Replace “exhibits” for “exhibit”.

Reply: We revised it in the revised manuscript.

Line 56–57. You mentioned “ilmenite grains exhibit more complex chemical responses to space weathering than lunar silicate grains”. Add a sentence where you specify what are the structural and chemical differences resulting from space weathering between these two mineral groups.

Reply: Thank you for your comment. we rewrite it as “Detailed microscopic analyses, including transmission electron microscopy (TEM) and atom probe tomography, suggest that space-weathered rims on lunar ilmenite grains exhibit complex chemical and structural responses to space weathering, such as the helium-bearing vesicles, significant structural defects, and variations of iron and titanium valence states, as compared to the amorphous rims on lunar silicates” in line 60 – 65 of the revised manuscript.

Line 76. In the manuscript, you never mentioned figures 1b,c,d,e, or f. You could add an introductory sentence where you state that from all the analyzed samples (Fig. 1a,b,c,d,e, and f) CE5C0200 is the sample that is the most space-weathered grain.

Reply: Thank you for your suggestion. We state it as “undamaged rims (CE5C0200 #3, CE5Z0806 #1, CE5Z0403 #1 and #2, CE5Z0204 #2) (Fig. 1e and Supplementary Fig. 2), densely damaged rims (CE5Z0806 #2) (Fig. 1d), vesicular rims (CE5Z1002 #2) (Fig. 1c), npFe₀-rich rims (CE5C0200 #2, CE5Z1002 #1) (Fig. 1b and Supplementary Fig. 3), and deeply damaged rims (CE5Z0204 #1) (Fig. 1f)” in lines 113 – 117 of the revised manuscript.

Line 76–77. Add a sentence expanding on why you think CE5C0200 is the most space-weathered grain.

Reply: Thank you for your comment. Grain size is an indicator of the maturity of the lunar soil, and finer grain sizes usually represent a more mature lunar soil. After careful consideration, we feel that this statement is uncritical. We rewritten this sentence as “Considering that the grain size of lunar soil is correlated with lunar soil maturity (Noble et al., 2001), ilmenite (CE5C0200 #1) selected from the finest fraction of shoveled Chang'e-5 lunar soils (<10 μm) could represent the most space-weathered ilmenite collected by the Chang'e-5 mission” in lines 86 – 89 of the revised manuscript.

Line 77. Replace “on the lunar surface” by “collected by the Chang'e-5 mission”.

Line 79. Add “(Fig. 2)” after “FIB section”.

Reply: We revised it in the revised manuscript.

Line 80. Are all npFeO particles elongated? I see npFeO with circular shapes in Fig 1a and Fig 1b. Describe the different shapes present. In addition, where you mentioned “60 nm in length”, is that an average or the maximum length? Again, the npFeO in Fig 1a and b are significantly smaller than that.

Reply: Thank you for your comment. Yes, there are spherical iron particles within the rim. we changed “elongate npFeO particles (~60 nm in length)” to “elongate and spherical iron particles” and added “The elongate-shaped iron particles are up to 60 nm in length, which is much larger than the observed spherical particles” in lines 92 and 95 – 96 of the revised manuscript.

Line 81. Did you perform sample tilting (zone axis) or FFT to corroborate that the rim was completely amorphous? Did you do the same for all the samples?

Reply: Actually, the procedure for all our studied ilmenite is to first tilt the host ilmenite to the low index zone – axis and obtain the selected electron diffraction patterns, followed by the observation of the surface structures. Therefore, any change in surface structure occurs on the basis of a high symmetry orientation of the crystal.

We realized that “completely amorphous” may not be accurate, as there are still a few deformed or short-range ordered crystalline planes present in Fig. 2.

We changed “completely amorphous” to “almost fully amorphous” in line 93 of the revised manuscript.

Line 83. Replace “(Fig. 2)” for “(Fig. 2a, b,c,d)”.

Reply: We revised it in the revised manuscript.

Line 84. For simplicity, please consider changing “EELS spectra show that the presence of helium is readily recognized (diagnostic peak at about 22 eV) in large vesicles” by “EELS spectra show the presence of helium (diagnostic peak at about 22 eV) in large vesicles” or similar.

Reply: Thank you for your comment. we changed it to “EELS spectra show the presence of helium (diagnostic peak at about 22 eV) in large vesicles” in lines 98 – 99 of the revised manuscript.

Line 88. Specify what is the energy of the L2 and L3 peaks.

Reply: We labeled the L₂ and L₃ edges of Ti in Fig. 2f.

Line 90. Can you explain which characteristics are you referring to?

Reply: Thank you for your comment. we changed it to “The thick structural damaged rim, the production of He-bearing vesicles, and the precipitation of large npFe⁰ particles suggest that this lunar soil grain had experienced extreme solar wind irradiation.” in lines 104 – 106 of the revised manuscript.

Line 100. Can you add the Ti EDX map to Fig 1a as you did for the other grains? Or is there a specific reason why you did not include it?

Reply: Thank you for your comment. Fig. 1a and Fig. 1d are different locations from the same grain (CE5C0200 #1 ilmenite). Fig. 1a is a TEM bright-field image, which is used to show features of texture and crystal structure in the nanoscale.

We chose the Fig. 1d location for the EDX map mainly because the iron particles in Fig. 1d are more visible, suggesting a more significant chemical alteration. We believe that Fig. 1a should have a similar Ti EDX map to the present Fig. 1d because it is from the same grain.

Line 106. Change the colors of the EDX maps on Fig 2. to make it consistent with the EDX maps on Fig.4 and the supplementary material.

Reply: Thank you for your comment. The EDX maps in Fig. 2 are single-element, to illustrate specific single-chemical element distribution features in the most space-weathered rims, and such colors were chosen to clearly exhibit the Fe- and Ti-rich regions. However, the EDX maps in Fig. 4 are a combination of multi-element maps, with the main focus on a more attractive color. We apologize that this color may be misrepresented in a later modification and may make the contrast less noticeable, so we intend to retain the original color scheme.

Line 107. Remove “most space-weathered” from the figure caption.

Reply: We revised it in the revised manuscript.

Line 118. Expand on how the rims are structurally consistent with the underlying ilmenite.

Reply: Thank you for your comment. We changed it to “The crystallographic orientation of the undamaged rim is consistent with the substrate ilmenite” in lines 139 – 140 of the revised manuscript.

Line 122. Do you mean Fig 4a instead of Fig 3a? also, add in the text the difference in at% of the O content between the rim and the underlying grain.

Reply: Thank you for your careful review. We changed it to “Fig. 4a” and added “(O_{rim} , 50.9 at% vs. $O_{\text{substrate}}$, 54.4 at%)” in line 143 of the revised manuscript.

Line 126. By “this group” do you mean the slightly damage rim category? Reword. In addition, consider replacing “weakest” by a more appropriate word (e.g., lowest).

Reply: Thank you for your comment, which was also raised by another reviewer. We realize that the undamaged rim may be more accurate as there was no structural damage observed. We changed the “slightly damaged rim” to the “undamaged rim” in the whole revised manuscript.

Line 133. Mention the value of O depletion in at%.

Line 133. Consider rewording “the degree of oxygen loss tended to decrease with increasing depth” it is confusing.

Reply: Thank you for your comment. We changed it to “the content of atomic oxygen tended to decrease with increasing distance from the surface” in lines 154 – 155 of the revised manuscript.

