Interactive comment on* "Surface paleothermometry using low temperature thermoluminescence of feldspar" *by Rabiul H. Biswas et al.

Anonymous Referee #2

This manuscript proposes a new method to study/reconstruct paleo-temperature using TL of feldspar from rock surface. Reconstructing paleo-temperature is an important topic in climatic change study, so the attempt of this study is important and worth for publication. However, there are some technical issues that are not settled well, which prevents convincing me that this is achievable. Here I summarise my major concerns.

1) Kinetic model: the authors considered three processes in their model, including dosing (growth), thermal decay and athermal decay (fading), which are represented by different terms in their equation 1. They then estimated the parameters based on TL measurements in different ways. For both the growth curve parameters (D0 and a) and fading parameters (rho'), my understanding is that they were based on the signals from different integrals of the TL glow curve (e.g., 200 – 250 C in 10C interval). However, for the thermal decay parameters (E, s and b), they used a Tm-Tstop method, in which the signal from each temperature interval is obtained from subtracting consecutive fractional glow curve. That means, the signals they used to estimate present-day charge population ($\bar{n}$), growth curve and fading are based on simple integral of TL signals at different intervals, which obviously are a mix of signals from a range of trapping energy levels, but the thermal kinetic parameters are based on single (or narrow-range) trapping energy levels. That says, the authors did not separate the TL signals for constructing their model using a similar way (Tm-Tstop method) that they did for estimating the thermal decay parameters. This is problematic, as the combination of different trapping levels are not linear, so their model (equation 1) simply becomes invalid when the signals being analysed are associated with a range of trapping levels are analysed. One need to makes sure that the different parameters in Equation 1 are all obtained from the same signal associated to a single or narrow-range energy level. I am not use if this can be achieved as the combination of MAR protocol (and fading test) and Tm-Tstop would be very difficult to achieve.

We do not assume that the thermal kinetic parameters are estimated on single trapping energy levels, as the number of traps are not known. In the absence of any information of the number of peaks it difficult to deconvolve the TL glow curve. So instead we use a 10 °C window and treat them as different 'thermometers', rather than single trapping energy levels. (Note that the rationale behind choosing a 10 °C window is the uncertainty of sample temperature during TL measurement in the laboratory.)

We constrain the growth parameters (D_0 and a) and fading parameter (ρ') for these different 10 °C temperature windows. To evaluate the thermal decay parameters (E, s, b) of each 'thermometer', we need to know the distribution of these parameters along the TL glow curve and find the most probable decay parameters of different thermometers from the distributions. Pagonis et al. (2014) demonstrated that the distribution of thermal decay parameters along a TL glow curve can be obtained using the *Tm-Tstop* method, subtracting the fractional glow curve and fitting with a TL equation. Biswas et al. (2018)

adopted this method for thermochronology and showed it could be used to extrapolate laboratory data to geological timescales, as a proof-of-concept.

The subtraction of consecutive fractional TL glow curves, obtained through the *Tm-Tstop* method, and fitting of the subpeak provides the characteristics (kinetic parameters) at that TL temperature. The integrated TL signal is a mix of different signals but the major contribution comes from the subpeak at that temperature range and a small fraction from the subpeaks either side of the temperature. Biswas et al. (2018) demonstrated (Fig. S4 in their supplement) that the life time distribution along the TL glow curve (present method) exactly match with classical lifetimes of the main dosimetric peaks of feldspar (as summarised in Aitken's TL book, 1985, Table E.1). This ensures the estimation of kinetic parameters of integrated TL signals from a distribution holds well. Moreover, this method was tested on well known (temperature) KTB borehole samples and predicted the sample temperature accurately (Supplement Fig. S6 of Biswas et al. 2018).

2) The authors simply assuming feldspar consist a continuous trapping energy level. However, it has been commonly accepted that band-tail states play important role in the luminescence process (including TL and IRSL) in feldspar (Poolton et al., 2002). I do not see any reason to discard the band-tail states from their model.

There is ample evidence from the literature suggesting that feldspar consists of continuous trapping energy levels (e.g. Grün and Packman, 1994; Pagonis et al., 2014; Strickertsson, 1985; Biswas et al. 2018).

We do not discard band-tail states in feldspar. Feldspar luminescence is complicated and several models that describe thermal detrapping processes in feldspar have been proposed (see summary in Gurlanik et al., 2015, RM, 81, 224-231). We have added this reference in the manuscript. Moreover, the TL of feldspar is a more diffusive process than IRSL which excites a resonant energy level below conduction band from which electrons either tunnel to recombination centers or hop through the band-tail states and recombine. The general order kinetic model is the simplest model available and the power term b (>1) accounts for the non-linearity that arises due to the presence of the band tail states (restricted mobility) or other complex process. This model has been used for a long time and most recently was used by Guralnik et al. (2015) for IRSL of feldspar and Biswas et al. (2018) for TL of feldspar.

