RC1's Comments on Chapter 3.1:

(Authors' responses are shown in purple after each comment)

We would like to thank you for the comments and feedback. Below we respond to each of your comments individually.

There are multiple major issues here:

- In figure 1 and 2, you did not state the criteria which decide if an element is 'present' or 'absent'.
  For Figures 1 and 2, elemental presence or absence was determined from the fitted µ-XRF spectra using the open-source software PyXRF v.1.0.28 (Li et al., 2017). Elements suggested by PyXRF automatic peak finding were evaluated against the summed µ-XRF spectrum for each mapped area and manually reviewed during spectral fitting. An element was considered present when characteristic fluorescence features at the expected emission line energies were supported by the measured spectrum and retained in the final fitted model. Elements were classified as absent when the expected spectral features could not be resolved or when the apparent signal could be attributed to overlap with another fitted component. We have added this description to the Methods section and figure captions.

- The selection of elements spanning the x-axis does not match the collection of maps in the supplement. In addition, the collection of maps is also not consistent over all samples. And the maps itself do not back the present/absend matrices
  Thank you for pointing this out. We identified an error in the code used to plot Figure 1, which caused the elemental absence/presence matrix to be inconsistent with the µ-XRF maps shown in the Supplementary Figures S3-S7. This error has now been corrected, and Figure 1 has been updated in the revised manuscript to ensure consistency with the supplemental maps. We also revised the text in lines 224-233 to reflect the correct mention of elements present within the revised µ-XRF maps in Figures S3-S7. We apologize for the oversight and appreciate the reviewer bringing this to our attention.

- The selection of elements seems random. Are those the elements which were presented as most likely matches by the software? However, this selection is not backed by geochemical considerations. In the revision, please include due to their ion radius likely elements in feldspars, like strontium and rubidium while excluding unlikely elements like for example copper

  The selection of the elements presented in the µ-XFP maps was not random. The elements were selected based on the fluorescence components resolved in the spectra collected during mapping and fitted using PyXRF. Since synchrotron based µ-XRF is sensitive to trace level elemental components the presence of an element in these maps does not necessarily imply that it is a major structural component of the feldspar.

  Our measurements were performed at an incident energy of 10 keV, which precludes excitation of the K-shell fluorescence of Rb and Sr. Therefore, these elements cannot be evaluated using the present dataset. We have clarified in the revised manuscript that the elemental maps represent elements detected under our experimental conditions.

- The selections of elements in figure 1 and 2 do not match.

  Yes, the elements listed in Figures 1 and 2 are not identical because the natural samples (X7363 and X7368) include Vanadium (V) and Neodymium (Nd) signals that were not detected in the µ-XRF spectra of the reference feldspars. These elements were therefore included in Figure 2. The occurrence of V and Nd in X7363 and X7368 are not

completely unexpected, as both can occur in trace elements in volcanic materials from the Turkana Basin, Kenya.

- The μ-XRF maps in the supplement lack units for their x-, y-axis and scales
  The axis information for the μ-XRF maps are provided in the figure caption. We recognize that including this information directly onto the figure may improve clarity. Both the X and Y axes are in pixels, with a spatial resolution of 1 μm per pixel.

- You did not state how the pixel values of the element maps were calculated from the XRF-spectra, please clarify this. I guess the software did not perform a spectrum deconvolution, which likely would have required a set of expected elements, but did return just the signal intensities at the dominating K-alpha or L- alpha lines of the specific elements. However, this approach can lead to severe misinterpretations if the data is not reviewed carefully. This considered, the uranium map shown in figure S3 and the uranium presence shown in figure 1 are probably not valid. An 10 keV synchrotron beam can only exicate the M shell of uranium, which by the way is indicated by the small plot title “U_M” in figure S3. However, the sensitivity of M-alpha is very weak. But the photon energy of uraniums M-alpha is very close to the K-alpha line of potassium. Thus, I’m very sure your uranium signals are ghosts, arising from the strong potassium signals. If you compare the “K_K” map with the “U_M” map in S3, this is also very obvious
  We have clarified μ-XRF data-processing procedure in the revised Methods section of the manuscript. The elemental maps were generated using PyXRF v 1.0.28, which performs spectral fitting rather than assigning pixel values from the intensity of a single K-alpha, L-alpha, or M-alpha emission channel. For each mapped area, elemental components were first identified through automatic peak finding and manually reviewed, with components added or removed as appropriate. The summed μ-XRF spectrum was then fitted using the selected elemental fluorescence components and associated fitting parameters. This resulting spectral model was then applied to the spectrum at each individual pixel, producing a two-dimensional map of the fitted fluorescence contribution for each element. The elemental fits incorporate the activated fluorescence lines associated with each selected element. We agree that careful evaluation of the fitted spectra is particularly important where spectral overlap may occur.