Line 143. I recommend using “small vesicles” rather than “tiny vesicles”.

Line 144. Delete “TEM”.

Reply: We revised it in the revised manuscript.

Line 143. What are the sizes and shapes of the vesicles and npFe⁰?

Reply: Thank you for your comment. we changed it to “spherical nano-vesicles (less than 5 nm in diameter) and a few nanophase iron particles (npFe⁰) (5–10 nm in diameter)” in lines 165 – 166 of the revised manuscript.

Line 150. What is the difference between “bubbles” and “vesicles”?

Reply: we changed it to “nano-vesicles” in the whole revised manuscript.

Line 159. Delete “shape”.

Line 162 –163. Add the values (at%) of the Ti enrichment and O depletion in the text.

Reply: We revised it. We changed it to “High titanium content (Ti_{outer layer}: 24.3 at% vs. Ti_{substrate ilmenite}: 22.5 at%) and oxygen deficiency (O_{outer layer}: 53.6 at% vs. O_{substrate ilmenite}: 56.8 at%)” in lines 187 – 188 of the revised manuscript.

Line 164. The sentence “The ubiquitous npFe⁰ particles in the outer layer are enriched in Fe” is redundant, consider rewording.

Reply: Thank you for your comment. we changed it to “The ubiquitous npFe⁰ particles in the outer layer are iron-rich in the EDX map” in line 190 of the revised manuscript.

Line 168. Remove “located beneath the outer layer” it is redundant.

Reply: We deleted it from the revised manuscript.

Line 187. Consider changing the word “slender” for “thin” or similar.

Reply: Thank you for your comment, we changed it to “parallel thin” in line 217 of the revised manuscript.

Line 189. Do you mean “identified” instead of “induced”?

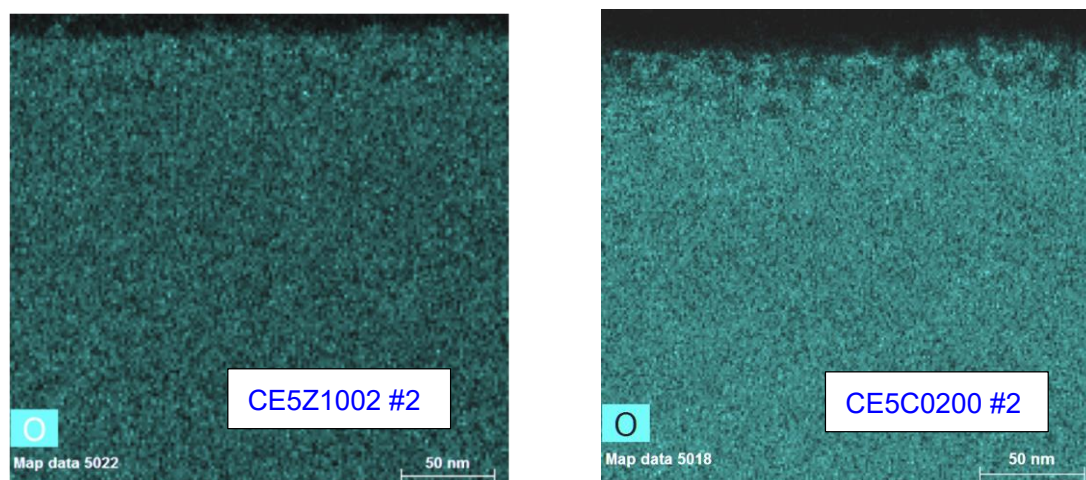
Reply: I’m sorry for our carelessness, we revised it in the revised manuscript.

Line 200. Include the O maps in the EDX layered image. Do you have any reason for including Si maps? Consider adding labels to the EDS maps and EELS spectra, maybe like a2, a3 where the BF image is a1. Change the numbering in the manuscript accordingly.

Reply: Thank you for your comment. Since oxygen changes continuously from the surface to the interior of ilmenite space-weathered rims, the contrast of the O maps is not very obvious (e.g., O maps of CE5Z1002 #2 and CE5C0200 #2 as follows).

As we all know, the surface of lunar soil particles is commonly distributed with vapor deposition layers due to frequent impacts, and an important criterion for the distribution of vapor deposition layers is the presence of exogenous elements. The reason for including Si maps here is to exclude vapor deposition of exogenous elements on the surfaces and to confirm that all the studied rim features are modifications of the host ilmenites. We added “The included Si element in the EDX maps is to distinguish the presence of exogenous vapor deposited material on the ilmenite surfaces.” in lines 235 – 237 of the revised manuscript.

We changed the numbering in the Fig. 4 as requested.



Line 200. What are the arrows showing the EELS spectra? In some EELS spectra you include Ti³⁺ and Ti⁴⁺ labels and in others no, be consistent.

Reply: Thank you for your comment. We add “The arrows in the EELS spectra (a3–e3) represent the splitting of the Ti $L_{2,3}$ peaks, and the decrease in splitting correlates with the presence of Ti³⁺” in lines 237 – 239 of the revised manuscript. We include the Ti⁴⁺ label in Figs. a3 – e3.

Line 216. Replace “Given the increasing depth of solar wind action and progressive complexity of microstructure features on ilmenite surfaces, we can classify space-weathered ilmenite, from weakly to strongly weathered rims ... ” with “Based on the thickness and the microstructural complexity of the rims present in the space weathered ilmenite surfaces, we can classify them from the least to the most altered as ...” or similar.

Reply: Thank you for your suggestion, we revised it in the revised manuscript.

Line 219. I think you can change the sentence “By combining previous in-situ ion irradiation simulation experiments and lunar soil observations, we clarifies the specific modifications of different degrees of solar wind irradiation on lunar ilmenite” to “By comparing previous in-situ ion irradiation experiments and analyzes of lunar soils, we could attribute specific characteristics to different degrees of solar wind irradiation on lunar ilmenite” or something similar.

Reply: Thank you for your comment. We changed it to “By comparing previous in-situ ion irradiation experiments and analyses of lunar soils, we could attribute specific characteristics in these Chang’e-5 ilmenite grains to different degrees of solar wind irradiation on the lunar surface” in lines 254 – 257 of the revised manuscript.

Line 223. Expand on what are you referring to as “crystal damage”.

Reply: we expanded it to “Structural damage (e.g., amorphization and crystal defects) to materials caused by solar wind irradiation” in line 258 of the revised manuscript.

Line 225. What do you mean by “certain orientation”?

Reply: Thank you for your comment. What we want to express is that the structure damage within the one ilmenite rim is approximately parallel, as shown in Fig. 3i. Its depth is consistent with the ion-irradiation simulation experiments under lunar conditions. We changed “The structural damage is concentrated on the surface of Chang’e-5 ilmenite, and its depth range (100–200 nm) coincides exactly with the results of energetic particle irradiation under simulated lunar conditions” in lines 260–262 of the revised manuscript.

Line 226. Add a reference about the penetration depth of solar wind particles on lunar surface materials.

