RC2's General Comments:

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

Thank you for the comments and feedback. Below we respond to each of your comments.

1. The synchrotron uXRF mapping is the key support of the conclusion drawn by this study, although it, to some extent, denies the main role of chemical composition on IR-RF behavior. It's better to state clearly the analytical constraints and detection limits by using 10 keV XRF and the accuracy and precision in determining the content of key elements, e.g., Fe, Mn, Ti, that may affect the luminescence production. Including the analysis of a standard sample is highly encouraged. Meanwhile, the authors need to bear in mind that the minor role of major element chemistry does not necessarily deny the contribution from the subtle roles of trace elements in forming recombination centers or altering the configuration of the crystal lattice.

   We agree that the analytical constraints of the 10 keV synchrotron μ-XRF measurements should be stated more clearly and we have expanded this discussion in the revised Methods section. The μ-XRF analysis in this study was used to identify fitted elemental fluorescence signals and their spatial distributions within individual grains of K-rich feldspar, rather than to determine quantitative elemental concentrations. Therefore, concentration-based detection limits, accuracy, or precision for Fe, Mn, Ti, or other specific elements were not determined as part of this experimental approach. We have revised the manuscript to clarify this distinction and the limitations on element detection with an incident energy of 10 keV.

   We appreciate the reviewer's suggestion to include a standard sample. However, the purpose of these μ-XRF measurements was qualitative elemental mapping rather than quantitative compositional analysis. A dedicated standard sample was not included in the experiment. Instead, reference feldspar samples of known mineralogical compositions were included to provide a comparative context for the elemental distributions observed in the natural samples.

   We agree with the reviewer that the absence of a clear relationship between the presence or absence of major elements and the IR-RF behavior does not exclude an important role for trace elements, lattice defects, or differences in the crystal structure in controlling luminescence behavior. This distinction is reflected in our Discussion, where we emphasize that the μ-XR results provide a first-order test of whether elemental presence or absence alone can distinguish IR-RF dose response curve categories, rather than a test of whether composition more broadly influences IR-RF.

2. The authors proposed to use the dose corresponding to 95% of the fitted asymptotic saturation level to screen out grains for IR-RF dating. It is better to have a sensitivity test using different functions to fit the dose response. In addition, I strongly encourage the authors to include two samples with independent age control, one fully bleached and the other not fully bleached, to show how the grain-level IR-RF characteristics would affect the dating accuracy.

   We appreciate this suggestion. In this study, we focused on the stretched-exponential function because it is the fitting approach most commonly used for IR-RF De determination. Here, we use D95 as a conservative and reproducible reference point in the high dose region of the fitted curve to compare apparent saturation behavior across feldspar compositions and synthetic mixtures, even where the absolute saturation point is not uniquely defined. Comparing saturation metrics calculated from different fitting

functions would be extremely valuable. However, this type of analysis would represent a broader methodological investigation that is beyond the scope of the present study. We specified in the text that the D95 values reported in this manuscript are specific to the stretched-exponential fits described in Erfurt and Krbetschek (2003). We have also included the equation for the stretched-exponential fits described in Erfurt and Krbetschek (2003) as well as the derived D95 equation in the revised manuscript.

We also agree that applying this single-grain level framework to samples with independent age control would provide an important test of how variability in single-grain IR-RF characteristics ultimately influences dating accuracy. This study was designed to characterize single-grain IR-RF behavior and establish the basis for a screening approach, rather than to perform a full validation using independently dated samples. We agree that this is an important next step and have emphasized this in the manuscript's conclusions.

RC2's Minor Comments:

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

1. In general, when I read through the m/s, I feel strongly that the authors need to balance the details of different sections. On one hand, the illustration of experimental results needs to be more accurate and concise. For instance, lines 273 to 284 are too descriptive; what do "sharp" increase or decrease mean exactly? I may suggest the authors adopt a more systematic way and simply claim whether the IR-RF DRCs are negative, positive, or not dependent upon the radiation doses. The use of a parameter, decay constant or growth constant to quantify such dependence is highly encouraged. In my opinion, it's better to avoid a long description of the curves. On the other hand, more details about the experiment and how you made the measurements need to be more detailed.

   We have revised and condensed the curve description in lines 273 to 284 to follow a systematic description of the curves. We defined a specific parameter to compare 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.

   We will include a more detailed description of aliquot preparation and measurement in Section 3.2.1.: Individual grains of each mineral (microcline, albite, anorthite, and sanidine) were carefully placed onto stainless steel discs using a silicone oil as an adhesive. Individual grain placement was performed under a microscope using a needle in our darkroom laboratory at SBU. The IR-RF signal of a single grain from each mineral was measured at SBU using the NIR-sensitive PMT and a 850/30 nm filter. Measurements were carried out at 70°C following the measurement and bleaching guidelines of Frouin et al. (2017), with irradiation continued up to a total dose of 12,000 Gy. This procedure was repeated for the 710/10 nm and 880/10 nm filters. Background measurements were conducted for each filter using the same measurement parameters to allow for direct comparison.