  We agree with the reviewer that the assignment of uranium in Figures 1 and S3 is not sufficiently supported by this dataset. At the 10 keV excitation energy used in this study, the uranium (U) L-shell cannot be excited, and uranium detection is therefore restricted to the comparatively weak U M-lines. These occur in a spectral region that strongly overlaps with the K K-lines, which is particularly problematic for the K-rich feldspars examined here. Upon re-examination, we also find that the spatial distribution of the fitted “U_M” map closely corresponds to the “K_K” map. We therefore cannot confidently distinguish the fitted U signal from spectral interference associated with K. We have removed the U assignment from Figures 1 and S3 and removed the corresponding mention of uranium from the manuscript. We note that removal of the U assignment does not substantially affect the broader conclusions of the study, as U was not used as a primary basis for interpreting feldspar composition or IR-RF behavior.

- There also other possible ghost signals in the data sets: For example, K-alpha of titatinum has nearly the same photon energy as L-alpha of barium. In order to distinguish between them you have to look also at L-beta of barium. Other example: K-beta of Fe overlaps K-alpha of Co. Thus, strong iron signals may lead to false-positive detection of cobalt.

We agree that careful evaluation of the fitted spectra is important, particularly where spectral overlap may occur. We have therefore re-examined the fitted µ-XRF spectra and elemental maps with particular attention to potential peak overlaps. We will clarify this evaluation in the revised Methods.

We paid particular attention to distinguishing barium (Ba) and titanium (Ti), during spectral fitting. The Ba L-series contains a greater number of emission lines than the Ti K-series, providing additional constraints for peak identification. When the expected Ti, V, and Cr K-line positions are compared with the measured spectra, together with the full Ba L-series, only the Ba L-lines reproduce the complete pattern of the observed peaks. We therefore assign these features to Ba rather than Ti for those samples.

For the presence of Co in the Figure S6, we agree with the reviewer that the Co assignment is not sufficiently supported and results from the overlap between Fe K-beta and Co K-alpha. Co was included in the fitting model only for this single map and was not included in the analysis of the remainder of the dataset. We have therefore removed the Co assignment from Figure 1 and S6 and updated the elemental description for anorthite in lines 240-241.

All this considered, the µ-XRF data needs to be re-analysed and the maps and figures reworked. Please consider the data analysis approach used in your own workgroup previously, see Sontag-Gonzales et al. (2024). In addition, I recommend to read through Riedesel et al. (2026), DOI:10.1007/s00269-026-01345-z. Especially their figure 2 shows, how geochemistry correlations of feldspars can also be resolved.
The µ-XRF data in this study were analyzed following the same method used in Sontag-González et al. (2024) and using the same open-source software PyXRF v.1.0.28 (Li et al., 2017). In response to this comment, we have also re-examined the analysis of the µ-XRF data and resulting elemental maps to confirm that the analysis is consistent with this approach.

We appreciate the recommendation to consider the work of Riedesel et al. (2026). Their geochemical correlation approach provides a valuable framework for resolving compositional relationships within feldspars. However, their analyses are based on electron microprobe measurements, whereas our data were collected using synchrotron based µ-XRF with an excitation energy of 10 keV on grains originating from different sample locations. The purpose of the present µ-XRF analysis is to identify elements detected within individual grains and assess broad compositional differences associated with the observed IR-RF behavior, rather than to derive quantitative chemical correlations. Our interpretation is limited to evaluating whether simple patterns of elemental occurrence correspond with IR-RF dose-response curve categories, while recognizing that more subtle compositional and structural controls likely influence IR-RF behavior.