Reply: We added references about the penetration depth (Keller et al., 2021; Christoffersen et al., 2011) in line 262 of the revised manuscript

Line 227. What are you referring to when you mentioned “at this structural damage”? Are you referring to the planar defects? If yes, expand on the formation on those and mention that they have not only been identified in lunar samples but also in solar wind irradiation experiments in oxides by Chaves, L.C., Thompson, M.S., Loeffler, M.J., Dukes, C.A., Szabo, P.S., Horgan, B,H,N. 2023. Evaluating the effects of space weathering on magnetite on airless planetary bodies. Icarus. 402, 1–14. <https://doi.org/10.1016/j.icarus.2023.115634>.

Reply: Yes, it is referring to the planar defects, and we revised it. The elongated defects

mentioned by Chave et al., (2023) are not the same as the planar defects we are referring to; planar defects refer to relatively deep (~100 nm), parallel thin defects (Fig. 3i).

We cited Chave's paper in line 260 of the revised manuscript, which is more appropriate.

Line 229. What do you mean by "this damage"? are you referring to the planar defects or to the different microstructural features observed (npFe₀, vesicles, planar defects, etc)?

Reply: Thank you for your comment. We revised it to "thus the solar wind ion implantation is the main cause of such planar defects" in line 264 of the revised manuscript.

Line 229. What is "earliest appeared shallow"? consider rewording.

Reply: Thank you for your comment, which was also raised by another reviewer. We changed it to "the earliest structural damage appears as localized nano-scale structural disorder" in lines 265 – 266 of the revised manuscript.

Line 231. By "intensity" are you meaning an increase in the energy of the solar wind particles or are referring to the exposure time? Consider rewording.

Reply: we reworded it to "accumulation" in line 267 of the revised manuscript.

Line 232. Correct "more deeper".

Reply: We revised it in the revised manuscript.

Line 235. What are you referring to when you mentioned "nano-vesicles"? to the "tiny vesicles" or "planar defects"? They have not been defined previously in the manuscript.

Reply: We have revised all "tiny vesicles" to "nano-vesicles" in the revised manuscript.

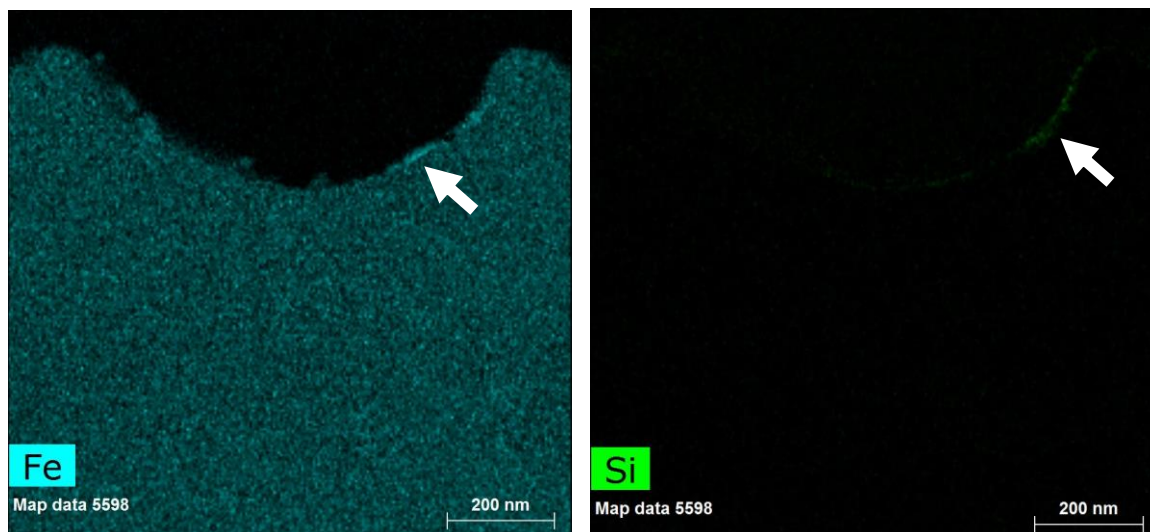
Line 241. Correct the typo "vesilces".

Reply: We revised it in the revised manuscript.

Line 263. Are npFe₀ present on the crater or not at all? If I am not mistaken, there are some scattered dark circular spots that look like npFe⁰.

Reply: Yes, you are right. There are a few npFe^0 particles in the top layer of the limited region of the crater. Our further investigations suggest that these npFe^0 particles are embedded in exogenously splashed silica-containing glass, rather than being formed directly by ilmenite during impact.

we added the “native” before “ npFe^0 particles” in line 298 of the revised manuscript.



Line 341. Change to “degrees of space weathering”.

Line 355. Correct the typo "micros".

Reply: we revised it in the revised manuscript.

Line 355. Were the lunar soils placed on top of a carbon substrate or on top of an SEM stub with a carbon substrate? Consider changing the word “stuck”.

Reply: Selection of lunar soils larger than 100 microns to be fixed on the top of a scanning electron microscope (SEM) stub with a carbon substrate” in lines 402 – 403 of the revised manuscript.

Line 367. What type of EDS detector?

Reply: We revised it to “SDD – EDS” in line 426 of the revised manuscript.

Line 371. For simplicity, change “Electron energy loss spectroscopy (EELS) is a useful method for determining the nanoscale vesicle components and valence states of

transitional elements in extraterrestrial samples, which is utilized in this study to characterize ...” to “Electron energy loss spectroscopy (EELS) was used to characterize ...” or similar.

Reply: Thank you for your comment. we revised it to “Electron energy loss spectroscopy (EELS) was used to characterize the elemental valence states of iron and titanium as well as the helium within vesicles in the Chang’e-5 ilmenite rims” in line 432 of the revised manuscript.

Line 384. Did you use any software to run the model? If yes, briefly mention the method/software used.

Reply: Yes, we used Python to run these models. We added “and these models were run by Python” in line 452 of the revised manuscript.

Reviewer #3 (K. Burgess, Remarks to the Author):

This manuscript presents TEM data of lunar ilmenite grains showing varying degrees of space weathering by solar wind irradiation. The imaging combined with EDS and EELS data provide evidence of how ilmenite rims form, from barely altered surfaces to intensely weathered, vesicle– and npFeO–rich rim more than 100 nm thick. The authors relate these differences in weathering to models of Chang’e-5 regolith emplacement and timing. The data presented support the overall conclusions and are an important contribution to our understanding of this understudied but important lunar phase. The results fit well within current understanding of solar wind irradiation processes and ilmenite rim structure, and clarify how intermediate space weathering stages can be recognized. The most novel part of the research is the attempt to directly link depth within the regolith (core) for each sample to ejecta from specific local craters and their exposure and timing.

Reply: Thanks for your approval of our research and your highly professional comments. We hope that our responses can address your questions.

However, several key points were not addressed by the paper. Previous work on ilmenite, which is cited, has shown clear importance of crystal orientation for the depth of

solar wind irradiation rims (Burgess & Stroud, 2018). This aspect of the grains here is not discussed, even as the difference in rim thickness is used to assume different levels of exposure. In the “NpFe⁰-rich rim”, the most heavily altered outer rim is in fact thicker than the outer rim of the “Deeply Damaged rim”, as Fig. 4 also demonstrates, even as planar defects are deeper in the latter. This does not alter the overall conclusions, but the less straightforward aspect of this variation as relates to exposure time needs to be addressed.

Reply: Thank you for your constructive comment. Yes, previous simulation experiments and analyses of lunar samples have shown that crystallographic orientation has a significant effect on the ion irradiation features of minerals (Burgess et al., 2018; Li et al., 2013), especially showing strong crystallographic controls of amorphous and planar defects processing as well as non-spherical npFe⁰ in the weathered rims.

