Response to review of Kadereit et al. A progressively elevated temperature (PET) IRSL SAR procedure – first experiments and results

Dear Svenja Riedesel,

Thank you for your detailed comments. To facilitate the reading of our response to others, we will begin by addressing your major points, as we understand them, before responding to each comment individually. Our response includes seven figures (**Fig. R-SR 1–R-SR 7**). To avoid repetition, we sometimes refer to our response to reviewer Jakob Wallinga and figures therein.

Response summary

(1) You assume that we essentially measure TL with "saddled-up" IRSL. This appears to be a misunderstanding of the underlying luminescence production process. PET-IRSL is a coupled phonon-photon stimulation process, and it should not be oversimplified as "TL + IRSL". Our experiments suggest that for two different IR stimulation power densities of 60 mW/cm² and 270 mW/cm² (Fig. S3.24) it shows a similar PET-peak position but different signal strengths. Our experiments with varying temperature-ramps (**Fig. R-SR 1**) let the PET-peak position shift to lower temperatures for lower heating rates. However, this does not indicate TL, but a heating-rate dependence of the signal, which is expected. In Figure 15 of our manuscript, we clearly show that TL is not an issue in our PET-IRSL SAR measurements.

(2) You suggest splitting the manuscript into two contributions.

We respectfully reject this request. When submitting the manuscript, we inquired whether the length might represent an obstacle for publication in GChron, and we explained that we do not prefer to split it up as this would lead to repetition without benefit to the reader. We regard the manuscript as a whole which does not consist of stand-alone aspects. After editorial inspection, the manuscript was passed as-is to the open discussion. Regardless, it does not seem to make sense to separately address issues raised here, e.g., partial bleaching detection, a-value determination, g-value assessment or any other aspect nowadays belonging integrally to a dating protocol. We show that all these aspects can be handled with PET-IRSL SAR.

(3) You request an explanation of the physics.

(3a) To date and to the best of our knowledge, no comprehensive numerical model for feldspar luminescence exists, unlike for quartz. We explain that our approach is based on the assumptions by Ankjærgaard & Jain (2010) and Jain & Ankjærgaard (2011), which we consider fully appropriate. Further speculations are beyond the scope of our contribution.

(3b) Regardless, we agree that additional in-depth investigations are desirable. They will require a changed experimental design and additional time-consuming measurements as performed, e.g., by Wang & Wintle (2013); Wang et al. (2014); Quin et al. (2015); or Riedesel & Duller (2022) for pIRIR and/or MET. However, such investigations are not a prerequisite for a manuscript introducing a new application procedure. Common practice for luminescence dating is to introduce new procedures, which includes pIRIR (Thomsen et al., 2008; Buylaert et al., 2009) and MET (Li and Li, 2011), whereas in-depth investigations are performed as more data become available or poorly understood age discrepancies become a burden, sometimes ten and more years later (e.g., Wang & Wintle, 2013; Wang et al., 2014; Quin et al., 2015; or Riedesel & Duller, 2022).

(3c) You seem to consider photon-transferred thermo-luminescence (PTTL) detectable as unstable post-IR isothermal TL (ITL) (Wang & Wintle 2013) an important issue. Yet presently, we have no indication that this might be of concern, as indicated, e.g., by poor dose recovery ratios (Quin et al., 2015). Flat end-to-end De-value plateaus recovering a given laboratory dose reliably can be produced with PET-IRSL SAR, if the thermal SAR parameter values are adjusted accordingly. We show this for each of our three test sediment samples and the commercially available feldspar specimen. While charge transfers by IR-stimulation is likely, PET-IRSL is a quasi-continuous process. There is no "time out" between a preceding and a subsequent IRSL readout like in pIRIR and MET, in which a potentially unstable ITL signal could be monitored. This also means that PET-IRSL fails the definition of PTTL that requires optical excitation to be observed in a subsequent TL signal. All we can provide are post-PET-IRSL TL curves as in **Figure R-SR 3ff** of the present response, but we would not know how to get in between individual measurement points of a signal curve without changing the energy regime fundamentally by either switching off IR-stimulation and/or temperature and thereby skewing the experimental data. This is different for pIRIR and MET which provides "natural" intervals for investigation. If we monitor TL-signals after we have turned off IR and/or temperature, this does not reflect the actual PET-IRSL process. To illustrate this reasoning with an example from geomorphology: Imagine a beach ridge, which is a pile of coarse-grained material deposited at the coast. New beach ridges can often be observed after a storm with heavy wave action at the land-ocean boundary. But was the ridge piled up while the waves were approaching the coast at maximum energy level – comparable to PET-IRSL with IR switched-on and temperature increasing to PET_{max}? Or was the material during that time in motion and the ridge was formed only when the system lost all transport energy as the direction of the water flow reversed on the upper beach in a down-slope direction? The loss of transport energy would compare to sudden IR-switch off and/or temperature cool-down. We assume that PET-IRSL with increasing temperature and IR switched-on compares to the high-energy wave system with pebbles on the coastline or charge carriers in the feldspar crystal in motion. The post-PET-IRSL "PTTL" signal would compare to the beach ridge that indicates that energy has been working in the feldspar crystal. It can proxy only the amount of total energy that was present very recently in the system, but not which pebble was deposited at which time or which fraction of "PTTL" contributed to which PET-IRSL signal integral, if it had a share in it at all.

Below, we will respond to your comments individually, using your text (here in black), and coloring our responses in blue.

The manuscript by Kadereit et al. presents a new feldspar luminescence dating protocol, which is based on a SAR IRSL protocol and follows the idea of a post-IR IRSL or MET-IRSL protocol. However, instead of performing individual IRSL measurements at increasing temperatures, the protocol measures the IRSL signal at continuously increasing temperatures, basically by performing a TL measurement while having the IR LEDs turned on.

We believe this comment oversimplifies the luminescence production process. You are right in stating that we ramp the heat while continuously stimulating with IRSL. However, it does not make it "TL plus IRSL". Both are separate processes, per definition and per underlying physics. If you were right, we could easily uncouple both processes, but this is not possible.

This is also relevant with respect to potential electron recycling from low-temperature to high-temperature readout. To minimize this potential draw-back as known from $pIR_{1st}IR_{2nd}$ and MET procedures, we consider it important to keep the (thermal) energy input constant (which we do during the initial 60 °C readout) and/or to increase the energy input during IR-stimulation (which we do during the high-temperature readout). You associate this issue of electron recycling with phototransferred TL (PTTL). We prefer the more general term "electron recycling", which we use in our manuscript, and which, we think, summarizes nicely the idea of Wang et al. (2014), that electrons responsible for the IR_{1st} signal are (to a larger part) also responsible for the IR_{2nd} signal, without further detailing the processes:

> *"During the initial IRSL stimulation electrons are retrapped in (phototransferred to) an intermediate trap correlated with a TL peak located between 200 and 250 °C. During the subsequent heating electrons are thermally evicted from this intermediate trap and a part of them is retrapped in electron traps responsible for*

the IRSL signal. Subsequent IR stimulation at an elevated temperature (e.g. 290 °C) gives the post-IR IRSL signal." (http://dx.doi.org/10.1016/j.radmeas.2014.03.006)

According to Wang et al. (2014) the electron-retrapping or recycling is a multi-step process of phototransfer and thermal transfer: **(1)** phototransfer from the IRSL-trap during IR_{1st} to TL traps previously emptied during the preheat step which function as a temporary/intermediate electron storage; **(2)** thermal transfer of these photo-transferred electrons from the intermediate storage during the temperature ramp for IR_{2nd} into the IR-trap previously sampled by IR_{1st}; **(3)** repeated IR-readout of these electrons from the IR-trap, this time by IR_{2nd}. Thus, the electrons are recycled successively "optically" (if IR is considered as optical), thermally and again optically. We are not sure whether PTTL describes this complex process adequately. As, however, we do not perform physical in-depth experiments to elucidate the electron reworking in the crystal, we feel happier with a less specific term, like electron recycling. For our measurement setup it is merely important to secure that any electron reworking during a lower-temperature step and/or in between a lower-temperature and a higher-temperature step is not reflected in a (significant) TL peak at the position of the respective temperature assisting the IRSL readout. In other words: the PET-peak is not actually a TL peak. That this is not the case, we showed with our experiment detailed in section 6.4, graphically illustrated in Figure 15 and further discussed in the discussion section 7.1.1, lines 626–634. Simply stated, PET-IRSL SAR is definitely not "TL plus IRSL".

