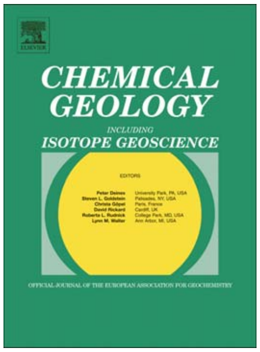

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# On the calibration of fission-track annealing models

S. Guedes[1], R. Jonckheere[2], P.A.F.P. Moreira[3], R. Hielscher[2]

[1] Departamento de Ciências Exatas e da Terra, Escola Paulista de Química, Universidade Federal de São Paulo, UNIFESP, Rua Professor Artur Riedel 275, 09972-270 Diadema, SP, Brazil.

[2] Geologisches Institut, Technische Universität - Bergakademie Freiberg, Bernhard-von-Cotta-Straße 2, 09596 Freiberg (Sachsen), Germany.

[3] Departamento de Raios Cósmicos e Cronologia, Instituto de Física "GlebWataghin", Cidade Universitria Zeferino Vaz, Universidade Estadual de Campinas, C.P. 6165, Campinas SP13083-970, Brazil.

## Abstract

To obtain thermal histories from fission track length data, the rates at which track lengths are reduced must be estimated. The usual method is to use empirical equations calibrated via laboratory analogues. A major concern is the extrapolation of the equations to predict temperatures on the geological time scale, which is far outside the range of the calibration data. Using objective statistical criteria, it is shown that the inclusion of geological data from age standards and deep boreholes is an important measure to ensure that model predictions of geological annealing rates are at least close to being correct. It is shown that high-temperature geological data should be the preferential target in the search for new field evidence. A discussion on the use and limitations of geological data and their consequences is presented.

Keywords: Experiment design; apatite annealing models; fission-track thermochronology; Kontinentale Tiefbohrung.

## 1. Introduction

Fission-track thermochronology is based on measurements of the number, lengths and orientations of fossil fission tracks in natural minerals (Fleischer et al., 1975, Durrani and Bull, 1987; Wagner and Van den haute, 1992; Gleadow et al., 2002). Fossil fission tracks are linear zones of lattice damage along the trails of the nuclear fragments released in spontaneous fission of $^{238}$U. The tracks crossing a polished internal mineral surface can be etched and counted with an optical microscope. Confined tracks in the grain interior are sometimes etched if they cut another track or a crack, which serve as conduits for the etchant. The etchable lengths of horizontal confined tracks appear to be shorter than the combined range of the two fission fragments (Paul and Fitzgerald, 1992; Jonckheere, 2003a, b) and are measured for fission track modelling. The combined action of time and temperature causes a restoration of the damage along a track; this effect, referred to as thermal annealing, results in a gradual reduction of its etchable length. Thus, with proper calibration, the length distribution of horizontal confined tracks can be used to infer the variations of temperature with time ($T$-$t$ path) that a mineral sample experienced during its geological history.

Apatite is the most commonly used mineral for this purpose. To establish the required calibration, the annealing kinetics of fission tracks in apatite has been investigated in laboratory experiments. For these experiments, the fossil fission tracks in a natural apatite are first erased through thermal treatment (e.g. 24 hours at 450 °C although the resistance to annealing varies with apatite composition). The sample is then irradiated in a nuclear reactor to produce a population of "fresh" tracks by thermal-neutron-induced fission of $^{235}$U and step-heated to a constant temperature between 100 and 350°C for a time interval of 1 to 1000 hours (Green at al., 1986; Crowley et al., 1991; Carlson et al., 1999; Ravenhurst et al., 2003; Barbarand et al., 2003; Tello et al., 2006). The track lengths are measured after each experiment ($T_i$, $t_i$). Empirical (Laslett et al., 1987; Crowley et al., 1991; Laslett and Galbraith, 1996; Ketcham et al., 1999, 2007a; Guedes et al., 2007) and semi-empirical (Carlson, 1990; Guedes et al., 2006a) annealing models have been proposed to describe the results. The model parameters have been determined by fitting the laboratory data. However, in geological applications, the annealing equations are applied to ($T$, $t$)-conditions far outside the laboratory range, i.e. to heating durations between 5 and 500 Ma and temperatures of 0 to ~120°C.