We were already considering the effect of the crystallographic orientation for the solar wind irradiation features on ilmenite at the very beginning of the study. However, the irregular shape of the lunar soils and the small size of the ilmenite particles make it difficult to achieve the preparation of FIB slices with a specific crystallographic orientation. Therefore, we had to adopt an FIB slicing strategy to crosscut the long-axis orientation of plate-shaped ilmenite grains (Fig. 1 and supplementary Fig. 2), to make all the FIB slices come from the direction along [001] as much as possible. In Fig. 1d-1f, we show the lattice fringes of the studied ilmenite grains. The ilmenite of the vesicular rim (Fig. 3f) shows a similar crystallographic orientation to the ilmenite of the deeply damaged rim ($[\overline{4}2\overline{1}]$ zone axis) (Fig. 3h), while the ilmenite of npFe⁰-rich rims show a $[4\overline{4}1]$ zone axis (Fig. 3g). Thus, in this aspect, we have tried to avoid differences in the solar wind irradiation characteristics of ilmenite induced by the crystallographic orientation. We hold that the orientation of solar wind irradiation on these naturally returned soils should be constantly changing during the continuous overturn of the lunar regolith, and the irradiation modification characteristic that the particles ultimately exhibit is a cumulative one of the irradiation by the solar wind during its exposure. All we can do is look at them in roughly the same crystallographic orientation. On the other hand, our categorization of ilmenite rims is not only based on the degree of structural damage, because the degree of structural damage may vary very much in different crystallographic orientations. As your paper proposed, the crystal orientation does

not seem to play a role in some of the micro-features of ilmenite (e.g., vesicles), especially the chemical variation. Here, we classify the rim groups not only by reference to the structural damage features but also features such as the degree of valence change of iron and titanium elements and the size of the vesicles. We have classified the weakly, intermediate, and highly weathered ilmenite after considering all the features together.

As you said, the “npFe⁰-rich rim” is, in fact, thicker than the outer rim of the “Deeply Damaged rim”, we also hold that this should be attributed to the differences in crystallographic orientation. We categorized them in the discussion section of our manuscript as highly weathered type (Line 368 of the revised manuscript), so that does not alter the overall conclusions.

We added “We extracted FIB slices that cut across the long-axis orientation of plate-like ilmenite grains (Fig. 1 and Supplementary Fig. 2), trying to have all FIB slices from the same crystallographic orientation of ilmenite” in lines 417 – 419 of the revised manuscript.

We added “The ilmenite with vesicular rim (f) show a similar crystallographic orientation with the ilmenite with deeply damaged rim ($[4\bar{2}1]$ zone axis) (h), while the ilmenite with npFe⁰-rich rims shows a $[4\bar{4}1]$ zone axis (g)” in lines 228 – 230 of the revised manuscript.

We added “Considering that crystallographic orientation has a significant effect on the ion irradiation characteristics of minerals, we have classified these rims not only concerning structural damage features but also to the degree of valence changes of the elemental Fe and Ti, as well as the size of the vesicles and npFe⁰ particles” in lines 108 – 112 of the revised manuscript.

We added “The outer edges of the deeply damaged rim are thinner compared to the npFe⁰-rich rims, which should be attributed to the difference in crystallographic orientation (Figs. 3g and 3h)” in lines 205 – 207 of the revised manuscript.

Additionally, the loss of O and increase of Ti seen in EDS data are used as evidence of different rim development stages, but the Methods section and supplementary table do not provide details of errors, software used, or quantification algorithms. Interpretation of both EDS and EELS data requires sample thickness information, which is also not noted. This could strongly affect both the ability to detect He and interpretation of changes in Fe valence, eg in Fig. S5c, where npFe⁰ apparently does not contribute to the signal (Ln 148–

150) but could be a small portion of the overall thickness and thus its lack is not meaningful.

Reply: Thank you for your comments, these issues have been raised by other reviewers and we have added more details of the test conditions in the methods section and errors in the supplementary table 1.

Yes, your suggestion is right, we added the information about the thickness of the FIB sections, which is crucial for all results in the STEM mode. There may indeed be an argument about the presence or absence of the Fe⁰ you mention only based on the ambiguous EELS data, as previous studies have detected signals of Fe⁰ on the surface of ion-irradiated material using the XPS method (Loeffler 2009).

We added the “and all these FIB slices were thinned to ~100 nm by Ga⁺ ion beam” in line 420 of the revised manuscript.

We added the “indicative of the extremely low content of Fe⁰ in the matrix” in lines 173-174 of the revised manuscript.

It is unclear if EDS composition information or the EELS spectra are from spots or summed from maps and, if spots, what the spot sizes are. Small circles are used in Fig. 5, but it is not clear if that is the actual size. Dwell times for each should also be noted. Ilmenite is usually robust against damage by the electron beam, but the amorphous rim could be less so, which could affect compositional measurements.

Reply: All these detailed test conditions are supplemented in the Method section.

I would really love to see how the adjacent mineral phases respond to space weathering and if similar trends can be related clearly to the ilmenite, potentially with “actual” exposure ages calculated for the silicates using solar flare track dating. I understand that would be a different manuscript, but encourage the authors to publish on that aspect of these samples as well.

Reply: Thank you for your suggestion. We have not observed solar flare tracks in these ilmenite grains, which are more common in plagioclase and olivine minerals (Keller et al., 2021). Solar flare tracks have been used as indicators of exposure history for decades. We think this is a very good idea, and we will try to study the characteristics of their adjacent

mineral phases to see if we can constrain an absolute exposure age in the following work. In addition, we think the combination of solar wind irradiation equipment and the in-situ TEM instrument can help us to in-situ and real-time to observe the modification of material by solar wind irradiation and provide an effective means of quantifying the specific characteristics of the lunar ilmenite rims in response to different levels of solar wind irradiation on the Moon.

We added “Many indices have been proposed in the last decades to estimate the exposure time of lunar soils, such as agglutinate content, Is/FeO parameter, noble gas contents, and the solar flare track density, and our study is an alternative approach to identifying soil exposures for cases where only a small amount of samples are returned, such as deep lunar regolith and asteroids” in lines 393 – 397 of the revised manuscript.

Several references are incomplete or not formatted correctly, including Ref. 4, 10, 27, 32. In particular, I cannot assess if Ref. 10 and 27 are the appropriate citations where they are used.

Reply: Thanks for your careful review. We have corrected formatting errors in the references and made them more appropriately cited.

Specific Comments:

Ln 45–46: While there is ongoing discussion of differentiating between different processes contributing to space weathering, the citations here are pre-2000, and significant work has been done on this topic in the past 25+ years, making these not the best references for the current state of the field or what “remains” an unsolved problem.

Reply: Thank you for your comment. We added some of the most recent papers here (Pieter et al., 2016; Denevi et al., 2023). We expanded this sentence as “effectively distinguishing between their effects in modifying lunar material remains a significant unsolved problem, especially for the formation mechanisms of optically active iron particles in lunar soil” in lines 48 – 50 of the revised manuscript.

Ln 56: I love ilmenite but do not see how it is “more complex” compared to the responses of silicates, which can have basically all of these same features (amorphization,

loss of O, npFeO formation, vesicle formation). In fact, its simplicity in having so few elements is part of its power in this context, as any depositional rims are easily recognized.