2. For Fig. 1, it is not clear whether the elements are mineral-limited or detection-limited. It's better to include both the consensus of the elements expected to be observed for different mineralogy and the actually detected elements.

   For Figure 1 the elemental presence/absence reflects what was detected and resolved in the μ-XRF spectra collected with an excitation energy of 10 keV, rather than the

assumption that elements not detected are mineralogically absent. Elemental identification was based on spectral fitting using the open-source software PyXRF v.1.0.28 (Li et al., 2017), with elements 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. We agree that the distinction between expected mineralogical composition and experimentally detected elements should be clearer. We have revised the Figure 1 caption to specify that the figure reports elements detected at 10 keV excitation energy, have added a better description of the criterion to the methods and figure captions, and we clarified in the Methods that non-detection reflects the analytical limits of the present measurements rather than confirmed mineralogical absence.

Fig 3 lacks the spatial scale for the u-XRF images, and there are also no labels for the X and Y axes.

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

Fig 4a, the Y-axis should also be IR-RF? I may suggest you integrate Fig. 4, 5, 6, and 8 into one figure to make the m/s more compact.

The Y-axis for Figure 4a is labeled RF because the ~710 nm signal is attributed to the broad red RF emission associated with the presence of iron (Fe), rather than the near-infrared emissions near 880 and 950 nm that are typically referred to as IR-RF (e.g., Erfurt and Krbetschek, 2003; Sontag-González et al., 2024, Kumar et al., 2026).

While we appreciate the suggestion, we disagree with the suggestion to integrate Figures 4, 5, 6, and 8 into a single figure. These figures present results from different experiments, each highlighting a distinct aspect of our single-grain IR-RF dataset. We feel that combining them could obscure the experimental distinctions and reduce the clarity of the results.

For the spatially resolved measurements, all data are shown in supplementary figures (Fig. S10 and S11), and no spatial information could be identified. I may suggest some attempt to align the IR-RF emission/characteristic maps with the u-XRF elemental maps and show the figure in the main text.

We have moved Figure S11 into the main text, with the addition of D95 values for each curve, and expanded more on this in the discussion. While we agree that an alignment between the SR-RF and μ-XRF would be extremely valuable, the spatial resolution of the SR-RF data and the μ-XRF maps are not directly comparable in this study. We are unable to reliably align individual IR-RF emissions with specific positions on the μ-XRF elemental maps. We have clarified this limitation in the revised manuscript and have included an example of one of our SR-RF images in the supplementary material.

3. For the mixing experiment, it would be better to specify more experimental details when synthesizing the DRC from individual grain curves.

We have included a more detailed description of aliquot preparation in Section 3.2.2.:

Individual grains of microcline, albite, anorthite, and sanidine, where applicable, were carefully placed onto stainless steel discs using a silicone oil as an adhesive. Individual grain placement was performed under a microscope using a needle in our darkroom laboratory at SBU.

For example, in the microcline-albite mixing experiment, one grain of albite was first placed onto a stainless-steel disc, and its IR-RF signal was measured at SBU using the NIR-sensitive PMT and a 850/30 nm filter. The same procedure was then repeated for one grain of microcline. Finally, a mixed aliquot containing one grain of microcline and one grain of albite was prepared by placing both grains on the same disc and measuring the combined IR-RF response. Each IR-RF measurement was completed using the same measurement parameters to allow for direct comparison between the individual grain and mixed-mineralogy aliquots. The same procedure was followed for the microcline-anorthite and microcline-sanidine mixing experiments.

References:

Erfurt, G. and Krbetschek, M. R.: IRSAR - A single-aliquot regenerative-dose dating protocol applied to the infrared radiofluorescence (IR-RF) of coarse-grain K-feldspar, Ancient TL, 21, 35-42, https://doi.org/10.26034/la.atl.2003.358, 2003.

Frouin, M., Huot, S., Kreutzer, S., Lahaye, C., Lamothe, M., Philippe, A., and Mercier, N.: An improved radiofluorescence single-aliquot regenerative dose protocol for K-feldspars, Quat. Geochronol., 38, 13-24, https://doi.org/10.1016/j.quageo.2016.11.004, 2017.

Kumar, R., Frouin, M., Sontag-González, M., Grandfield, T., Schwenninger, J.-L.: Spatially resolved infrared-radiofluorescence (SR IR-RF) measurements for single-grain feldspar dating., Rad. Phys. and Chem., 240, 113930, https://doi.org/10.1016/j.radphyschem.2026.113930, 2026.

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.

Sontag-González, M., Kumar, R., Schwenninger, J.-L., Thieme, J., Kreutzer, S., and Frouin, M.: Short communication: Synchrotron-based elemental mapping of single grains to investigate variable infrared-radiofluorescence emissions for luminescence dating, Geochronology, 6, 77-88, https://doi.org/10.5194/gchron-6-77-2024, 2024.