The annealing equations are such that points of equal reduced track length ($r = L/L_0$) lie on straight or curved (iso-retention; iso-annealing) lines that are either parallel or meet in a single (fanning) point in Arrhenius space ($1/T$, $\ln(t)$). The iso-retention lines corresponding to different annealing equations behave in a similar fashion within the ($T$,$t$)-range of the experimental data but deviate from each other when extrapolated outside the data range, in particular to geological timescales. It is therefore becoming more and more common for researchers to compare geological predictions with field evidence. Green et al. (1989) and Green (2004) claim that the model of Laslett et al. (1987), the most used in geological applications, is compatible with data from boreholes in the Otway Basin (Australia). Ketcham et al.

(1999) and Kohn et al. (2002), in contrast, argue that this model underestimates the length reduction at low temperatures on a geological timescale. Issler (1996) and Ketcham et al. (1999; 2007a) proposed to calculate specific index (benchmark) temperatures and lengths from the model equations for comparison with field evidence and showed that the benchmark temperatures for the Laslett et al. (1987) equations are in less than perfect agreement with the Otway Basin data. This has led researchers to acknowledge the limitations of extrapolating laboratory-based equations and to suggest alternative calibrations. Kohn et al. (2002) proposed to change the initial mean track length ($L_0$) in the Laslett et al. (1987) model but this was shown to be an inconsistent procedure (Guedes et al., 2006b). Others have suggested including geological data in the fitting procedure for estimating model parameters (Guedes et al., 2005; Jonckheere et al., 2005).

The importance of geological information for evaluating, adjusting or even fitting annealing parameters is clear. It is unclear, however, *how* important geological constraints are relative to laboratory data and whether one geological benchmark is as effective as another, or - if not - which provides the tighter constraints on the annealing parameters. There are also other considerations related to the inclusion of geological annealing data: [1] Jonckheere and Wagner (2000), Barbarand et al. (2002) and Jonckheere, (2003a) suggest that the annealing kinetics of fossil fission tracks differs from that of induced tracks; if this is confirmed, then the significance of laboratory data for calibrating annealing equations is diminished; [2] in geological samples, fresh tracks accumulate at the same time as older tracks are annealed, posing a problem of *how* geological information can be exploited for estimating annealing parameters. Using objective statistical criteria, it is shown that geological information from age standards and deep boreholes is essential to ensure a reliable calibration of the annealing equations. The statistical approach and the approximations needed to incorporate geological information and their associated errors are discussed.

## 2. Method

### *2.1. Annealing equation*

The basis of the calculations is an annealing equation with a convenient mathematical form that gives reasonable predictions at laboratory and geological timescales. The chosen equation is that of Laslett and Galbraith (1996), which is a development of the simple fanning linear Arrhenius model of Laslett et al. (1987):

$$L = L_0 \left[ 1 - \exp\left( C_0 + C_1 \frac{\ln(t) - \ln(t_c)}{\frac{1000}{T} - \frac{1000}{T_c}} \right) \right]^{1/3} \tag{1}$$

$L_0$ ($\mu_{max}$ in Laslett and Galbraith, 1996), $C_0$, $C_1$, $t_c$ and $T_c$, are parameters to be estimated from the data; $1/T_c$ and $\ln(t_c)$ are the temperature and time co-ordinates of the fanning point in the Arrhenius diagram. The "initial" mean track length, $L_0$, in particular, is a free parameter in order to account for the fact that the "true" "initial" length can exceed the measured mean length of induced tracks. Donelick et al. (1990) observed a continuous reduction of the mean induced track length until up to a month after the irradiation. Thus, all measured mean track lengths must be considered underestimates of the track length at the time of track formation. In addition, apatites of different composition can have different "initial" mean track lengths (Carlson et al., 1999). The purpose of equation (1) in the present context is to provide a general description of the variation of $L$ with $T$ and $t$. Because this variation is independent of the scaling factor $L_0$, its exact value is of no consequence and $L_0$ can be considered as a constant. We do not consider apatite composition or anisotropic annealing: the annealing rates of different apatites are related through the equations of Ketcham et al. (1999) and Ketcham et al. (2007a) and the annealing rate along one orientation is related to that along another direction through the c-axis conversion of Donelick et al. (1999) and Ketcham et al. (2007b). Equation (1) with constant $L_0$ is thus an adequate general indicator of the variation of $L$ with $T$ and $t$.

## *2.2. Experimental space*

In an annealing experiment, an apatite aliquot is step-heated to a constant temperature, $T_i$, which is maintained for a time $t_i$. After cooling and etching, the mean horizontal confined track length, $L_i$, is measured. Let each pair ($T_i$,$t_i$) be called an "experiment" and "experimental space" the set containing all possible experiments. In general, the experimental space is limited to the region of interest for a given application but also by practical considerations. In the case of fission-track annealing, the temperatures of interest range from values at which almost no length reduction is observed to temperatures at which annealing is complete. For 1 hour annealing experiments on apatite, this range is ~100 - 350°C and for 1000 hours the temperature for total annealing is ~300°C. The temperature boundaries of experimental space are thus irregular. The time dependence of the annealing process is logarithmic and the times of interest extend to ~500 Ma. The time limits of experimental space are therefore determined by practical factors. The longest published annealing experiment lasted 800 days (Ravenhurst et al., 2003) but most are limited to 1000 hours.