Reply: Thank you for your comment. Ion simulation experiments have shown that ilmenite exhibits more complex microstructural rims than silicates under ion irradiation (Christoffersen et al., 2011). The studies of Apollo mature lunar soils also exhibit key differences in response to space radiation processing relative to silicates, such as structural defects and more pronounced trapping helium (Burgess et al., 2018; Christoffersen et al., 1996). The complexity we are talking about here is that the weathered rims of ilmenite can contain all of these features (e.g., helium-bearing vesicles, significant structural defects, and nanophase iron particles) simultaneously, which is difficult to observe simultaneously within the amorphous rim of silicates.

We rewrote this sentence as “exhibit complex chemical and structural responses to space weathering, such as the helium-bearing vesicles, significant structural defects, and variations of iron and titanium valence states, as compared to the amorphous rims on lunar silicates. More critically, the simplicity of ilmenite elemental composition (Fe, Ti, and O) makes it easy to identify impact-induced vapor deposition on lunar soil, thus making the ilmenite the most appropriate target for studying solar wind irradiation on the Moon” in lines 62 – 68 of the revised manuscript.

Ln 81: “vesicles...entrained” in the rim implies a process that is not accurate, as it has an implication of “dragging along with” or picking up during movement.

Reply: We rewrote it as “embedded” in line 93 of the revised manuscript.

Ln 90: I think the sentence beginning “All of these microscopic characteristics...” is lacking a verb such as “show”?

Reply: Thank you for your comment. We revised it in the revised manuscript.

Ln 137–138: “the densely damaged rims should be a Ti³⁺-bearing oxide” I am confused by this statement and the use of “However” in the following sentence. Does this mean that the Ti³⁺ component would not initially be incorporated into the ilmenite? And as noted above, small FeO metal grains could be very difficult to see in the EELS data. This

seems to say that Ti^{3+} oxide could not form without change in Fe valence, but localized loss of Fe (relative increase of Ti) could have similar effects.

Reply: Thank you for your comment, there's no logical relationship between these two sentences. We had deleted the word "However" here. As you said, the formation of Ti^{3+} oxides should not depend on the reduction of iron.

Ln 147: "at" should be "in".

Reply: We revised it in the revised manuscript.

Ln 225: I am not sure what "certain orientation" means here. The grain surface?

Reply: Thank you for your comment, which was also raised by another reviewer. We have changed it to "The structural damage is concentrated on the surface of Chang'e-5 ilmenite, and its depth range (100–200 nm) coincides exactly with the results of energetic particle irradiation under simulated lunar conditions" in lines 260 – 262 of the revised manuscript.

Ln 229–232: There are several grammatical errors here that make this set of sentences difficult to understand.

Reply: Thank you for your comment. We rewrote this sentence as "In Chang'e-5 ilmenite grains, the earliest structural damage appears as localized nano-scale structural disorder in the most surface layer, and with the accumulation of solar wind irradiation, the most surface layer is gradually amorphous, and the structural disorders and planar defects appear at a deeper depth" in lines 265 – 268 of the revised manuscript.

Ln 271: Use of "decisive" is confusing here to me. Do you mean it is the only heating required? I would use "sufficient" instead.

Reply: Thank you for your suggestion, we revised it in the revised manuscript.

Ln 355: Typo on "microns" and "to be stuck" is awkward.

Reply: Thanks for your careful review. We rewrote this sentence as "lunar soils larger than 100 microns to be fixed on the top of a scanning electron microscope (SEM) stub with a

carbon substrate” in lines 402 – 403 of the revised manuscript.

Fig. 2b,c: white lines are not labeled. Are they meant to show crystal orientation?

Reply: Thank you for your comment, we added labels to these Figures. We added “and the white lines represent the (110) and (018) lattice fringes of npFe^0 and ilmenite, respectively” in lines 132-133 of the revised manuscript.

Fig. 3b: scale bar doesn't show.

Reply: We are sorry for our carelessness, and we added a scale bar in the Fig. 3b.

Decision letter and referee reports: second round

6th Jun 24

Dear Professor Li,

Your revised manuscript titled "Diverse space-weathered rims on ilmenite: Implications for the exposure history of Chang'e-5 lunar soils" has now been seen again by our 3 reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are retained. We remain interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication. In particular, two reviewers would like the issues relating to the map color scheme to be addressed and reviewer 1 notes that the FIB section thicknesses are still not appropriately included in the manuscript.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and the completed checklist:

link redacted

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We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

Please do not hesitate to contact us if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Candice Bedford, PhD
Editorial Board Member

Communications Earth & Environment
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Joe Aslin
Deputy Editor
Communications Earth & Environment

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Please refer to our data policies at <http://www.nature.com/authors/policies/availability.html>.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have sufficiently addressed most of the first round of reviewer comments and have made corrections to the manuscript and should be commended for their extensive efforts. However, there are a few minor concerns remaining.

1) The authors have not adequately addressed the concerns raised by Reviewer 1 and Reviewer 3 regarding the EDS methodology and the sample thickness. Saying that they used an “interactive TEM quantitative method” is not enough to ensure reproducibility of the data. The Bruker Esprit software they used will indicate information about the quantification method being used. An example statement type: “EDS data were quantified using the Cliff-Lorimer method without absorption correction and with standard k-factors”. This also underscores the importance of knowing the thickness of the sample for EDS (and EELS) to determine if absorption correction is needed. Judging thickness in FIB is an estimate with error up to several tens of nanometers, it would be ideal if they measured thickness instead from the zero-loss-peak which is routine during EELS analysis.

2) The authors have not adequately addressed the concerns raised by Reviewer 2 for L106 regarding maintaining a consistent color scheme throughout the manuscript. This is stylistically important and, in this case, it would be best to reassign the colors in the single element-maps in Fig. 2 to match those of Fig. 4.

3) There is a labeling issue with the data repository. The data are available, but the files at the shared location are completely mislabeled and so it not possible to know which file goes with each figure.

Additional specific comments:

L31: Correct “proved” to “prove” to stay in active voice.

L58: The study in reference 10 (Christoffersen et al. 2011) compares silicates and sulfides, not silicates and oxides, so not sure why it is included. And Reference 11 (Christoffersen et al. 2007) does not identify that ilmenite's resistance above that of pyroxene is due to its close-packed crystal structure. I would recommend removing "owing to its close-packed crystal structure". Reviewer 3 brought this up too.

L64: Remove "the" after "making"

L395: Replace "to be" with "were"

L396: Remove the comma after "substrate" and insert "and" before "coated"

L403: Replace "confirmed" with "characterized"

L407: Should be "focused" not "focus"

L428: Replace "of STEM mode" with diameter.

Reviewer #2 (Remarks to the Author):

This manuscript focuses on the chemical and microstructural characterization of ilmenite grains returned by the Chang'e-5 mission through transmission electron microscopy, energy dispersive X-ray spectroscopy, and electron energy loss spectroscopy. Ilmenite grains exhibit different degrees of space weathering and their alteration has been attributed to the influence of the solar wind on the lunar surface. In addition, different degrees of alteration were used to reconstruct the stratigraphic profiles from the sampling region to elucidate regolith overturn processes. The results from this manuscript will contribute to a better understanding of the space weathering processes on the Moon.