3) How sensitive is the model to dose rate ($\dot{D}$)? The dose rate would play an important role in filling the traps during natural process, so I would expect that it will somehow influence the model results. Unfortunately, the dosimetry of the samples is poorly described. How did the author estimate the dose rate of K-feldspar? The author appears to simply crush the rock and select 150 –250 um grain size range. What are the original grain sizes of the K-feldspar minerals in the rock? Did the authors make any rock slide to investigate this? This is critical as there are a large contribution of internal dose rates for Kfeldspar.

The dose rate values were taken from Lehman et al. (2019b). It is mentioned in title of Table 1. They mention in their paper (supplement) as follows: "Environmental dose rates were calculated using DRAC

online calculator (Durcan et al., 2015), assuming a grain size between 750 and 1000 μm and water content of 2%". The grain sizes were estimated through thin section analysis.

4) The authors applied the NCF method to overcome sensitivity change issue for TL measurements. They do realise the limitation of this method as it is based on extrapolation of the NCF values from low temperature to high temperature region, which is unreliable. Although the authors tested the effect of initial sensitivity changes on the modelling results and found very little changes for their sample, it does not guarantee that this applies to other samples and situations. The reason that the sensitivity changes did not affect the results is simply because that their samples are young and the growth of signal still lies on the linear part of growth curve, so any systematic changes in the sensitivity result in a proportional changes of different signal integrals. For older samples or high-De samples, however, this may result in non-proportional changes among different signal integrals (because the different D0 values for different integrals), and, hence, different model results in paleo-temperature. This potential problems should be appropriately acknowledged as at present it gives false impression that the initial sensitivity change does not matter.

We agree and have included appropriate caveats. Sensitivity change is a limitation for TL measurements and as such we carefully screened the data. As for all TL measurements this will need to be done in further studies but as this is well known we do not emphasise the point here, but rather give a detailed account of how we addressed this challenge.

To circumvent the initial sensitivity change the NCF was introduced (Singhvi et al. 2011) as shown in Fig. 8a. The limitation of this method is that the NCF can only be measured for the lower temperature part of the TL signal (<200 °C) where our region of interest (ROI) is 210-250 °C TL. In the absence of a strong correlation (dose response) between low temperature TL and the ROI, we extrapolated. As the reviewer notes, this approach is appropriate for our sample suite.

Minor comments:

5) Line 21: Credits should be given to Li and Li (Li and Li, 2012) who firstly proposed the idea of multiple-thermometers using TL (although not implemented in their study), and they also first introduced the rate equation to investigate the effect of single growth and saturation on OSLthermochronology.

Reference added.

6) Figure 1: The authors should at least provide some typical TL glow curves for their samples before showing the kinetic results.

Amended.