Besides introducing the protocol, the authors also present a range of quality control tests, which are meant to characterise the performance of the protocol.

While I very much appreciate the considerable effort the authors have clearly put into this study, I have major reservations about the current presentation of the results. The authors have performed a very large number of experiments, which is visible by the length of the paper (57 pages), which includes 18 Figures (each of them consisting of multiple sub-figures), and the length of the supplementary material (161 pages). Regardless of the content, I find this paper too long and too complex. I would like to ask the authors to cut the paper considerably and consider splitting it into two papers.

We addressed this point above. We don't believe it is in the communities best interest to split the paper.

Besides the length, I find the structure of the paper not ideal and I am so far not convinced why this protocol is an improvement to existing measurement procedures. I would like to ask the authors to, when revising their manuscript, consider, which information is really important and how this information can be supported by a selection of clear figures and well-structured text.

See comments above.

Right from the beginning, I was wondering, why no introduction of the physical processes governing the proposed protocol was given and why the authors did not comment on the possibility of phototransfer affecting their signal (see e.g. Wang and Wintle, 2013; Qin et al., 2015; Riedesel and Duller, 2022). This is then addressed in section 6.4. However, the figures are not very clear and zoomed in version would be helpful and a further discussion of the drawbacks of phototransferred TL would be useful. At the end of section 6.4 the reader is left wondering, if one should now be concerned about the possibility of PT-TL or not.

The description of the physical processes is summarized in our manuscript, e.g., in lines 91–96 in the introduction and lines 989–992 in the conclusion. We do not assume any different electron trap(s) and recombination with electron holes as for the classic pIRIR and MET IRSL, and refer to the respective literature instead. We here focus on the application aspect of the new approach. A detailed assessment of the physics involved is not the scope of the present manuscript.

In the revised version we will mention the possibility of phototransferred TL (PTTL), or rather electron recycling, right in the introduction, as not to surprise the reader with this issue in section 6.4. Although we think that the term PTTL makes an analogy that is not fitting and somewhat bends the original definition of PTTL.

In lines 554–559 we describe the relevant parts of the signal curves and provide a hint that the slight signal increase resembling that of the zero-dose signal curve should not be an issue. As it is good practice in scientific papers to separate results (as in section 6.4) from the discussion, we discuss the issue in more detail in section 7.1.1 in lines 626–634. Our experiments (see Figure 15) do not give cause for concern regarding (significant) electron recycling. Further, if electron recycling was an issue, we expect we couldn’t produce end-to-end plateaus of De-values with appropriate SAR protocol parameters. Poor dose recovery is regarded to flag problems with “PTTL” in pIRIR and MET dating procedures (e.g., Quin et al., 2015).

We agree that more experiments investigating the physics of IRSL, pIRIR IRSL, MET IRSL and also PET IRSL are desirable. However, protocols (including SAR, pIRIR, MET) were developed and their basic application performance tested (as we did here) and subsequently more elaborate experiments were performed behind those protocols. Even if PET IRSL is to some degree flawed with the same drawbacks as the two- and multi-step pIRIR and MET procedures, we still see the clear advantage of evaluating the quasi-continuous data curves. This approach is especially important regarding assessment of the quasi-continuous De-value curves which are useful for bypassing the need for “wiggle-matching” ages across different MET-readout temperatures.

Please advise on statements we find contradictory, particularly regarding the number of figures which are pointed out as including too many and too few figures. The latter include zoomed in versions of Fig. 15; or extra graphs showing shifting PET-peak positions; or additional graphical presentations of what one might expect from the here presented measurement approach in the style of Bateman et al.

We detailed our expectations in lines 81–84, but will make it clearer that these are not just generally desirable issues but the specific aims of the present study.

Figure 15

You are critical of our Figure 15, which illustrates our experiments to test whether the PET-peak is a “hidden” TL-peak. The subfigures illustrate that this is not the case. As a focus of your critique was PTTL as proxied by TL peaks measured in between IR$_{1st}$ (usually in the range 50–60 °C) and IR$_{2nd}$ (usually in the range 225–290 °C), we reproduced the graph for HDS-1776 in Fig. 15a in a higher-resolution quality (**Fig. R-SR 2**). In addition, we include zoomed-in insets of the background signal, which rises slightly with increasing readout temperature. One blow-up shows the background data on linear scales (**Fig. R-SR 2b**), the other one on logarithmic scales (**Fig. R-SR 2c**).

The background increases with each abbreviated PET-IRSL SAR cycle (no normalization-dose cycles included or used for signal normalization in this experiment). This is expected, as thermal readout is only up to 320 °C (conforming to the preheat procedure of the PET-IRSL SAR measurements) or 280 °C in the PET-IRSL step, also conforming to that of the PET-IRSL SAR measurements. We deliberately did not clear all luminescence traps up to 500 °C, as is usual praxis for experiments investigating PTTL in pIRIR, as we aimed to avoid altering the feldspar crystal or feldspar charge-trap conditions by high temperatures.

The increase in the background signal with increasing temperature is also recorded by the normalization dose and the SAR procedure corrects for the increasing signal during a complete SAR measurement appropriately (cf. our response to Jakob Wallinga, lines 94–114 and Fig. R-JW 1). The background consists of several – likely three – components, as illustrated by the zoomed-in insets in **Fig. R-SR 2b** and **R-SR 2c**. We assume that a smaller fraction of this composite background signal could be due to “PTTL”. The major part(s) are likely due to charge building up from one measurement cycle to the next, which was not cleared by strong hot-bleaches or high temperature TL in our experiment compiled in Fig. 15 of the manuscript.

For the revised manuscript we will provide better resolved graphs for each sample in Fig. 15a–15c of the manuscript, following **Fig. R-SR 2**.

New experiment

As you probably expect a figure in the style of Riedesel & Duller (2022), we performed an additional experiment. But please note that the measurement conditions with TL up to 500 °C are not those of the sediment dating. Further, for PET-IRSL there is no "natural window" for monitoring potential "PTTL" as it exists for pIRIR and MET in between the individual IR-measurement steps. To monitor something like "PTTL", one has to change the energy regime drastically, by either switching off IR and/or temperature, to observe changes in the shape of TL peaks, like the ca 410 °C feldspar peak, which is regarded as the source of the IRSL signal (Murray et al., 2009). The experiment was conducted on reader LR03 (PULSE).

<u>Experimental setup</u>

For our new experiment, we used a previously bleached aliquot (3600 s solar simulator) of sample HDS-1776. First, the aliquot was "prepared" by an initial preheat TL at 320 °C for 60 s (isothermal TL) and a subsequent TL 25–500 °C with subsequent isothermal TL (ITL) at 500 °C for 60 s.