We have made the analytical scope and these limitations clearer. We will mention the findings of Riedesel et al. (2026) in the updated manuscript.

Regarding your selection of elements, I recommend to go just with the elements used by Riedesel et al.(2026) and those listed by the reference compositions stated in table S1. Taken the limitations arising from your µ-XRF set-up into account, this leaves you with Ca, K, Fe, Mn and maybe Ba and Ti, if you can ensure they are spectrally resolved. In case you switch from your yes/no matrices to an more quantitative display, you may also add Si and Al.
We do not think that restricting the elemental maps solely to the elements reported by Riedesel et al. (2026) or listed in the reference compositions in Table S1 would best represent the

purpose of our μ-XRF analysis. Our measurements were collected using synchrotron μ-XRF with an excitation energy at 10 keV, and the elemental maps presented here are based on elements for which fluorescence signal has been identified and spatial distributions revolved in the measured spectra. The goal of this analysis is not to provide a quantitative geochemical classification of feldspar to directly compare elemental concentrations, as our experimental setup does not provide the analytical capability required for this type of quantitative assessment. Additionally, very low-Z elements such as aluminum (Al) and silicon (Si) cannot be detected with this experimental configuration because their fluorescence lines are strongly attenuated in air and will not be discussed. Here we document the elemental signals detected within individual grains for this experimental setup and explore board compositional relationships with their IR-RF behavior.

Regarding the supplement. Please ensure that the μ-XRF maps meet basic scientific standards and correlate to the findings and figures in the manuscript. In addition, I recommend to add one full set of maps for one aliquot of X7363 and X7368 each to the supplement. Also, please add some example XRF spectra of points marked in the maps to the supplement.
We again apologize for the inconsistency between Figure 1 and supplementary Figures S3-S7. We have corrected the underlying plotting error and have ensured that the revised μ-XRF maps are consistent with the findings and figures presented in the manuscript. We have also added a set of μ-XRF elemental maps for representative grains from X7363 and X7368 to the Supplementary Material. In addition, we have included an example of the summed μ-XRF spectra for the mapped areas used in our PyXRF analysis. We clarify in the revised Supplementary Material that these spectra represent the full mapped area rather than individual point measurements.

RC1's Specific Comments:

(Authors' responses are shown in purple after each comment)

Line 41: Please refer also to the photon energy of the 880 nm emission
We have updated the text to include the photon energy of 1.41 eV for the 880 nm emission.

Line 50: You refer to the 'natural' RF curve but you did not define beforehand that (1) returns you the natural RF curve
Thanks for pointing this out, we have updated the wording in (1) to clarify that it is the 'natural' RF curve.

Line 54: I guess you referred to Frouin et al. (2015) instead of Frouin et al. (2017)
This was meant to reference the PhD work of Frouin, 2014, the text has been updated to reflect this.

Line 60: Please add a single sentence of the origin of the IR-RF signal according to the band model. Something like: "According to band model used in the early studies, the IR-RF signal arises from
Done.

Line 60/61: The sentence does not sound scientific. I suggest something like: "At the onset of the DRC, the signal intensity is at maximum … etc."
Done.

Line 108: Please refer to the SBU albite the same style as the other samples, by writing for example "Albite SBU Amelia". Also, is there a reference to this sample?

We have updated the naming of the Albite to reflect the style of the other samples. There is not a specific reference linked to this sample. However, we have included the elemental composition of the sample in Table S1.

Line 117: Again, please keep the style consistent, by writing for example: "… sanidine FCs (An1Ab24Or2; Zimmerman and McIntosh 2011) from Fish Canyon Tu (Cebula et al. 1986)"
Done.

Line 129: Small writing error: "… significance to Pleistocence …"
Done.

Line 143: "… they targeted …". To which work do you refer? Sontag-Gonzales et al. or Nazaretzki et al.? Please clarify in the text.
'They' refers to the work done by Sontag-González et al. (2024). We have updated this sentence in the text to be more clear.

