Supplement of

Surface paleothermometry using low-temperature thermoluminescence of feldspar

Rabiul H. Biswas et al.

Correspondence to: Rabiul H. Biswas (biswaarbisu@gmail.com)

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S1: Theoretical Model

The rate equation that describes the trapped charge population corresponding to a particular luminescence trap is described by Biswas et al. (2018) for thermoluminescence (TL) of feldspar for general order kinetics, and is shown below.

\[
\frac{d}{dt} (\bar{n}(r', t)) = \frac{\dot{D}}{D_0} \left(1 - \bar{n}(r', t)\right)^a - se^{-\frac{E}{kT} (\bar{n}(r', t))} - \bar{s}e^{-\rho r^{\frac{1}{2}} (\bar{n}(r', t))} (S1)
\]

where \( \bar{n} \) is equal to \( n/N \) (where \( n \) is the number of trapped electrons at time \( t \) and temperature \( T \), and \( N \) is total number of available traps), \( \dot{D} \) is the dose rate due to ambient radioactivity (Gy ka\(^{-1}\)), \( D_0 \) is the onset of dose saturation (Gy), \( a \) and \( b \) are the kinetic orders of trapping and thermal detrapping respectively, \( E \) is the trap depth or activation energy (eV), \( s \) and \( \bar{s} \) are the thermal and athermal frequency factor respectively (s\(^{-1}\)), \( \rho \) is the athermal fading parameter (Tachiya and Mozumder, 1974), and \( r' \) is a dimensionless distance that characterizes the probability of athermal escape (Huntley, 2006). The validity of the model for TL of feldspar has been demonstrated for samples from a controlled thermal environment, the KTB borehole (Biswas et al., 2018).

The above-mentioned model is valid for a single trapping energy. However, TL of feldspar arises from a continuous distribution of trapping energies (Biswas et al., 2018; Duller, 1997; Grün and Packman, 1994; Pagonis et al., 2014; Strickertsson, 1985). Thus, it is necessary to obtain the distribution of the model parameters along the TL glow curve to access the parameters for multiple TL thermochronometers (200-250 °C, 10 °C interval).

S1.1: Constraining model parameters

The kinetic parameters for growth (\( \dot{D} \), \( D_0 \) and \( a \)), thermal decay (\( E \), \( s \) and \( b \)) and athermal decay (\( \bar{s} \) and \( \rho' \)) can be constrained in the laboratory through three different experiments.

S.1.1.1: Growth Parameters

The growth of TL due to ambient radioactivity in nature can be reproduced in the laboratory using a multiple aliquot regenerative dose protocol (MAR; Aitken, 1985) and strong beta irradiation (0.22 Gy/s). The resultant dose response curves (DRC) follow the first term of Eq. 1 (Eq. S2).

\[
\frac{d}{dt} (\bar{n}(r', t)) = \frac{\dot{D}}{D_0} \left(1 - \bar{n}(r', t)\right)^a \tag{S2}
\]

The growth parameters (\( D_0 \) and \( a \)) are deduced by fitting the DRC using Eq. S3, obtained by solving Eq. S2.

\[
\bar{n} = 1 - \left(1 + (a - 1) \frac{D}{D_0} t\right)^{-\frac{1}{a-1}} \tag{S3}
\]

The dose rate parameter (\( \dot{D} \)) can be deduced separately by measuring radioactive element U, Th and K concentrations and converting them into an equivalent dose rate (Gy/ka) using DRAC (Durcan et al., 2015). The method has been described by King et al. (2016).
S.1.1.2: Thermal decay parameters

For the general case, the thermal detrapping of trapped electron from a specific center (with distinct $E$ and $s$) can be described by the equation given by May and Partridge (1964) (second term of Eq. 1):

$$\frac{dn}{dt} = -se^{-\frac{E}{kT_R}b}$$  \hspace{1cm} (S4)

If the sample is heated with a linear heating rate ($\beta$), the intensity ($I$) of the single TL peak can be described by the following equations, which is a truncated series approximation (Kitis et al., 1998).

$$I(T) = I_M \times b_{\frac{b}{b-1}} \times \exp\left(\frac{E}{kT} \times \left(\frac{T - T_M}{T_M}\right)\right) \ldots$$

$$\times \left[1 + \left(b - \frac{1}{2}kT_M \times \frac{2kT_M}{E} \right) + \left(b - 1\right) \times \left(1 - \frac{2kT}{E}\right) \times \left(\frac{T^2}{T_M^2} \times \exp\left(\frac{E}{kT} \times \left(\frac{T - T_M}{T_M}\right)\right)\right)\right]^{-\frac{b}{b-1}}$$

and,

$$s = \frac{2kT_M}{E} \left(1 + \left(b - 1\right) \frac{2kT_M}{E}\right)^{-1} \exp\left(\frac{E}{kT_M}\right)$$  \hspace{1cm} (S5)

where $I_M$ and $T_M$ are the maximum intensity and peak temperature respectively. Note that $T_M$ differs from $T_m$ as the former is the theoretical peak temperature of the given $E$, $s$ and $\beta$, and the latter is the maximum temperature of the fractional glow peak.

The above mentioned equation is valid for a single trapping energy level. However, TL of feldspar results from a continuous distribution of traps and the whole TL glow curve cannot be analyzed directly. To circumvent this Pagonis et al. (2014), proposed that a composite glow curve that arises from a continuous distribution of traps can be described as a linear combination of a large number of sub-peaks, each corresponding to a different but closely located energy level, $E$. Each sub-peak can be obtained by subtracting the consecutive fractional glow curves, obtained through the $T_m - T_{stop}$ method (McKeever, 1980). The details of the method are described by Pagonis et al. (2014) and Biswas et al. (2018).