Afterwards, five different measurements were performed, repeating each one three times to check the signal's reproducibility. In each cycle, the aliquot was given a dose of ca 300 Gy (2500 s beta-irradiation time), before being preheated to 320 °C for 60 s. These two steps compare to steps (8) and (1) of the PET-IRSL SAR protocol in Fig. 2 of the manuscript. (Please note, that we will restructure Fig. 2 when revising the manuscript, as suggested by reviewer Jakob Wallinga.)

The following step was either **(1)** a TL step to 500 °C with subsequent ITL at 500 °C for 60 s; or **(2a)** a PET IRSL step for 120 s at 60 °C, completed by TL up to 500 °C with subsequent ITL at 500 °C for 60 s; or **(2b)** a PET IRSL step for 230 s up to PET_{max} 280 °C (like step (2) of the PET-IRSL SAR protocol in Fig. 2 of the manuscript), completed by TL up to 500 °C with subsequent ITL at 500 °C for 60 s; or **(3a)** a PET IRSL step for 120 s at 60 °C; afterwards a separate TL step 25–500 °C with subsequent ITL at 500 °C for 60 s; or **(3b)** a PET IRSL step for 230 s up to PET_{max} 280 °C (cf. step (2) of the PET-IRSL SAR protocol in Fig. 2 of the manuscript); afterwards a separate TL step 25–500 °C with subsequent ITL at 500 °C for 60 s.

For better comparison of TL- and PET-peak positions, the temperature ramp of the preheat, the TL to 500 °C and the PET-IRSL readout was always 2 K/s. This is somewhat different from the PET-IRSL SAR measurements of our study, in which only PET-IRSL with PET_{max} 280 °C was measured with a ramp of 2 K/s. 25 °C is the starting temperature of any TL or preheat TL ramp on the *lexsygresearch* readers and compares to the room-temperature in the luminescence laboratory.

Results of the experiment

High-temperature TL peak

The results are shown in **Fig. R-SR 3–Fig. R-SR 7**. The high-temperature TL peak (here after a preheat of 60 s at 320 °C; post-PHT_{320} $TL_{25–500}$), which is considered as the source of the IRSL signal, is situated at ca 390 °C (**Fig. R-SR 3**). The difference of ca 20 K to the TL-peak position as observed by Murray et al. (2009), is owed to a lower temperature ramp of 2 K/s in our experiment. As the signal curves are well reproducible, in the following presentations of **Fig. R-SR 4–R-SR 7**, only one of the three measured replicates is shown in each case.

Changes of the TL-peak after IR-readout

Changes of the shape of the 390 °C TL peak after IR stimulation are shown in **Fig. R-SR 4** and **Fig. R-SR 5.** In **Fig. R-SR 4** the PET-IRSL signal curves are continued as TL signal curves after IR-switch-off, once after 120 s IR at 60 °C (PET-$IRSL_{60}$$\rightarrow$$TL_{60–500}$), and once after reaching PET_{max} 280 °C (PET-$IRSL_{60-280}$$\rightarrow$$TL_{280–500}$), respectively. When IR is switched off at PET_{max} 280 °C (dark-red line), the PET-IRSL signal drops to ca 200 counts, before it rises from that elevated level again as a TL signal towards the 390 °C TL peak. The signal of the 390 °C TL peak decreases after previous IR stimulation, and broadens on the low-temperature side. Both changes augment with increasing duration of the PET-IRSL readout (here 120 s IR at 60 °C *vs* a complete PET-IRSL readout 60–280 °C for 230 s). These results conform to earlier observations after IR_{1st} and IR_{2nd}, e.g., by Murray et al. (2009).

In **Fig. R-SR 5** we contrast the TL signal 25–500 °C (plus 60 s ITL at 500 °C) after 60 s preheat at 320 °C (plus 60 s ITL at 320 °C) (post-PHT_{320} $TL_{25–500}$) with TL signals 25–500 °C measured after PET-$IRSL_{60}$ (post-PET-$IRSL_{60}$ $TL_{25–500}$) and PET-$IRSL_{60–280}$ (post-PET-$IRSL_{280}$ $TL_{25–500}$), respectively. This means that the PET-IRSL readout was not continued as a TL-readout as illustrated in **Fig. R-SR 4**, but a separate TL measurement step starting from room-temperature or 25 °C, respectively, was performed after

PET-IRSL readout. The IRSL-source TL-peak at 390 °C shows similar changes as in **Fig. R-SR 4**, but as the post-PET-IRSL$_{280}$ TL-signal is not “concatenated” to the PET-IRSL$_{60–280}$-signal, it is better discriminable/isolatable for further data analysis.

Deriving summed-up/accumulated “PTTL”

For further data processing, we subtracted the pre-PET-IRSL TL-signal (post-PHT$_{320}$ TL$_{25–500}$) from the post-PET-IRSL signals (post-PET-IRSL$_{60}$ TL$_{25–500}$; post-PET-IRSL$_{280}$ TL$_{25–500}$) to derive the signal that might be invoked by “PTTL” (**Fig. R-SR 6**). As the post-PHT$_{320}$ TL$_{25–500}$-signal is close to the machine background for most of the PET-IRSL readout time, there is hardly any difference between the gross and the net TL-signals. The post-PET-IRSL$_{60}$ TL-signal appears to be composed of two components, a first one up to ca 110 °C, and a second one > ca 110 °C. They might correspond to the first two components observed in **Fig. R-SR 2**.

The post-PET-IRSL$_{280}$ TL-signal is close to zero for most of the TL-measurement time, but it starts to rise ≥ ca 235 °C. As observed in **Fig. R-SR 5**, this rise corresponds to the rising flank of the 390 °C TL peak, which is (one of) the source(s) of the IRSL signal. As, however, all TL-traps up to 320 °C had been emptied by the preheat, and as the post-PHT$_{320}$ PET-IRSL readout was conducted up to PET$_{max}$ 280 °C, the TL signal observed in the range ca 235–280 °C could not recover during the PET-IRSL ramp up to 280 °C. The combined photon- and phonon-energy of a temperature of 280 °C and IR-stimulation would correspond to a temperature equivalent > 280 °C. This does not allow electrons occupy TL traps ≤ 280 °C. The increase in the TL signal ≥ ca 235 °C must therefore be owed to charge settling once IR and temperature have been switched off. It would have to be regarded as “PTTL” accumulating during the complete course of the PET-IRSL readout rather than “PTTL” that contributes to the PET-IRSL signal.

We then contrast the net TL-signals with the PET-IRSL signal in the range 60–280 °C (**Fig. R-SR 7**), expressing the net TL-signals after PET-IRSL readout at 60 °C and up to PET$_{max}$ 280 °C, respectively, in both cases as a fraction of the PET-IRSL signal that was read out up to PET$_{max}$ 280 °C (cf. dark-red line in **Fig. R-SR 4**; PET-IRSL$_{280}$ → TL$_{280-500}$ up to 280 °C).

The net post-PET-IRSL$_{60}$ TL$_{25–500}$-signal amounts to a constant fraction of ca 2.5 % of the PET-IRSL$_{280}$-signal up to ca. 230 °C. Thereafter it rises slightly to ca 4 %. It is difficult to imagine that TL traps exist along the whole temperature range contributing an almost uniform fraction to the PET-IRSL signal. We therefore assume that the signal originates from electron charge expelled from an unstable 100 °C-TL peak (cf. slightly increased values in **Fig. R-SR 7** in the range ca 90–130 °C), that filled during the cool-down to 25 °C prior to TL$_{25-500}$. Once stimulated by TL, the charge sems to provide a continuing signal, which may represent phosphorescence. We do not think that the signal is representative for the active PET-IRSL readout. But assuming a worse-case scenario, in which the continuous signal contribution represents true "PTTL", it would contribute ca. 2.5–4 % of the signal used for dating.