Line 153: Please put "at excitation energies of" before "10 keV" to prevent a misinterpretation that you mapped the 10 keV fluorescence
Done.

Line 174: I suggest to add the preferred wavelength ranges of the PMTs
We have added the wavelength ranges for each PMT for both machines mentioned.

Line 175: Did you mean IRSL instead of IRPL? If not, aren't the RF PMT better suited to the NIR emission of IRPL?
This was a typo and was meant to refer to IRSL. We have updated the text to reflect the correction.

Line 179/180: "…, which has been reported to … light sensitivity". Please, either provide a reference or remove this half-sentence. I suggest to remove it, as this statement should be part of either the introduction or discussion.
We have removed the latter half of the sentence.

Line 192 – 194: I would not list the used lenses here as this is usualy not done in publications like these. Is this the same optic as used in Kumar et al. (2026)? However, you could add the lens information to the caption of supplement figures S1 along with a statement that "reflection and transmission loses caused by the lenses are not taken inte account"
Yes, these are the same optics described in Kumer et al. (2026).

While we understand the hesitation to include the details of the optics in the text, we feel that it is important to retain these details in the instrument descriptions since the lenses differ between the two separate machines. We feel that providing the information will also establish a clear record of the machine configuration, which could facilitate comparisons with future studies. Transmission losses caused by the stated lenses were in fact taken into account for Figure S1.

Line 201: Not an issue with this manuscript but a technical recommendation: Those cameras support lower readout rates with lower readout noise. The frame-transfer will be slower, but given you channel time of 5 sec, there is suicient time available to improve the signal-to-noise ratio.
Thank you for the suggestion, we will take this into consideration in our future measurements.

Line 267: So, the measurements were done after bleaching with settings like described

in Frouin et al. (2017)? And figure 4 displays RF_reg curves of single grains measured with the NIR-sensitive PMT? And did you use the SBU or the Oxford device? Please clarify in the text.
Correct, our measurements were done after the bleaching and measurement parameters following Frouin et al. (2017). Figure 4 displays the IR-RF regenerative curves of the single-grains measurement with the NIR-sensitive PMT. These measurements were conducted with the SBU device. The text and figure caption have been updated to reflect the machine configuration.

Figure 4: The curve describtions you did in line 273 to 284 are hard to follow when looking at the graphs. A lin-log or log-log scaling of the axes would be better for this purpose. However, I admit that changing the axes of fig. 4 and 5 would make the diagrams styles worse and harder to grasp. Please consider to add such figures to the supplement.
While we agree that the curve descriptions are not the most quantitative approach, when we tried alternative ways of plotting the data in Figure 4 (log-log, lin-log, or normalized plots), these approaches substantially reduced the readability of the figure. We therefore feel that the current figure provides the clearest representation of the data. However, if truly desired, we would be happy to include a log-log version of Figure 4 into the supplementary materials for further comparison. We have revised and condensed the curve description in lines 273 to 284 to follow a systematic description of the curves. We have compared the mean signal within the first 500 Gy to the mean signal over the last 1000 Gy of the measurement to classify the curves as increasing, decreasing, or a relatively flat response.

Figure 4: The y-axis title states a bin width of 2 Gy, which is a channel time of 10 sec if this is the SBU device. Is this correct? This info don't has to be part of the caption but should be mentioned in line 267.
Correct, this is the SBU device. We have specified this in line 267 and have added it to the figure caption for consistency.

Figure 4: Please add more information to the caption (bleached, single-grain, PMT, RF70 protocol) and add the bandpass width of the filters (850/30 instead just 850).
We have updated the figure caption with additional measurement parameters.

Line 301: The behaviour of which particular curves do you mean? 850 nm of sanidine? Please clarify.
The behavior of curves that demonstrate an increase in signal intensity with dose. We have clarified this in the text.

Line 303 – 311: You did already a curve discussion in lines 273 to 284. It would serve the readability and your argumentation string (line 312 builds upon line 302) if you in incoperate this paragraph into the previous curve discussion in a brief manner.
We have made this more concise and condensed this paragraph with the curve discussions in line 273 - 284.