See additional comments about the manuscript in the attached file.

Reviewer #3 (Remarks to the Author):

The authors have successfully addressed the comments from the reviewers, and the manuscript is much clearer and easier to read. The least damaged rim category is now "undamaged" in the main manuscript, but "slightly damaged" in the Supplementary Material. I also was aware of several minor typos although nothing that affects understanding.

Comments to the authors of the manuscript

Second round

“Diverse space-weathered rims on ilmenite: constraining the exposure history of Chang’e-5 lunar soils”

Line 106. Consider rewording “extremely intense solar wind irradiation” as it is not clear what are you referring to (fluence, flux, exposure time).

Line 112. Consider replacing the word “delineated” to “defined” or similar.

Line 127. You mentioned in the rebuttal letter that you considered the colors used in Fig 4 more appropriate for visualization of the chemical changes. Then, consider changing the colors in Fig 2 to be consistent with the colors used in Fig 4.

Line 167. In the manuscript, you used both EDX and EDS to refer to energy dispersive X-ray spectroscopy. Please choose only one of the acronyms and modify the manuscript accordingly.

Line 189-190. Again, for clarity consider rewording “are iron-rich in the EDX map” to “are shown as iron-rich regions in the EDX map” as saying that npFe^0 are iron-rich is redundant.

Line 203. In the last two rows in the supplementary table, is challenging to know which sample is associated to which type of rim. Consider adding the type of rim below the sample numbers and maybe leave the right column only as substrate ilmenite, rim, outer rim, etc.

Line 235. Expand on the interpretation of the presence of Si in c2, e2, and d2 layered maps as it is clear that Si is present in the top surface of the lamellas. If concerned about the presence of exogenous materials (e.g., vapor redeposition from surrounding mineral phases), why you did not include other elements present in the adjacent silicates (e.g., Mg, Na, Ca, etc)?

Line 265-268. The entire sentence “In Chang’e-5 ilmenite grains, the earliest structural damage appears as localized nano-scale structural disorder in the most superficial layer, and with the accumulation of solar wind irradiation, the superficial layer is gradually amorphous, and the structural disorders and planar defects appear at a deeper depth” is difficult to understand. Therefore, change to “In the Chang’e-5 ilmenite grains, the early-stage structural damage appears as localized ~~nano-scale structural disorder~~ (be specific about the features) in the most superficial layer (include the depth), and with the accumulation of solar wind irradiation, the superficial layer become amorphous, and the structural disorders and planar defects appear at a greater depth” or similar.

Line 283. You mentioned “oxygen deficiency and elemental Fe -Ti reduction are the most pronounced chemical alteration at the weathered ilmenite rims”. However, you didn’t include O maps in your Fig. 4 layered maps. I consider it critical to include those maps. It is clear the variation in the O in the EDS maps of samples CE5Z1002 #2 and CE5C0200 #2 that you included in the rebuttal letter. Including O maps is important to show the O variations between

the different rim categories. If you are concerned about the visualization of the layered maps, try playing with the map colors.

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Line 426. Define “SDD” before using the acronym.

Line 452. Run “in” Python.

Ms. Ref. No.: COMMSENV–24–0071A

Title: Diverse space-weathered rims on ilmenite: Implications for the exposure history of Chang'e-5 lunar soils.

Authors: Zhuang Guo, Mingwei Zhang, Bojun Jia, Yang Li, Wenzhe Fa, Yong Pang, Chenxi Zhu, Yuanyun Wen, Xiongyao Li, Jianzhong Liu, Ziyuan Ouyang

Communications Earth & Environment

Reviewer #1 (Remarks to the Author):

The authors have sufficiently addressed most of the first round of reviewer comments and have made corrections to the manuscript and should be commended for their extensive efforts. However, there are a few minor concerns remaining.

1) The authors have not adequately addressed the concerns raised by Reviewer 1 and Reviewer 3 regarding the EDS methodology and the sample thickness. Saying that they used an “interactive TEM quantitative method” is not enough to ensure reproducibility of the data. The Bruker Esprit software they used will indicate information about the quantification method being used. An example statement type: “EDS data were quantified using the Cliff-Lorimer method without absorption correction and with standard k-factors”. This also underscores the importance of knowing the thickness of the sample for EDS (and EELS) to determine if absorption correction is needed. Judging thickness in FIB is an estimate with error up to several tens of nanometers, it would be ideal if they measured thickness instead from the zero-loss-peak which is routine during EELS analysis.

Reply: Thanks for your approval of our revisions and your new professional comments. We have carefully considered your suggestions and made changes to our manuscript. A precise description of the quantitative methodology and the calculation of the FIB thickness based on the EELS zero-loss peak has been added to the revised manuscript.

We changed “The quantification of TEM-EDS data was calculated by an interactive TEM quantitative method in Bruker Esprit software” to “TEM-EDS maps were quantified using the Cliff-Lorimer ratio technique with standard k-factors in Bruker Esprit software for the Talos” in lines 435–437 of the revised manuscript.

We added “The relative thickness (t/λ) of FIB films was calculated based on the equation

of $\frac{t}{\lambda} = \ln \frac{I}{I_0}$, where t is the absolute thickness of the FIB films, λ electron mean-free-path, and I is the total integrated area under the EELS spectrum and I_0 is the area under the zero-loss peak (ZLP). The relative thickness (t/λ) of these studied FIB films was extracted from their EELS ZLP spectra using the Gatan Microscopy Suite (GMS) software, with values ranging from 0.34 to 0.87. Additionally, the λ value of the FeTiO₃ was calculated to be approximately 90 nm by using the Mean Free Path Estimator of DigitalMicrograph script (available at <http://www.dmscripting.com/meanfreepathestimator.html>). Therefore, the absolute thickness (t) of these studied FIB films was determined to be 30.6–78.3 nm” in lines 434-443 of the revised manuscript, and we also added the calculated thickness results in Figure 4. The original FIB foil thickness data and results are publicly available in the provided data repository (<https://figshare.com/s/f67768f7856487675aed>).

In this cases, the A and F corrections during the TEM-EDS microanalysis are generally omitted because the sample is so thin (tens of nanometers).

2) The authors have not adequately addressed the concerns raised by Reviewer 2 for L106 regarding maintaining a consistent color scheme throughout the manuscript. This is stylistically important and, in this case, it would be best to reassign the colors in the single element-maps in Fig. 2 to match those of Fig. 4.

Reply: Thank you for your comment. We have reassigned the color scheme in Figure 2 to be consistent with Figure 4.

3) There is a labeling issue with the data repository. The data are available, but the files at the shared location are completely mislabeled and so it not possible to know which file goes with each figure.

Reply: We apologize for our carelessness. We have rechecked and ensured that all original data is labeled correctly. In addition, we have added the results of the sample thickness calculations to the data repository (<https://figshare.com/s/f67768f7856487675aed>).

Additional specific comments:

L31: Correct “proved” to “prove” to stay in active voice.

L58: The study in reference 10 (Christoffersen et al. 2011) compares silicates and sulfides,

not silicates and oxides, so not sure why it is included. And Reference 11 (Christoffersen et al. 2007) does not identify that ilmenite's resistance above that of pyroxene is due to its close-packed crystal structure. I would recommend removing "owing to its close-packed crystal structure". Reviewer 3 brought this up too.