The relevant time span for geological applications extends far beyond the experimental space of laboratory experiments. Therefore experimental space is redefined as comprising a "laboratory" and a "natural" space, so as to include the geological data available for calibration, on condition that well-documented geological samples in terms of ($T_i$,$t_i$) and $L_i$ can be interpreted in the same way as laboratory experiments despite the difficulties mentioned in section 1. The boundaries of natural experimental space are determined by the properties of the available samples. The obvious candidates are the accepted and candidate apatite age standards that are believed to have remained at constant near-ambient temperatures (~25°C) for almost all the time since the formation of the oldest remaining track: Durango (DUR; 31.4 Ma; Naeser and Fleischer, 1975; Green, 1985; McDowell et al., 2005); Fish Canyon Tuff (FCT; 27.8 Ma; Hurford and Gleadow, 1977; Naeser et al., 1981; Hurford and Green, 1983; Green, 1985; Hurford and Hammerschmidt, 1985; Schmitz and Bowring, 2001; Lanphere and Baadsgaard, 2001; Schmitz et al., 2003); Mount Dromedary (MTD; 98.8 Ma; Williams et al., 1982; Hurford and Green, 1983; Green, 1985; Spell and McDougall, 2003) and Limberg t3 (LIM; 16.4 Ma; Kraml et al., 1996, 2006; Rahn et al., 2001). For the present calculation, we select the accepted age standards (DUR and FCT; Hurford, 1990a; b) for which it has been established that their independent fission-track ages are in agreement with their reference ages (Jonckheere, 2003b; Enkelmann et al., 2005) and the low-temperature benchmark proposed by Ketcham et al. (1999; 2007a) that never experienced temperatures much in excess of ~15°C (ODP; 115 Ma; Vrolijk et al., 1992).

The (*T*, *t*, *L*)-data provided by age standards cluster around low temperatures and modest degrees of annealing. High-temperature benchmarks are available from deep boreholes with well-documented thermal histories. Although several borehole studies have been carried out (e.g. Naeser, 1979; 1981; Briggs et al., 1981; Gleadow and Duddy, 1981; Coyle et al., 1997; House et al., 2002; Hendriks, 2004; Lorencak et al., 2004) or are in progress (Jonckheere et al., 2005; 2006) the results have hitherto not been used as part of the data sets used for calibrating annealing equations. In view of their different geological and tectonic settings, ages, thermal histories, sampling densities and methods of analysis (age determination or confined track length measurements), it is difficult to establish which reliable high-temperature benchmarks these studies provide. However, this is not a requirement for the present investigation, which addresses the question of how important borehole samples *as such*, i.e. as opposed to age standards and annealing experiments, are for calibrating the annealing kinetics of fission tracks in apatite. For this reason, and with a view to future work, we concentrate on the KTB borehole (Kontinentale Tiefbohrung). The advantages of the KTB are: [1] it spans the entire range from ambient temperatures (10°C) at the surface to complete annealing at 4 km depth (130°C); [2] all the samples remained close to their current temperatures since the onset of track accumulation (Jonckheere, 2003a); [3] there is little lithological variation and almost no variation of apatite composition throughout the depth interval 0-4km (Jonckheere et al., 2006); [4] the available dataset is the most comprehensive in terms of the number of samples (sampling interval: ~50m) and the number of confined-track-length measurements (>3000 and increasing; Wauschkuhn, 2005).

We assume that the KTB provides 25 useful benchmarks (natural experiments) at 5°C intervals; all samples are ~65 Ma (Figure 1). The age standards (DUR and FCT) and the ODP sample each contribute one benchmark. The number of laboratory experiments is in principle unlimited. Based on representative experiments (Green at al., 1986; Crowley et al., 1991; Carlson et al., 1999; Ravenhurst et al., 2003; Barbarand et al., 2003; Tello et al., 2006), laboratory space was subdivided in 44 separate experiments (11 annealing temperatures at 25°C intervals and 4 annealing times; Figure 1), minus 6 for which the annealing equation predicts complete erasure of the tracks. This is close to the minimum that might be considered sufficient. On the other hand, crowding the laboratory space with what the statistical criteria might later dismiss as redundant experiments that can be deselected at little cost in favour of natural experiments would overemphasize the latter. Our aim is to evaluate the importance of geological benchmarks relative to a representative but not an inflated set of laboratory experiments. The set composed of 38 laboratory experiments, 3 standards and 25 KTB samples constitutes our experimental space (Figure 1).