Line 315: Please remove "all"
Done.

Figure 5 : Please add the background curve if available or a short statement that no background correction was done. Please state also, that these are single-grain measurements.
We have added the background measurement to the figure, and updated the figure caption to reflect that these are single-grain measurements completed with a PMT.

Line 320: Please correct meaning regarding “850-880nm”
This refers to the IR-RF dose response behaviors using the 850/30 nm and 880/10 nm filter. We have rewritten the sentence as follows:

“The contrasting IR-RF dose-response behavior observed between microcline and sanidine across measurements using the 850/30 and 880/10 nm filters, together with variability among the sanidine samples measured using the 850/30 nm filter, suggests that the IR-RF response is controlled by more than bulk feldspar composition.”

Line 327: Please write “K, Na and ..” and “ … and lead (Pb)”.
Done.

By the way, as you could not detect Sr and Pb appearance (and others) due to the too low excitation energy of your µ-XRF setting, you may discuss this briefly in the discussion.
Yes, that's a fair point. We have mentioned this in the revised discussion.

Line 329: Please provide a reference about the effects of inclusions in volcanic feldspars.
We have revised this sentence to be as follows:

“Grain-scale heterogeneity may also arise from composite mineralogy, micro-inclusions, or other chemically distinct phases that are common in feldspar grains derived from volcanic provenances (O’Gorman et al., 2021), and these distinct domains may contribute to spatial variations in luminescence behavior.”

Line 336: Please write “decreasging-signal grain population”.
Sure, we have updated the wording here and for the increasing-grain population found in line 337.

Line 351: Interesting idea! Please add the equation from Erfurt and Krbetschek (2003) here. Please add also the actual equation of how to calculate D95 from the fitting parameters of the Erfurt-Krbetscheck equation.
This is a good point, we have added both equations to the text, near line 353.

Supplement figure S8: I recommend to print the fitting parameters as well as R^2 and D95 to this figure. In addition, I recommend to add the same fitting diagrams for the other two samples shown in figure 6 to S8.
We have included the R^2 to this figure and updated the others to include R^2 and the fit with stretched-exponential function as shown in Figure S8.

Line 373: Which SLS settings did you use? If you used those of Frouin et al. (2017), why the longer bleaching?
We used the SLS settings described in Frouin et al., 2017. We used a longer bleaching time in this experiment to demonstrate that the shifts observed in the mixing experiments (microcline-alite and microcline-sanidine) result from the mineral mixing itself, rather than from repeated measurement and bleaching of the same grains.

Figure 7: Please add to caption that this are IR-RF measurements with the 850/30 (?) filter and that the grain was bleached for 6h between each cycle.
Yes, this was measured with the 850/30 nm filter using the SBU device, after a 6hr bleaching between each cycle. We have updated the figure caption to specify the measurement parameters.

Figure 8: It would be nice to have D95 points plotted in this figure. Please add also a short information about IR-RF with 850/30 filter and no backround correction to the

caption.
We have added both the D95 points and the background measurement to the figure. We have also updated the figure caption to specify the measurement parameters.

Line 404: I wonder if stretched exponential fitting is still suitable for the microline + three-sanidine aliquot? The curve goes up after ~ 1500 Gy. This can not be modelled by a stretched exponential. How does this effect the D95 value? Please elaborate.
This is a good point! While the one-microcline + three-sanidine aliquot starts to increase in IR-RF intensity above ~1500 Gy, the stretched-exponential fit to the full curve yields a D95 of 872 Gy with an R^2 of 0.943. To evaluate the inclusion of the higher-dose region signal increase, we repeated the stretched-exponential fit using only data up to 1500 Gy. The resulting D95 increases to ~1134 Gy and an R^2 of 0.998. This is ~30% increase in the D95 and improvement of the R^2 value suggests that the increase in the IR-RF signal in the high-dose region does influence the stretched-exponential fit and consequently the calculated D95. We have clarified this limitation in the revised manuscript and note that the D95 value for this aliquot, and others that exhibit any increase in dose should be interpreted with caution. This also highlights the need for further work evaluating the applicability of stretched-exponential fitting to IR-RF dose-response curves.