The fraction of the net post-PET-IRSL$_{280}$ TL$_{25-500}$-signal stays close to zero up to ca 235 °C before it rises to a maximum of up to ca 6 % at PET$_{max}$ 280 °C. This signal rise conforms to the third background component in **Fig. R-SR 2**. We consider it as plausible to recognize the three components of **Fig. R-SR 2** in **Fig. R-SR 7**. The main, but for this observation not relevant, difference between the experiments in **Fig. R-SR 2** and **Fig. R-SR-7**, are the TL readout temperature (320 °C *vs* 500 °C) and the number of measurement cycles of abbreviated PET-IRSL readouts (eleven *vs* two), which lead to an overall larger background increase in **Fig. R-SR 2** but provide a similar three-component structure.

Interpretation

While the fraction of post-PET-IRSL$_{60}$ TL$_{25-500}$ varies between ca 2–4 %, the fraction of post-PET-IRSL$_{280}$ TL$_{25-500}$ amounts to ca. 6 % at maximum of the PET-IRSL signal. Wang & Wintle (2013) assumed that charge from PTTL signals observed after IRSL-readout at low temperature (IR$_{1st}$), which could be incorporated in the high-temperature IR$_{2nd}$-signal, could show similar signal instability (fading) as IR$_{1st}$. However, this is not what PET IRSL does. Therefore, we can quantify a pseudo-PTTL contribution, but our data do not support an effect with practical relevance.

We did not perform any stability tests on the respective net TL signals. But as the fraction of net post-PET-IRSL TL$_{25-500}$ increases in the high-temperature range, as shown in **Fig. R-SR 7**, one would expect increasing g-values with increasing readout time. Yet, a contrary trend is observed, as illustrated in Fig. 10a of our manuscript. g-values decrease from ca 2 % per decade to ca 1 % per decade in the range ca 120–280 °C temperature assisting the IR-readout.

Summary

We do not have evidence that unstable "PTTL" is an issue for PET-IRSL SAR beyond the statements conceded in lines 626–634 of our manuscript. We do not have any indication of poor dose recovery on the samples studied, which could point to a problem with instable "PTTL". Background signals increasing from one PET-IRSL SAR cycle to the next, are recorded by the normalization-dose signal and they are appropriately corrected by the SAR procedure for Lx/Tx calculation (cf. also Fig. R-JW1 and Fig. R-JW 2 in our response to reviewer Jakob Wallinga).

To address the raised concerns, we had performed an experiment to monitor “PTTL” in the course of pIRIR and MET procedures. However, the experiment does not conform to the stimulation conditions during PET-IRSL, only after PET-IRSL. Therefore, only a summed-up/accumulated “PTTL” signal can be monitored, but no “PTTL” reflecting each individual point in time during PET-IRSL readout. At present, we would not know how to monitor “PTTL” and its contribution to the PET-IRSL signal during active PET-IRSL “switched-on”. Regardless, the results of the summed-up post-PET-IRSL signal of what might be regarded as “PTTL” (cf. **Figs R-SR 3–R-SR 7**) and which would overestimate its potential contribution to the PET-IRSL signal at any point in time of the 230 s lasting readout anyway, do not give indication of a significant problem with “PTTL”. For this reason, we cannot support the reviewer’s hypothesis that “PTTL” is an issue for PET and we do not wish to allocate more space to the issue of potential “electron recycling”, as we prefer to name it for reasons detailed above, in our manuscript.

Many figures are presented to illustrate the data, however, most of them look similar and while they of course present the data, the key message each figure is supposed to convey is not directly visible. Regarding the figures I would like to offer the following suggestions:

- Please display all doses in Gy and not in s.
    - Thank you for spotting this. We will address doses in Gy and irradiation times in s.
- Currently there is text pasted below each subfigure. I would suggest adding this labelling into each subfigure (text type and sample: for example “Dose recovery test HDS-1849 (PHT 320 °C)”)
    - We will look at this and see what can be changed.
- I give further comments regarding individual figures below.
    - We will address each comment below.

Suggestion ofrestructuring:

Overall, I would suggest splitting the paper into two or restructuring it significantly:

(1) One paper or the first part of a paper presenting the concept of the protocol, with an explanation of the physics behind it and the potential influence of PT-TL on the measured signal. It could then also include a sketch-like summary figure which presents the types

of De plateaus expected. This would be particularly helpful to be able to follow the discussion in section 7.1.4. Bateman et al. (2003) present a conceptional, sketch-like figure (their Fig. 1) to illustrate De distributions they would expect for certain bleaching processes. I would envisage something like for the expected De plateaus. It would also be interesting to add a figure showing the variation in peak position.

Again, we do not consider it as reasonable to split the paper (for details see our comments above). Nor do we think that PTTL, or rather more general, electron recycling/retrapping (see above), is a major issue for the studied samples. We addressed the issue explicitly with an experiment (see Fig. 15 and **Figs. R-SR 2–R-SR 6**). We will mention that issue in the introduction as a possible challenge for pIRIR and MET and also cite Riedesel and Duller (2022).

We will prepare a conceptual sketch for the graphical summary.

(2) A second paper or second part of a paper could then evaluate various quality control parameters, such as DRT performance or size of residual dose. Here a few key figures could be shown to support the data. I don't think it is necessary to show 18 figures in the main text. Instead of showing many similar figures, summary figures could be presented.

Some hints on which figures you consider as redundant would be helpful.

We are developing a new type of "spaghetti-plot"-like graph to present De-value estimates from PET-IRSL SAR measurements (cf. our response to reviewer Jakob Wallinga, lines 316–324, Fig. R-JW 6).

For these two papers or sections of a paper it is important that they make it clear, why this protocol is an important contribution to the existing suit of measurement protocols. Why should a user deviate from their routine procedures to test this new protocol? What are the advantages?

We believe that we abundantly addressed the novelty of our approach without diminishing existing protocols.

In many instances throughout the text, the authors use abbreviations or deviate in their description of their experiments from common terminology. This makes it hard to follow the text, as it requires constantly remembering how certain tests and procedures are termed in the current paper. For example: an NRM should be called test dose, a zero-LAB DRT is a residual dose measurement.

As for the term "test dose", we prefer to call it a normalization dose, as nothing is tested but normalized. By definition it is used for intra-aliquot normalization for the SAR procedure (as opposed to former inter-aliquot normalization procedures for MAR or

MAA, e.g., by short-shine normalization), mathematically expressed as Lx/Tx.

A residual or remnant dose, we would associate with a partially bleached sample. If, however, a thoroughly bleached sample (here usually 1 h solar bleaching in the lexsyg reader and once even 12 h bleaching "super-bleach" experiment), gives a dose after application of a SAR protocol, this is a product of the way the sample is treated in that measurement procedure. Probably all SAR parameters, including the number and size of the regeneration and normalization dose points, have an effect on the size of the recovered dose despite sufficient bleaching and no laboratory dosing. The strongest impact might come from the preheat temperature (and duration), as suggested by our preheat tests with bleached but not dosed samples (cf. also lines 715–724 of our manuscript). Inconsistently, we also use "residual dose", e.g., in line 720, without putting it in quotation marks, as we did not want to question also that term. But we decided deliberately for dose-recovery test (DRT) with a zero-laboratory dose, as we consider the recovered dose as a measurement artifact. This is also supported by our dose-recovery test added to our reply to reviewer Jakob Wallinga (cf. **Fig. R-JW 3** and **Fig. R-JW 4**).

We will omit some of them, like, e.g., PBL tests, and include a list of abbreviations in the appendix.

Further comments:

Throughout the text: Please present all doses in Gy and not in s. I can see that you use both in the figures (two axes) and the text, but I don't see the necessity in this. It adds to the complexity of both, the text and the figures.