S.1.1.3: Athermal decay parameters

The athermal loss of the TL signal of feldspar is thought to be due to quantum mechanical tunneling of trapped electrons and recombination with the nearest recombination center (Wintle, 1973). The distance dependent athermal or anomalous fading is described by Huntley (2006) (third term of Eq. 1):

$$\frac{d}{dt}(\bar{n}(r', t)) = -\bar{s}e^{-\frac{\beta r}{3\gamma}}\bar{n}(r', t)$$  \hspace{1cm} (S7)
Solving the above equation according to the formulation of Huntley (2006) the athermal loss of TL can be described as:

\[ n = \exp\left\{-\rho'\left[\ln(1.85t^*)\right]^3\right\} \]  \hspace{1cm} (S8)

where \( t^* \) is the time delay since the irradiation is ceased plus half of the laboratory irradiation time (Auclair et al., 2003).

**S1.2: Discretization of parameters from continuous distribution**

Several studies suggest that broad TL glow curve from feldspar arises from a continuous distribution of trapping energies, which is suggested by several methods, like \( T_m - T_{\text{stop}} \), the initial rise method, and analysis of fractional glow curves (Biswas et al., 2018; Duller, 1997; Grün and Packman, 1994; Pagonis et al., 2014; Strickertsson, 1985). Regardless, it is difficult to isolate a single trap with distinct kinetic parameters. Instead we assign the most probable kinetic parameters for each thermometer (glow curve temperature) from the distribution of parameters along the TL glow curve (as shown in Fig. 1 of main text). This is the method that we have adopted here and in Biswas et al. (2018). We then arbitrarily choose 10 °C TL temperature windows as distinct thermometers. A continuous distribution of trapping energies can be assumed as the sum of a large number of discrete traps (Pagonis et al. 2014). Thus a continuous distribution of trapping energies is discretized as shown in the figure below for the activation energy (\( E \)).

![Graph showing discretization of trap depths](image)

**Fig. S1:** The evaluated continuous distribution of trap depth (\( E \)) of sample MBTP1 (circles with error bar) and its discretization in 10 °C windows (green box; width is 10 °C and height 5 % of the median value).
Table S1: Kinetic parameters of sample MBTP9. Note that in the thermal decay parameters (E, s and b) no errors are mentioned. The mean values of these parameters were calculated from the distribution and an arbitrary error of 5% was considered. The dose rate ($\dot{D}$) value was taken from Lehmann et al. (2020).

| TL (°C) | $\dot{D}$ (Gy/ka) | $D_0$ (Gy) | $a$ | E (eV) | log.(s) | b | log.(s') |
|---------|-------------------|------------|-----|--------|---------|---|----------|
| 200-210 | 844 ± 44          | 1.00 ± 0.47| 1.21| 11.53  | 1.50    | -6.02 ± 0.07 |
| 210-220 | 773 ± 41          | 1.00 ± 0.03| 1.25| 11.63  | 1.49    | -6.13 ± 0.09 |
| 220-230 | 680 ± 37          | 1.00 ± 0.01| 1.29| 11.72  | 1.49    | -6.18 ± 0.10 |
| 230-240 | 625 ± 40          | 1.18 ± 0.33| 1.32| 11.79  | 1.49    | -6.33 ± 0.17 |
| 240-250 | 502 ± 36          | 1.10 ± 0.27| 1.36| 11.85  | 1.49    | -6.51 ± 0.24 |
| 250-260 | 371 ± 29          | 1.00 ± 0.22| 1.39| 11.90  | 1.48    | -6.60 ± 0.30 |
| 260-270 | 274 ± 33          | 1.00 ± 0.31| 1.43| 11.99  | 1.48    | -6.65 ± 0.41 |
| 270-280 | 205 ± 37          | 1.00 ± 0.44| 1.46| 12.05  | 1.48    | -20 ± 0.00  |
| 280-290 | 166 ± 42          | 1.00 ± 0.52| 1.50| 12.14  | 1.48    | -20 ± 0.00  |
| 290-300 | 140 ± 43          | 1.00 ± 0.79| 1.53| 12.20  | 1.47    | -20 ± 0.00  |

S2: Random thermal histories for inverse modeling

For random thermal history generation, Greenland ice core $\delta^{18}O$ data up to 60 ka (Svensson et al., 2008) as shown in Fig. S1a was scaled in the following way.

$$T(t) = T_{\text{base}} + T_{\text{amp}} \times f(\delta^{18}O)$$

(S9)

where $T_{\text{base}}$ is the temperature at 20 ka and $T_{\text{amp}}$ is temperature difference between the present day ($T_{\text{present}}$) and $T_{\text{base}}$ as illustrated in Fig. S1b. The scale was chosen in a way such that the temperature at 20 ka was fixed to zero and the present-day temperature randomly varied from 0 to 40 °C. $T_{\text{base}}$ was then randomly varied from -20 to 30 °C. Only a few random paths are shown in Fig. S1c out of the large number of random paths simulated (300,000).

Fig. S2: a) Greenland ice core $\delta^{18}O$ data up to 60 ka (redrawn after Svensson et al., 2008). b) illustration of scaling of $\delta^{18}O$ into a time Temperature (t-T) path. c) Few random paths are shown here and a total of 300,00 paths were generated for inverse modeling.
S3: Constraining kinetic parameters of natural sample

Fig. S3: Plots for kinetic parameters of sample MBTP1. a1-a4 are the growth of four thermometers (210-250 °C, 10 °C interval). The natural point is average of three discs. b1-b4 are the athermal fading of the four thermometers. c, d and e are the distribution of thermal kinetic parameters (E, s and b) and the four thermometers’ regions are illustrated.
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