Line 441: "X7468" is probably a typo for "X7368"
Correct, thanks for catching this.

Line 452: I suggest to include figure S11 in the main text. The figure is too interesting to be hidden in the supplement. I recommend to also plot D95 points here. If you decide against this suggestion, some other figure, for example a D95 distribution, would be nice to underpin your main conclusion.
Agreed, we have decided to move Figure S11 into the main text and have updated it to include the D95 values for each curve. Thanks for this suggestion.

Line 462: I already wondered, figure 3 appeared misplaced earlier in the text. Just a suggestion: Maybe it fits better here?
Agreed, we have re-order the figures to have ‘Figure 3’ appear here.

Line 478/479: But why could red TL be an explanation for sanidines IR-RF behaviour? I do not see the phyisical link here. Please explain closer or remove this sentence.
We agree that the physical link is not clear in the original text. We did not intend to suggest that the red TL directly controls the IR-RF behavior of sanidine. We intended to refer to the broad Fe-related emission centered near ~710 nm that has been reported to interfere with the wavelength region used for IR-RF detection. We have revised the sentence to make the distinction clear and have removed the implication that the red TL provides a mechanism for the observed sanidine behavior.

Line 501: There is no Fig. 08 or is Fig. 3 meant? However, sorting the (newly analyzed) µ-XRF occurences into the IR-RF curve categories is a good idea.
This is a typo and is meant to reference Figure 3.

Line 520: You conclusion about the value of D95 is reasonable. But D95 depends on the validity of the fitting model. A stretched exponential might not be a good representation of an IR-RF curve in all cases. And in cases were it is not, D95 would contain a large systematic error. Thus, in order to be statistically valuable, D95 should come with a statistical measure of likelihood, like a p-value, a Chi² or a residual sign test, I don’t know what suits best. It is not necessary to provide such a measure in this manuscript, however you should at least discuss it in one or two sentences.

We agree with this point and feel that it raises a direction for future research. For the purpose of this manuscript, we will include the need for a statistical measure of likelihood for using the D95 value. However, specifying a specific one would require more experiments, samples, and an evaluation of the stretched-exponential fitting model.

Line 534: Please make clear, that the conclusions in this paragraph are only valid for SR IR-RF measurements and not for multi-grain PMT measurements. Even more, your conclusions are excellent arguments in favor of SR IR-RF.
We have clarified that the conclusions in this paragraph are specific for SR-RF measurements and should not be directly extended to multi-grain measurements. However, we have also emphasized that the variability observed at the single-grain level adds to a growing body of work suggesting the need for caution when evaluating multi-grain IR-RF measurements.

Line 551: You did develop a “quantitative, transferable screening criterion” with D95. Please consider to rephrase this sentence.
We have changed the wording of this sentence to reflect this.

References:

Frouin, M.: Les feldspaths comme support pour la datation par luminescence des gisements archéologiques et des séquences quaternaires d’Aquitaine, Ph.D. thesis, École Doctorale Montaigne Humanités, Université Bordeaux Montaigne, France, 391 pp., 10.70675/c7cc32f3z7150z468bza993z47118f93f4c9, 2014.

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Li, L., Yan, H., Xu, W., Yu, D., Heroux, A., Lee, W.-K., Campbell, S. I., and Chu, Y. S.: PyXRF: Python-based X-ray fluorescence analysis package, in: X-Ray Nanoimaging: Instruments and Methods III, SPIE Optical Engineering and Applications, 38-45, https://doi.org/10.1117/12.2272585, 2017.

O’Gorman, K., Tanner, D., Sontag-Gonz´ alez, M., Li, B., Brink, F., Jones, B.G., Dosseto, A., Jatmiko, Roberts, R.G., and Jacobs, Z.: Composite grains from volcanic terranes: internal dose rates of supposed ‘potassium-rich’ feldspar grains used for optical dating at Liang Bua, Indonesia, Quat. Geochronol., 64, https://doi.org/10.1016/j.quageo.2021.101182, 2021.