We will use dose in [Gy] and irradiation time in [s] as displayed and are convinced that the reader will overlook the complexity. However, we will change the axis labeling to "irradiation time [s]" instead of "De [s], which was not correct.

Abbreviations: Many abbreviations are used throughout the text. I am particularly confused by the use of LAB, NRM and PBL. NRM is usually referred to as test dose in the literature. I would ask the authors to keep in line with the general use of terms (see also the naming in Murray and Wintle 2000, their Table 1).

We will use laboratory dose, normalization dose and partial bleaching (test) instead.

Lines 31-40: This is a very long and unnecessary introduction. Is there the possibility to shorten this and target the first paragraph of the introduction to convey the necessity of this study?

We wanted to address readers, who are not familiar with feldspar luminescence dating and introduce relevant abbreviations like, De, SAR, IR. But we will change the introduction as requested.

Lines 57-58: I disagree in calling band tail hopping “temperature-dependent fading”.

Here we may refer to Guérin & Visocekas (2015; http://dx.doi.org/10.1016/j.radmeas.2015.05.003) “the temperature dependent or “frozen” part of the fading is relevant to a different mechanism, which is “hopping”, already proposed in the mid-sixties, which preserves the experimental logarithmic fading decreasing law”.

Line 62: “Firstly, near-neighbor electron…” There is a preheat prior to the low temperature IRSL step, which also results in electron hole-recombination.

Sure. But here we explain the $pIR_{1st}IR_{2nd}$ protocol and why there are two consecutive IR-steps. Prior to laboratory preheat there is storage in the sediment archive which also allows for near-neighbor electron recombination. We consider it legitimate to abstract from these distracting issues.

Lines 95-96: I would recommend citing Jain and Ankjærgaard (2011) instead of Riedesel et al. (2019).

We will include Jain and Ankjærgaard (2011) in line 59 and cite Jain and Ankjærgaard (2011) in lines 95–96.

Lines 99-128/Section 2.1 (sample material): I would suggest moving the information into a table. Please explain why these samples were chosen, although they don’t have any independent age control available.

These samples do have age constraints. Please refer to our response to reviewer Jakob Wallinga, lines 40–52.

As suggested, we will move the information on the samples into a table, if this saves space.

Line 264: Intensity rather than count data?

Ok.

Lines 396-399: This is very long and complex sentence.

We will split it up.

Line 412: Referring to fading as luminescence decay is rather uncommon.

We will change it to "signal loss".

Line 425: Please change to: All g-values are in the range of 0.5-4 % per decade, indicating a signal loss over time.

Ok.

Lines 441-455/Section 6.1: This section could be cut. It comes very suddenly and only conveys the information that the samples exhibit a blue emission. This could have been added to the sample description.

We consider it as relevant to present the results of our spectral measurements to assess the validity of our measurements and measurement setup for a new approach. Besides, so far, to our knowledge nobody has presented a PET IRSL SAR spectrum before. As first the PET IRSL SAR approach has to be introduced before a PET IRSL spectrum can be presented, it does not make logically sense to show it earlier because the results should not come before the introduction of the general concept.

Line 555: What is meant by "late background"? I can see from Figure 3 that late background describes the signal measured after the LEDs have been turned off, but that is the instrumental background and not the signal background.

We will reword our sentence to "These background signals show a slight steady increase, …".

In contrast to the "machine background" at constant 60 °C (e.g., in Fig. 3), these background signals with an increasing TL temperature up to PET_{max} conform more to the signals of the zero-dose regeneration dose points (and may be equivalent to them).

Line 593: Higher instead of stronger.

Ok.

Lines 600-614: A figure just showing the peak positions would be useful.

We do not understand that comment.

(1) Three different feldspar specimens with different potassium contents show that in each case after a preheat temperature of 320 °C the peak positions shift from ca 188 °C, to ca 204–212 °C and ca 220 °C, respectively, with increasing potassium content.

(2) We investigated whether there is a systematic shift of the PET-peak after different preheat temperatures and show this systematic shift in Figure 14.

Or did you want to suggest to remove Figure 14?

Line 675 and elsewhere: Please do not call them zero dose tests. What is measured is the residual dose remaining in the samples after laboratory bleaching.

Technically our expression is correct and hence we disagree (see our comments above). The recovered dose is a measurement product/artifact of the sample treatment during the SAR protocol, most likely dependent on the strength of the preheating, which transfers charge into the IRSL trap. If you consider the De-value curves to be without merit, they might at least serve for illustrating this issue nicely.

Lines 767-774: I cannot follow the reasoning. G-values vary significant between samples.

We do not understand that comment. Why should g-values from different samples from different regions not vary significantly, especially if they have been preheated differently, either up to only 200 °C or up to 280 °C, respectively? But the values compare, e.g., for HDS-1849 from the Nebraska Sandhills with IR$_{60}$ and pIR$_{170}$ g-values measured by Buckland et al. (2019) for another sample from that area.

Line 781: I would say the g-values decrease with De integral?

We will rephrase that sentence (see also comment by reviewer Jakob Wallinga).

Line 825/section 7.1.4: I don’t understand what the authors mean by partial bleaching test and how they were conducted. I understand this as investigation into the shape of De plateaus during a De determination using the proposed protocol.

The procedure used to conduct the partial bleaching (PBL) tests is described in section 6.2. Therein, we also present the results of the partial bleaching (PBL) tests. We tried to strictly separate “results” in section 6 from the advanced “discussion” in section 7. In lines 827–836 we explain that now we want to compare the results of De-value curves from samples with a natural palaeodose to those of the experiments gained by the partial bleaching (PBL) tests. Or rather: Following the partial bleaching (PBL) tests, we are now able to recognize similarities with those from the natural samples. We acknowledge that fading might also have changed the natural De-value curves, an issue which we have not investigated in the present study. Please see our response to reviewer Jakob Wallinga, lines 262 ff. and Fig. R-JW 5).

Fig. 1: Why is there a secondary y-axis displaying a change in power density? I thought the density was set at e.g. 60 mW/cm2 and not changed during the measurement? I guess the green box is inserted to show the power density? However, with the actual scale on the secondary y-axis it seems like the power density should be changed during the measurement.

As denoted in the figure caption, the graphical representation follows that of the LexStudio2 Sequence Editor (Richter and Kumar, 2024); in higher resolution and with a unit error corrected. The upper boundary of the green box shows the chosen power density, which is the same from 60–290 s measurement time, i.e., here at 60 mW/cm². It is correct that we did not change the power density during a measurement but only the temperature.

Fig. 2: Here especially: Please report all doses in Gy and not in s. You could then also remove most footnotes to make the figure more readable.

As detailed elsewhere, we will now display doses in Gy and irradiation time in s.

Fig. 3: In the legend please use lines instead of points.

The graph is the graphical output of the R package “Luminescence”, which uses points for the colour code: “*Graph as exported by the R package “Luminescence” function “analyse_SAR.CWOSL” (slightly modified).*” We want to keep it consistent.

Fig. 5: Replace “expected value” by “given dose”, you could even add a little label to the red line in the figure. This would also help the reader to immediately spot that this data was gathered by performing a dose recovery test.

We will replace “expected value” by “given dose”.

We consider adding labels to the lines as inappropriate as this would overload the figures. There might be space for a label in Figure 5, but not in other figures. Would the reader then miss the label? We prefer to be consistent.

Fig. 5 and Fig. 6: These figures are referred to asl NRM tests. I thought they show the results from a dose recovery test and the NRM is the normalisation dose, which is usually called a test dose. Or am I mistaken?