L64: Remove "the" after "making"

L395: Replace "to be" with "were"

L396: Remove the comma after "substrate" and insert "and" before "coated"

L403: Replace "confirmed" with "characterized"

L407: Should be "focused" not "focus"

L428: Replace "of STEM mode" with diameter.

[Reply: Thank you for your careful review, we have corrected all these issues you raised.](#)

Reviewer #2 (Remarks to the Author):

This manuscript focuses on the chemical and microstructural characterization of ilmenite grains returned by the Chang'e-5 mission through transmission electron microscopy, energy dispersive X-ray spectroscopy, and electron energy loss spectroscopy. Ilmenite grains exhibit different degrees of space weathering and their alteration has been attributed to the influence of the solar wind on the lunar surface. In addition, different degrees of alteration were used to reconstruct the stratigraphic profiles from the sampling region to elucidate regolith overturn processes. The results from this manuscript will contribute to a better understanding of the space weathering processes on the Moon.

See additional comments about the manuscript in the attached file.

[Reply: Thanks for your approval of our revisions and your new professional comments.](#)

Specific comments:

Line 106. Consider rewording "extremely intense solar wind irradiation" as it is not clear what are you referring to (fluence, flux, exposure time).

[Reply: Thank you for your suggestion. We changed it to "long-term exposure to solar wind irradiation". Line 103 of the revised manuscript.](#)

Line 112. Consider replacing the word "delineated" to "defined" or similar.

[Reply: We changed it to "defined" in line 109 of the revised manuscript.](#)

Line 127. You mentioned in the rebuttal letter that you considered the colors used in Fig 4 more appropriate for visualization of the chemical changes. Then, consider changing the colors in Fig 2 to be consistent with the colors used in Fig 4.

Reply: Thank you for your comment; other reviewers have also raised this issue. We have reassigned the color scheme in Figure 2 to make it consistent with Figure 4.

Line 167. In the manuscript, you used both EDX and EDS to refer to energy dispersive X-ray spectroscopy. Please choose only one of the acronyms and modify the manuscript accordingly.

Reply: Thank you for your careful review. We have used "EDS" throughout the text.

Line 189-190. Again, for clarity consider rewording “are iron-rich in the EDX map” to “are shown as iron-rich regions in the EDX map” as saying that npFe0 are iron-rich is redundant.

Reply: Thank you for your suggestion. We changed it to “shown as iron-rich regions” in line 189 of the revised manuscript.

Line 203. In the last two rows in the supplementary table, is challenging to know which sample is associated to which type of rim. Consider adding the type of rim below the sample numbers and maybe leave the right column only as substrate ilmenite, rim, outer rim, etc.

Reply: Thanks, we have added the type of rim below the sample numbers in the supplementary Table.

Line 235. Expand on the interpretation of the presence of Si in c2, e2, and d2 layered maps as it is clear that Si is present in the top surface of the lamellas. If concerned about the presence of exogenous materials (e.g., vapor redeposition from surrounding mineral phases), why you did not include other elements present in the adjacent silicates (e.g., Mg, Na, Ca, etc)?

Reply: Thank you for your comment. As we mentioned in our manuscript, the simplicity of ilmenite elemental composition (Fe, Ti, and O) makes it easy to identify impact-induced vapor deposition on lunar soil, and the Si element in the EDS maps an effective criteria for judging the presence of exogenous vapor deposited material on the ilmenite surfaces. Impact-induced selenium-bearing layers are not discussed in detail in the paper as this study focuses on solar wind alteration of ilmenite. Nevertheless, we objectively describe

the presence of silica material intermediate (Figure 4c2) and highly weathered (Figure 4d2 and 4e2) ilmenite grains, and more explanation was added in the revised manuscript.

We added “In addition, trace amounts of Si enriched on the uppermost surface of the ilmenite grain (Figure 4c2)” in lines 164-166 of the revised manuscript.

We added “and also shows the signal of the Si element” in lines 189 of the revised manuscript.

We added “A distinct Si-rich layer (~5 nm) was observed on the surface of the ilmenite grain, which is most likely a product of impact deposition and is not related to the solar wind alteration of ilmenite, thus excluding the Si-rich layer from the classification of the three sublayers described above (Figure 4e2)” in lines 203-206 of the revised manuscript.

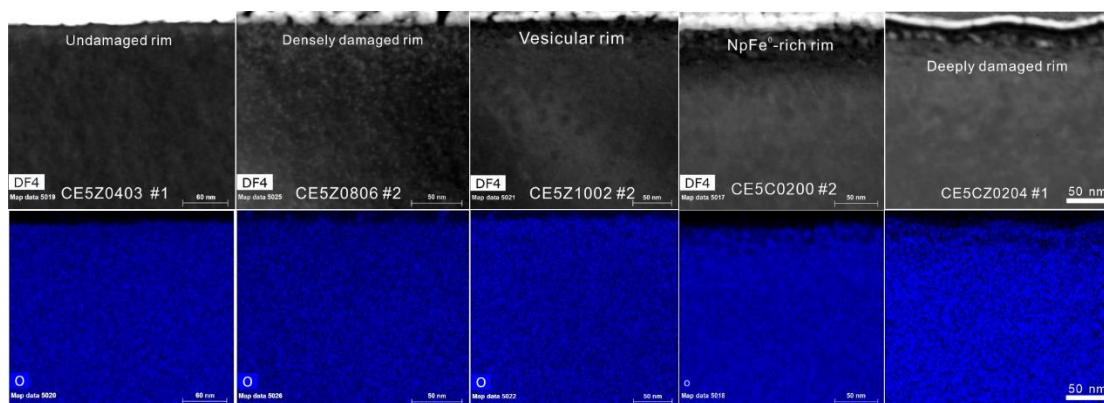
We added “Vapor deposit layers are widely distributed on the exposed surfaces of lunar soil and always exhibit elemental Si enrichment²¹. Therefore, the exotic Si compositions detected in the intermediate (vesicular rim) and highly weathered (npFe₀-rich rim and deeply damaged rim) ilmenite grains also suggest that these grains have experienced a long-term space exposure” in lines 257-261 of the revised manuscript.

Line 265-268. The entire sentence “In Chang’e-5 ilmenite grains, the earliest structural damage appears as localized nano-scale structural disorder in the most superficial layer, and with the accumulation of solar wind irradiation, the superficial layer is gradually amorphous, and the structural disorders and planar defects appear at a deeper depth” is difficult to understand. Therefore, change to “In the Chang’e-5 ilmenite grains, the early-stage structural damage appears as localized nano-scale structural disorder (be specific about the features) in the most superficial layer (include the depth), and with the accumulation of solar wind irradiation, the superficial layer becomes amorphous, and the structural disorders and planar defects appear at a greater depth” or similar.

Reply: Thank you for your suggestion. We have changed this sentence to “In the Chang'e-5 ilmenite grains, the early-stage structural damage appears as localized nano-scale structural disorders in the most superficial layer (~50 nm), and with the accumulation of solar wind irradiation, the superficial layer becomes amorphous, and the structural disorders and planar defects appear at a greater depth (up to 120 nm)” in lines 271-275 of the revised manuscript.