### *2.3. Statistical criteria*

The aim of an annealing dataset is to calibrate an annealing equation i.e. to determine its parameters with the tightest possible confidence intervals. If a practical constraint, such as time, prevents that all possible experiments be carried out, so that just a fixed number *n* can be performed, then a specific set of experiments must be selected from experimental space. de Aguiar et al. (1995) showed that the combination of *n* experiments that minimizes the confidence ellipsoids of the parameters is the one that minimizes the D-criterion:

$$\phi_D = \det(\mathbf{X}'\,\mathbf{X})^{-1/p} \tag{2}$$

in which $\mathbf{X}$ is the model matrix, $\mathbf{X}'\,\mathbf{X}$ is Fisher's information matrix (Fisher, 1925) and $p$ the number of model parameters $a_1, \ldots, a_p$, . The model matrix is an $n \times p$ matrix, containing, in the columns (j), the partial derivatives of the model equation with respect to its different parameters, calculated at ($T_i$, $t_i$), and, in the rows (i), the different experiments. Thus, for a model equation $f(t, T; a_1, \ldots, a_p)$, the model matrix elements are:

$$x_{ij} = \left.\frac{\partial f}{\partial a_j}\right|_{(T_i, t_i)} \tag{3}$$

The D-criterion can only be applied to sets with the same number of experiments. Another useful quantity, A, is equal to the trace of the dispersion matrix or the sum of the elements on its main diagonal. The dispersion matrix is the inverse of the information matrix: $(\mathbf{X}'\,\mathbf{X})^{-1}$. Its trace A is proportional to the predicted variance. Thus the best set of *n* experiments is the one that minimizes this variance, or minimizes A:

$$A = \mathrm{tr}((\mathbf{X}'\,\mathbf{X})^{-1}) \tag{4}$$

The A-criterion can be used to compare sets of different sizes. The D and A criteria can be combined to determine both the minimum number of experiments for an efficient estimation of the model parameters and the most efficient set of that size. Fixing the number of experiments at *n*, the D-criterion is used to find the most efficient set of size *n*. Then, the value of *A* is calculated for that set. The calculation is repeated for different *n* until the variation of A is less than a given limit or until A tends to a plateau (Moreira et al., 2005).

## 3. Application

A first estimate of the model parameters is needed to proceed with optimization. In this case, their values have been determined by fitting the Durango data of Carlson et al. (1999). For reference, let experimental space be restricted to laboratory experiments and their number be equal to the number of possibilities (38; Figure 1). The corresponding A and D criteria are listed in Table 1. Now let the standards be included but the number of experiments remain the same: all three standards are selected at the expense of laboratory experiments in the mid ($T$, $t$)-range (Figure 1a), reflecting the intuitive rule that a system is best characterised by its boundaries. The much lower values of $A$ and $\Phi_D$ (Table 1) further indicate a significant improvement of parameter estimation. Next the KTB data are included, keeping the number of experiments constant. Optimization under these conditions leads to the distribution in Figure 1b: 15 natural experiments have been selected at the expense of as many laboratory experiments in the mid-temperature range, resulting in a further significant improvement of the statistical criteria (Table 1). The experiments appear to be so distributed as to cover, on the one hand, the conditions where $L_0$ can be best estimated (low temperatures) and, on the other, those where $L$ varies most as a function of $T$ (high temperatures).

Figure 1 presents a plan of how best to proceed in order to estimate the parameters in equation (1) from scratch. This does not reflect the current situation, however, because comprehensive sets of laboratory data are already available and annealing parameters have been fitted to them. Data on age standards are also available. The real question is thus if, and how much, additional investigation of borehole samples can improve the current parameter estimates and how to proceed for maximum effect. To answer this question within our conceptual framework, we assume that laboratory space is exhausted and that all standards have also been investigated, and we examine the effects of supplementing them with a variable number of borehole experiments. The immediate answer to the question how many experiments should be added is that every possible experiment should be carried out because more data means better parameter estimates. The law of diminishing returns, on the other hand, implies that each successive addition improves the estimates by a decreasing margin. A cost/effect calculation is thus required to set a practical limit.