This is correct. When testing different normalization doses (NRM), we call these tests NRM tests, as detailed in lines 302–304: “*… we performed DRTs with varying NRM (NRM tests). These tests represent combined normalization-dose and dose-recovery tests.*”

Fig. 7: In the caption you refer to the residual dose tests as zero-LAB DRT. Please refer to them as residual dose measurements/test. There is no dose to recover.

We agree that zero-LAB DRT is not a good abbreviation and will omit it. For why we do not use “residual dose” please see our comments above.

Fig. 9: The orange star is missing in b.

The orange star is not missing. As the sample was preheated only to 200 °C, and as PET_{max} was only 170 °C (see top x-axis) and PET-IRSL SAR was read out with a ramp of 1 K/s, a PET-peak has not developed (see, e.g., lines 206–208 and 622–624 of the manuscript).

Fig. 16: The overall message these figures should convey is unclear to me. Is it also necessary to show all Figures 16, 17 and 18?

As stated in section 7.2 in lines 863–868, the suites of De-value curves reflect different degrees of bleaching and therefore different palaeo-environments ranging from poorly bleached alluvium to moderately bleached reworked loess to well-bleached dune sand. We found it absolutely thrilling that the De-value curves reflect/reveal the bleaching conditions so obviously.

Unfortunately, at present, there do not exist any smart graphs to present the De-value results of PET-IRSL SAR measurements, but we are up for developing them. One example, for sample HDS-1776, is given in the response to reviewer Jakob Wallinga in Fig. R-JW 6. But we are open for further suggestions.

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- Murray, A.S., Wintle, A.G. Luminescence dating of quartz using an improved single-aliquot regenerative-dose protocol Radiation measurements 32, 57-73, 2000.
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- Wang, X. L. and Wintle, A. G. Investigating the contribution of recuperated TL to post-IR IRSL signals in perthitic feldspar. Radiation Measurements, 49: 82–87, 2013.

Thank you very much for giving us notice of these publications.

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Ankjærgaard, C. and Jain, M.: Optically stimulated phosphorescence in quartz over the millisecond to second time scale: insights into the role of shallow traps in delaying luminescent recombination, Journal of Physics D: Applied Physics, 43, 255502, https://doi.org/10.1088/0022-3727/43/25/255502, 2010.

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Murray, A. S., Buylaert, J. P., Thomsen, K. J., and Jain, M.: The effect of preheating on the IRSL signal from feldspar, Radiation Measurements, 44, 554–559, https://doi.org/10.1016/j.radmeas.2009.02.004, 2009.

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Riedesel, S. and Duller, G., 2022. Measuring photo-transferred thermoluminescence from feldspars in post-IR IRSL procedures using a new user defined command for the Risø

TL/OSL reader. Ancient TL 40(1): 1-11. https://doi.org/10.26034/la.atl.2022.557, 2022.

Thomsen, K. J., Murray, A. S., Jain, M., and Bøtter-Jensen, L.: Laboratory fading rates of various luminescence signals from feldspar-rich sediment extracts, Radiation Measurements, 43, 1474–1486, https://doi.org/10.1016/j.radmeas.2008.06.002, 2008.

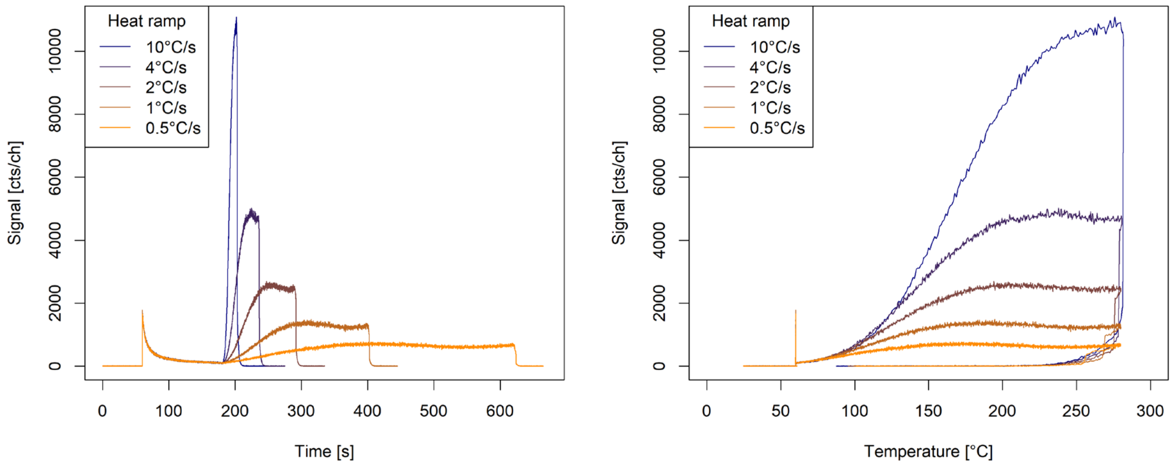
Kind regards,
Svenja Riedesel
Cologne, 9th February 2026

With kind regards

Annette Kadereit, Mariana Sontag-Gonzáles, Sebastian Kreutzer, Marco Colombo, Christoph Schmidt and Paul R. Hanson

5th March 2026

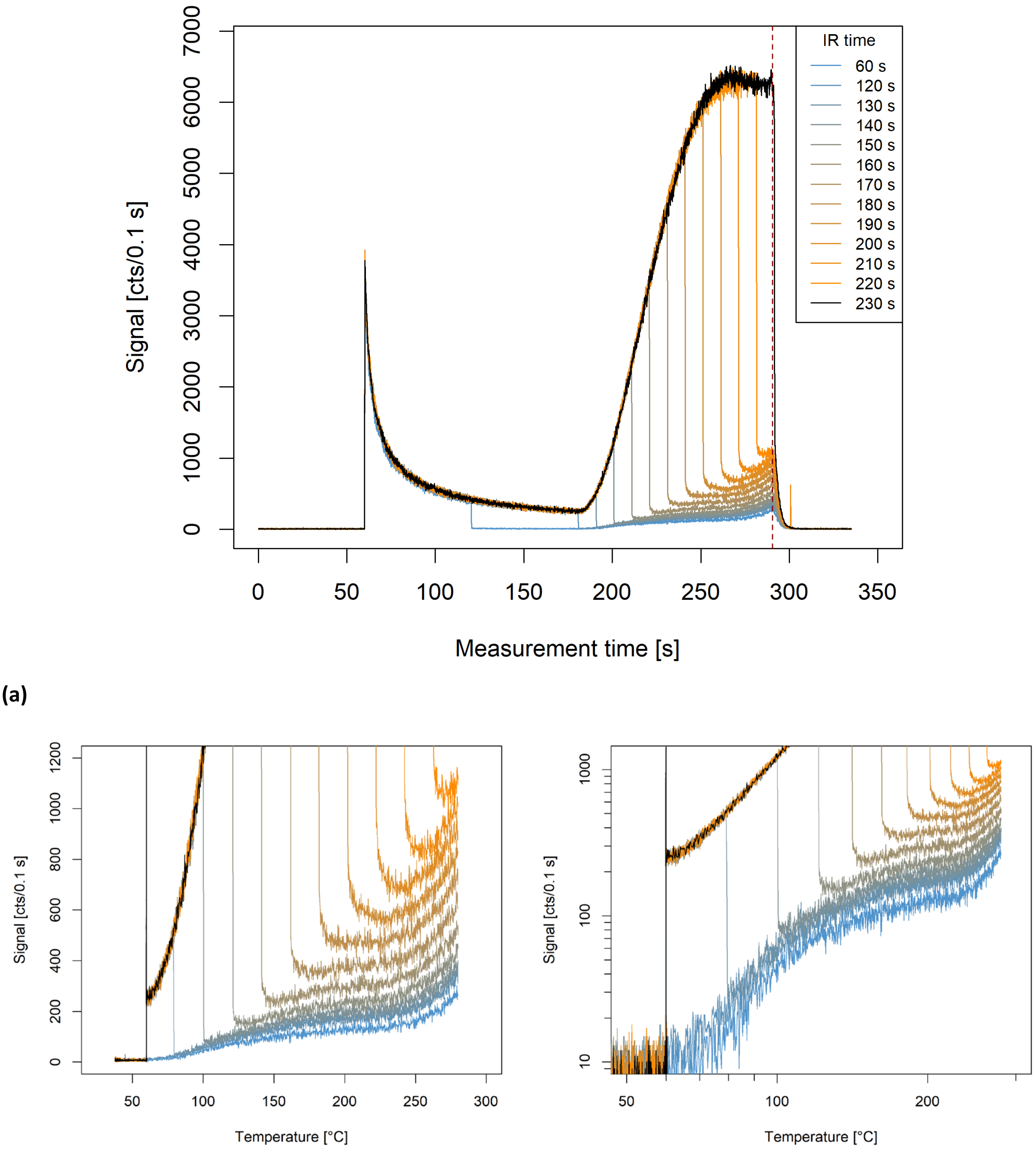
Response to the review of Svenja Riedesel of the manuscript ‘A progressively elevated temperature (PET) IRSL SAR procedure – first experiments and results’ by Kadereit et al.