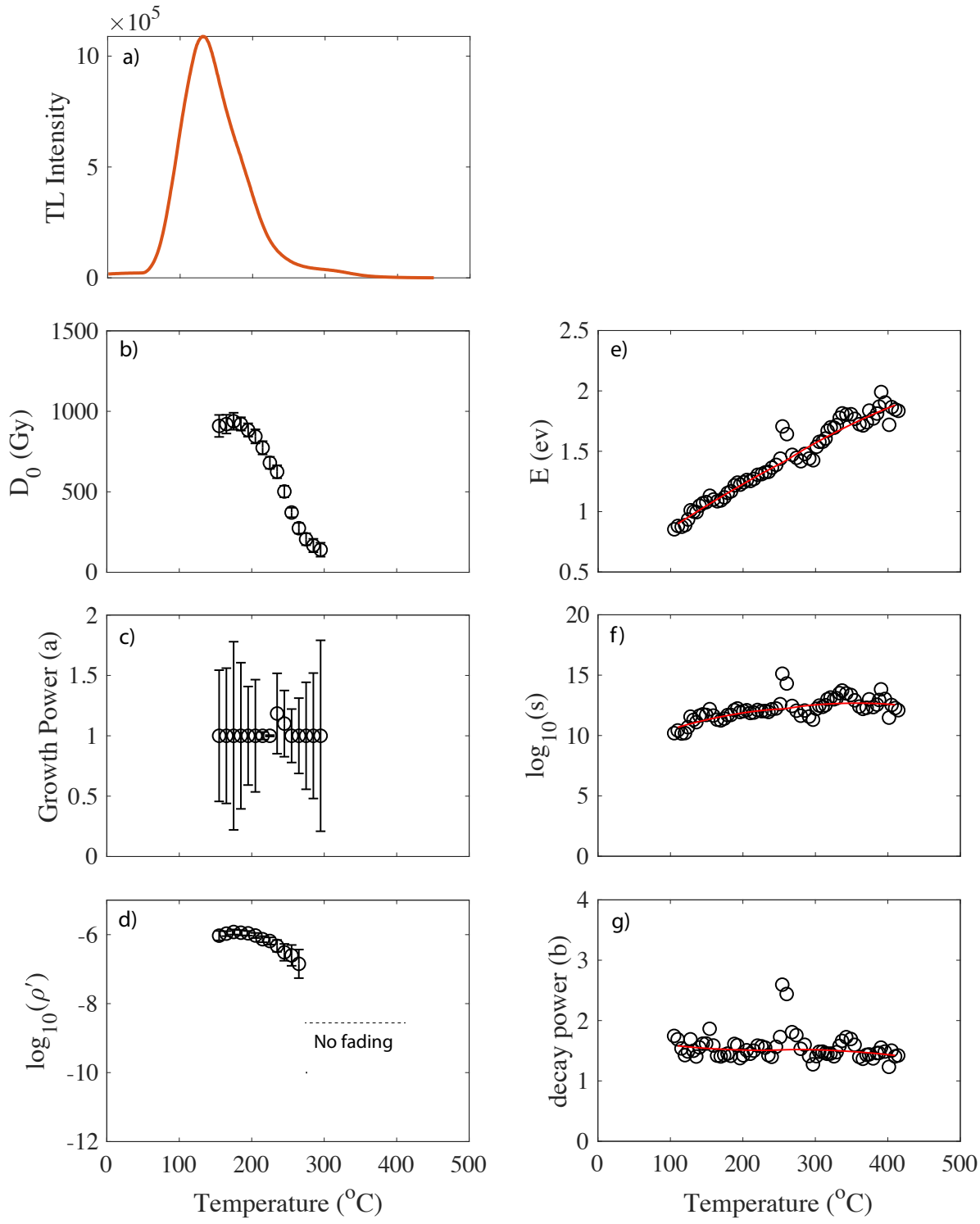

Figure 1: Laboratory TL glow curve (a) and inferred TL kinetic parameters (b-g) along the TL glow curve of sample MBTP9. The method used to constrain these parameters in the laboratory is followed by Biswas et al. (2018) and described in supplementary material S1.

7) Figure 1c: The fading parameter (rho') shows systematic change as a function of temperature up to 280C, but it suddenly become 'no fading'. This is surprising. The different integral signals represent a continuous mixture of signals of different athermal features, why one can obtain a sudden change in the fading? Is it because that the fading rate has large uncertainty range consistent with zero fading? In this case, it would be problematic to say 'no fading', as statistically it is also like to be 'fading'.

Beyond 270 °C, the fading parameter (ρ') is less than 10^{-10} with large uncertainty and is considered to be a non-fading signal. Even a fading curve with $\rho'<10^{-7}$ looks parallel to the time axis.

8) Figure 8b: Why not plot the results for other temperature range (e.g., 120 – 150 C)?

The objective is to see whether NCF_{100} is dose dependent or not.

9) Figure 8c: what are the errors for the NCF at high temperature range (200 – 250C)? Have you incorporated the NFC errors into the final results?

The errors for the NCF at the high temperature range (200 – 250 °C) are stated in Table 1. We did not incorporate NFC errors into the final results.

10) Table 1: Why there are no errors for E, s and b? Why an arbitrary error of 5% is assumed, rather than their actual analytical errors?

See Fig. 1. The distribution of E, s and b are a bit scattered and we spline fit the trend. So we did not calculate the regression of the trend and rather assumed an working error of 5%.

11) Figure S3: why there is only one natural point but 3 regenerative points for each thermometer? Did the author just measure one aliquot for natural?

The natural point is the average of three discs.

References

Guralnik, B., Li, B., Jain, M., Chen, R., Paris, R.B., Murray, A.S., Li, S.-H., Pagonis, V., Valla, P.G., Herman, F., 2015. Radiation-induced growth and isothermal decay of infrared-stimulated luminescence from feldspar. Radiation Measurements 81, 224-231.

Li, B., Li, S.-H., 2012. Determining the cooling age using luminescence thermochronology. Tectonophysics 580, 242-248.

Poolton, N.R.J., Ozanyan, K.B., Wallinga, J., Murray, A.S., Bøtter-Jensen, L., 2002. Electrons in feldspar II: a consideration of the influence of conduction band-tail states on luminescence processes. Physics and Chemistry of Minerals 29, 217-225.

Other references mentioned here are cited in the manuscript.