Line 283. You mentioned “oxygen deficiency and elemental Fe -Ti reduction are the most pronounced chemical alteration at the weathered ilmenite rims”. However, you didn’t include O maps in your Fig. 4 layered maps. I consider it critical to include those maps. It is clear the variation in the O in the EDS maps of samples CE5Z1002 #2 and CE5C0200 #2 that you included in the rebuttal letter. Including O maps is important to show the O variations between the different rim categories. If you are concerned about the visualization of the layered maps, try playing with the map colors.

Reply: Thank you for your comment. We have given quantitative results for oxygen in Supplementary Table 1, which show a clear change in O content with increasing surface depth of ilmenite grains. In the O map, the oxygen is constantly changing from the surface to the interior of the ilmenite spatial weathering edge, and the contrast in O content in the maps is not very clear. Undoubtedly it does provide data support for the important conclusion that oxygen deficiency and elemental Fe-Ti reduction should be correlated in our manuscript. We have therefore summarized the EDS O maps in the Supplementary Information as additional Fig. S7.



Line 298. In the manuscript, be more specific about what are you referring to when you mentioned “native”

Reply: Thank you for your comments. We want to clarify here that these npFe⁰ particles are not directly derived from the host ilmenite, they are most likely exogenous. We are sorry for the confusion about the word "native", and we have revised it to “indigenous npFe⁰ particles derived from the host ilmenite” in line 305 of the revised manuscript.

Line 352. Fig. 6 shows that the pre-C1 paleoregolith region only has ilmenites that present

slightly damage and deeply damage rims despite having experienced a significant number of overturn events. Can you expand on that?

Reply: Thank you for your comment. In the main text, we mentioned that “The most space-weathered ilmenite grain (deeply damaged rim) was identified from the CE5Z0204 lunar soil, suggesting that lunar soil depths greater than 65 cm must have experienced a relatively long exposure history”. I want to emphasize that although the exposure age of particles in a layer varies widely as expected, the presence of extremely space-weathered grains indicates that this layer of lunar soil has undergone a long exposure, which is exactly what we are focusing on.

However, as you say our observations also revealed the presence of weak weathered ilmenite particles in deep-drilled lunar soils (CE5Z0204), which seems contradictory to the long exposure it experienced. There are two main points to be made here; (1) We studied only a few particles, and even though exposed for a long time of this layer, there are still allow for the possible presence of grains from the newly-formed impact crater at that time, like the heterogeneous exposure histories of shoveled Chang’e-5 lunar soils (CE5C0200). (2) Our model constraints on the evolution of deep lunar soils are limited because their formation age and impact crater ejecta are unclear. The only thing we can confirm is that the CE5Z0204 is most likely from the pre-C1 paleoregolith layer exposed on the lunar surface prior to the formation of the Xu Guangqi crater, resulting in a distribution of ilmenite weathering features similar to that of the topmost layer of shoveled Chang’e-5 lunar soils. We added “Weakly weathered ilmenite grain (CE5Z0204 #2) was also found in the deep-drilled CE5Z0204 lunar soil, reflecting the possible presence of ejecta from the then-newly formed impact craters during the formation and evolution of the pre-Xu Guangqi ancient regolith. The result implies that unexposed grains may be present at any depth of the lunar regolith, but the presence of highly space-weathered grains should indicate that the lunar regolith has experienced long-term exposure” in lines 350-356 of the revised manuscript.

Line 395. Consider changing “such as” to “from”.

Line 402. Consider rewording “Selection of lunar soils larger than 100 microns to be to be fixed on the top of” to “Lunar soil grains larger than 100 microns were placed on top of” or similar

Line 426. Define “SDD” before using the acronym.

Line 452. Run “in” Python.

Reply: Thank you for your careful review and suggestions, we have corrected and revised all these issues you raised.

Reviewer #3 (Remarks to the Author):

The authors have successfully addressed the comments from the reviewers, and the manuscript is much clearer and easier to read. The least damaged rim category is now "undamaged" in the main manuscript, but "slightly damaged" in the Supplementary Material. I also was aware of several minor typos although nothing that affects understanding.

Reply: Thanks for your approval of our revised manuscript and careful review. We have double-checked our manuscript and corrected the spelling errors in the manuscript suggested by you and other reviewers to ensure that there were no typos throughout the text.

Decision letter and referee reports: third round

10th Jul 24

Dear Professor Li,

Your manuscript titled "Diverse space-weathered rims on ilmenite: Implications for the exposure history of Chang'e-5 lunar soils" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Candice Bedford, PhD

Editorial Board Member

Communications Earth & Environment

orcid.org/0000-0002-0783-1064

Joe Aslin

Deputy Editor,

Communications Earth & Environment

<https://www.nature.com/commsenv/>

Twitter: @CommsEarth

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have made satisfactory revisions to the manuscript and their response addressed all outstanding concerns raised by the reviewers.

Reviewer #2 (Remarks to the Author):

The manuscript has significantly improved as the authors have addressed all the comments from the second round of reviews. Please resolve the minor comments described below.

Line 56. Expand on why ilmenite is more resistant to amorphization than silicates.

Line 61. Specify which are the “significant structural defects”

Line 96. Was the EELS spectra acquired from panels 2a, b, c or d? If yes, show the location from where these spectra were extracted. Also, in panel 2d, what is the Ti-rich arrow showing?

Line 144. Consider removing the word “also”

Line 144. You mentioned that the EELS data show no change in the Ti and Fe L-edge peaks but you do

not show the Fe spectrum in Fig 4a3.

Line 147 Consider changing “weakest” to “lowest” or similar.

Line 154. Delete “atomic”

Line 154-155. Supplementary Figure 4 shows the opposite, an increase of O content with increasing distance from the surface. Also, what are you referring to as “disorders” in supplementary figure 4b?

Line 163. Can you specify which are the structural damage characteristics that you are referring to?

Line 168. After “(figure 4c2)” you could add “were identified” or similar.

Line 168. Replace “at” for “in”.

Line 172. Add in the parenthesis “Supplementary Table 1” that shows the O loss

Line 186-187. Consider rewording “ shows the signal of the Si element;” to “shows the presence of Si.” or similar. Also, refers the readers to the respective figure.

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but are not showing any Fe EDS map. Maybe refer the readers to Supplementary figure 7. Also, the size of the “O” labels in supplementary figure 7 is not consistent between the panels nor the thickness of the scale bars, and some maps have a label “map data”.

Line 210. Consider changing the word “encapsulated” to “present” or similar.

Line 276. What are you referring to as “disorders”? Please elaborate.

Line 396. Consider rewording the phrase “is highly likely were” to “was likely”.

Line 434. Remove “method, and the FIB method is a common technique for the in-situ preparation of

transmission electron microscope (TEM) foils”

Line 485. Please change “anonymous reviewer” to “L. Chaves” as I agreed to disclose my name.
Thank

you.

Ms. Ref. No.: COMMSENV-24-0071B

Title: Space-weathered rims on lunar ilmenite as an indicator for relative exposure ages of regolith.

Authors: Zhuang Guo, Mingwei Zhang, Bojun Jia, Yang Li, Wenzhe Fa, Yong Pang, Chenxi Zhu, Yuanyun Wen, Xiongyao Li, Jianzhong Liu, Ziyuan Ouyang

Communications Earth & Environment

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Line 485. Please change “anonymous reviewer” to “L. Chaves” as I agreed to disclose my name. Thank you.

[Reply: Thank you for your careful review, we have corrected all these issues you raised in the revised manuscript.](#)