(a) **(b)**

Fig. R-SR 1 PET-peak position shifting with varying temperature ramp. PET-IRSL signal curves plotted against **(a)** readout time, **(b)** temperature assisting the IR-readout. PET-peak positions at ca 237 °C for heat ramp 4 K/s; ca 199 °C for heat ramp 2 K; ca 185 °C for heat ramp 1 K/s; ca 170 °C for heat ramp 0.5 s. The peak for heat ramp 10 K is cropped at 280 °C by the high heating rate. The somewhat awkward backward course of the signal curves in **(b)** correspond to the final PET-IRSL decline in **(a)** (cf. also Fig. 3 of the manuscript). The curve ramped with 2 K/s was used in our study for PET-IRSL readout up to PET_{max} 280 °C (sample HDS-1776). The curve ramped with 1 K/s was used in our study for PET-IRSL readout up to PET_{max} 170 °C (samples HDS-1827 and HDS-1849). In both cases, IR was switched-off after 290 s measurement time or 230 s IR-readout time, respectively (cf. position of the final PET-IRSL decline of the dark-brown signal curve at 290 s measurement time). Therefore, no PET-peak was observed for measurements with the 1 K/s ramp (light-brown signal curve).

Response to the review of Svenja Riedesel of the manuscript ‘A progressively elevated temperature (PET) IRSL SAR procedure – first experiments and results’ by Kadereit et al.



(a)

(b)

(c)

Fig. R-SR 2 Variable durations of PET-IRSL readout for samples HDS-1776. After IR-switch-off the readout is continued as TL. The figure is basically a reproduction of Fig. 15c of the manuscript, but omitting the first PET-IRSL signal curve up to PET_{max}, which due to a considerable sensitivity change after the first measurement cycle deviated from the course of all the other signal curves. **(b)** and **(c)** are magnified versions of the background signal displayed **(a)** on linear scales and **(b)** logarithmic scales.

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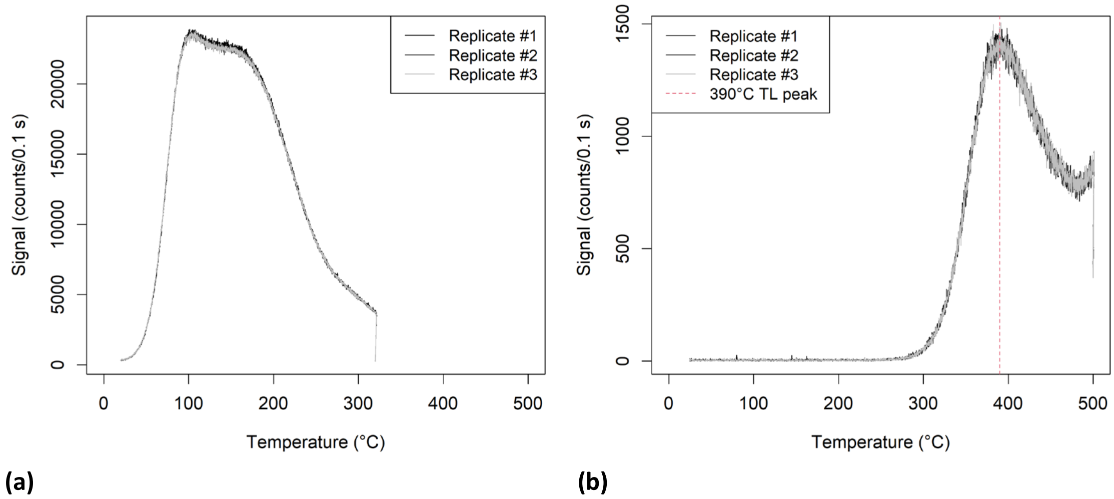

Fig. R-SR 3 Contrasting TL 25–500 °C after preheating at 320 °C for 60 s (post-PHT_{320} $TL_{25–500}$) with PET-IRSL at 60 °C for 120 s continued as TL 60–500 °C (plus 60 s ITL at 500 °C) (PET-$IRSL_{60}$ → $TL_{60–500}$) and PET-IRSL to PET_{max} 280 °C continued as TL 280–500 °C (plus 60 s ITL at 500 °C). (PET-$IRSL_{280}$ → $TL_{280–500}$). Replicates **(a)** PHT_{320}, **(b)** post-PHT320 $TL_{25–500}$.

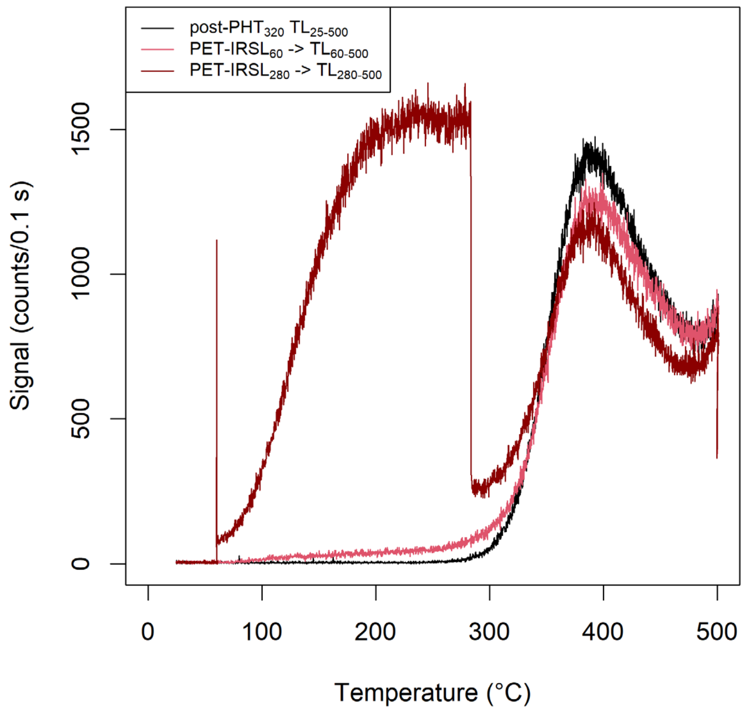

Fig. R-SR 4 Contrasting TL 25–500 °C after preheating at 320 °C for 60 s (post-PHT_{320} $TL_{25–500}$) with PET-IRSL at 60 °C for 120 s continued as TL 60–500 °C (plus 60 s ITL at 500 °C) (PET-$IRSL_{60}$ → $TL_{60–500}$) and PET-IRSL to PET_{max} 280 °C continued as TL 280–500 °C (plus 60 s ITL at 500 °C) (PET-$IRSL_{280}$ → $TL_{280–500}$). The upward spike-like signal course at 60 °C (only dark-red and light-red lines) represents 120 s IRSL at 60 °C of the PET-IRSL readout. The downward spike-like signal course at 500 °C represents ITL 500 °C.

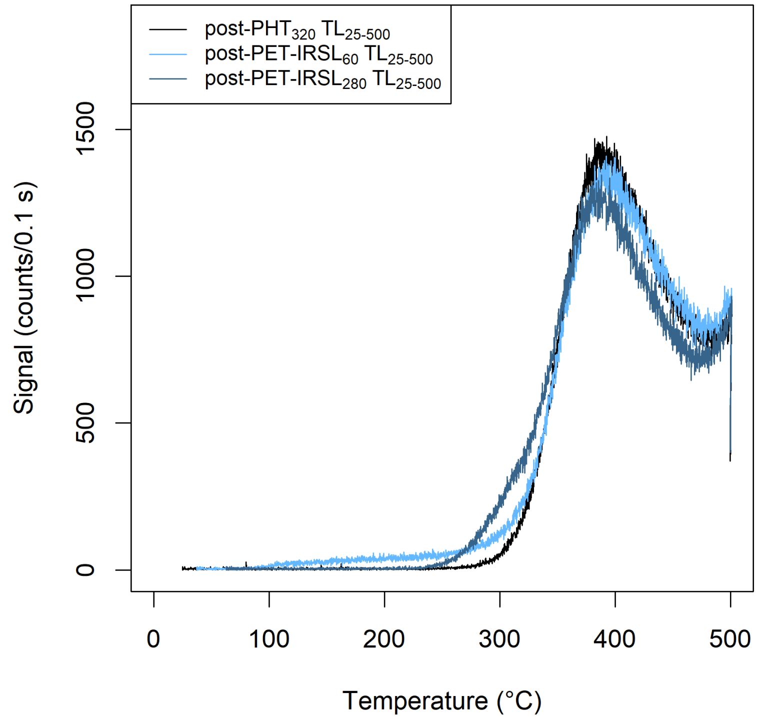

Fig. R-SR 5 Contrasting TL 25–500 °C after preheating at 320 °C for 60 s (post-PHT$_{320}$ TL$_{25–500}$) with TL 25–500 °C (plus 60 s ITL at 500 °C) after PET-IRSL at 60 °C for 120 s (post-PET-IRSL$_{60}$ TL$_{25–500}$) and after PET-IRSL to PET$_{max}$ 280 °C (post-PET-IRSL$_{280}$ TL$_{25–500}$).

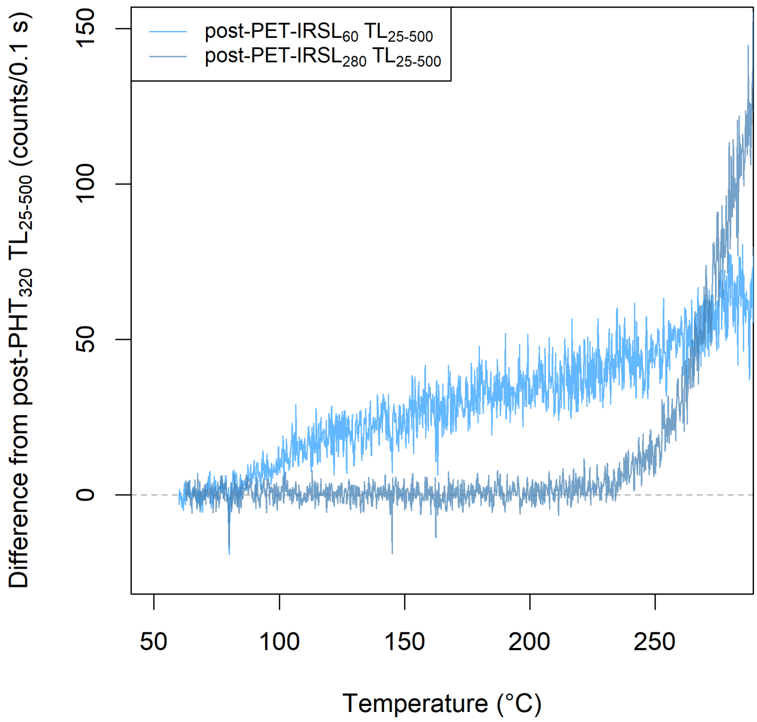

Fig. R-SR 6 Net signals of TL 25–500 °C (plus 60 s ITL at 500 °C) after PET-IRSL at 60 °C for 120 s (post-PET-IRSL$_{60}$ TL$_{25–500}$) and after PET-IRSL to PET$_{max}$ 280 °C (post-PET-IRSL$_{280}$ TL$_{25–500}$), each after subtraction of TL 25–500 °C after preheating at 320 °C for 60 s (post-PHT$_{320}$ TL$_{25–500}$).

Response to the review of Svenja Riedesel of the manuscript 'A progressively elevated temperature (PET) IRSL SAR procedure – first experiments and results' by Kadereit et al.

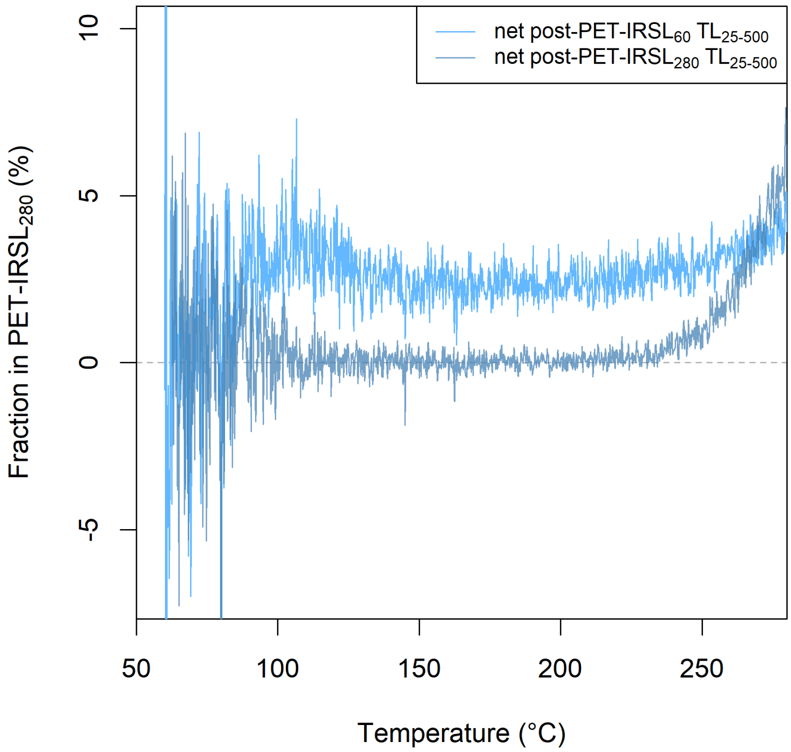

Fig. R-SR 7 Net post-PET-IRSL$_{60}$ TL$_{25–500}$ signal (light blue) and net post-PET-IRSL$_{280}$ TL$_{25–500}$ signal illustrated as a fraction of the PET-IRSL signal 60–280 °C.