This is done as follows: all laboratory experiments and standards are selected and the reference values of $A$ and $\Phi_D$ are calculated. Then, a single borehole experiment is added. The optimization is then performed using the D-criterion and the A-criterion is calculated for the resulting configuration. In the next step, two borehole experiments are allowed. D-optimization is performed again and the A-criterion calculated for the new configuration. This is repeated for $n = 3, 4 \ldots$ borehole experiments. $A$ is plotted as a function of the number of D-optimized experiments in Figure 2a. It is seen that $A$ decreases fast at low values of $n$. This means that a few well-selected borehole samples have a significant positive effect on the estimation of annealing parameters even when a full set of laboratory experiments and standards is available. $A$ decreases more slowly as $n$ increases, reflecting the diminishing return. It is possible to conclude from Figure 2a that, in this particular case, not much is gained by including more than about 15 borehole experiments, on the express condition that their selection is at all times the most effective (D-optimal).

It has been noted that the D-optimal set with $n+1$ experiments differs from that with $n$ in the addition of the new experiment but not in the distribution of the first $n$ experiments. This is not the general case but a feature of the question addressed here. It is interesting to take advantage of this to illustrate another point. Figure 2b shows the sequence in which the borehole data are added to the set of laboratory and standard data, i.e. according to their relative impact on the estimation of the annealing parameters. The experiments selected first are concentrated in the high temperature range. The first low-temperature experiment is selected at $n = 17$ and thus has little effect on the parameter estimation. This is because $L$ varies little in low-temperature range, i.e. the factor multiplied with $L_0$ in equation (1) remains close to 1, so that the annealing parameters therein are difficult to estimate from low-temperature data. For the same reason, low-temperature data are efficient at estimating $L_0$ but it appears that its value is already well constrained by the laboratory experiments and the standards. It is also worth noting that the 15 most important borehole samples are all situated in what is still referred to as the partial annealing zone, i.e. the interval of accelerated variation of the fission-track age with temperature or depth (Wagner and Van den haute, 1992) and thus, according to equation (1), the temperature range from which $C_0$, $C_1$, $t_c$ and $T_c$ can best be estimated. This highlights the dependence of the D-criterion on equation (1) and suggests a possible complication related to the fact that it does not provide an accurate description of the reduction of mean confined track length as a function of $T$ and $t$ in geological samples; the consequences are discussed in section 4. If the experiment is repeated without the standards, the result is about the same (Figure 2c). The implication is that laboratory data provide by far the tightest constraints on $L_0$. The practical point is that, although all geological experiments improve the annealing parameter estimates, high-temperature data, i.e. borehole samples from the partial annealing zone should be the main target when looking for field evidence.

## 4. Geological data

Using geological data for calibrating annealing equations involves an approximation because there is a constant addition of new tracks during the annealing episode, so that later tracks have been annealed for a shorter length of time than earlier ones. This gives rise to a superposition of track populations with mean lengths ranging from the "correct" value for the assumed annealing time for the oldest tracks to the unannealed track length for the most recent. The mean track length in a geological sample is thus longer than that of a fictitious laboratory experiment of the same $(T_i, t_i)$. The impact of track accumulation on the overall mean length is mitigated by the logarithmic dependence of $L$ on $t$ (equation 1), which causes the track lengths to be bunched towards the lower end of the range. To address this problem, we define an "equivalent temperature" $T_{eq,i}$, i.e. a fictitious temperature, higher than $T_i$, that, over a time $t_i$, produces the same overall mean length reduction in a geological sample as in a laboratory sample at $T_i$. To establish a numerical relationship between $T_{eq,i}$ and $T_i$, $t_i$ is set at a fixed value (0.1, 1, 10, 100, 1000 Ma; the relationship between $T_{eq,i}$ and $T_i$ is not sensitive to $t_i$ because of the logarithmic dependence of $L$ on $t_i$). For a given $T_i$, the mean track length in a geological sample is calculated in the usual manner: $t_i$, is divided in $N$ intervals of duration $t_i/N$ and $N$ values of $L$ are calculated for $(1 - \frac{1}{2}/N)t_i$, $(1 - \frac{3}{2}/N)t_i \ldots \frac{1}{2}/N\ t_i$. The length-biased mean (Laslett et al., 1982), $L_m$, of these $N$ values is the mean track length in a geological sample that remained at a constant temperature $T_i$ for a time $t_i$; substituting $L_m$ and $t_i$ in equation (1) and solving for $T$ gives $T_{eq,i}$. It appears from the numerical results that the relationship between $T_{eq,i}$ and $T_i$ is well described by a straight line:

$$T_{eq,i} \approx A_1\, T_i - A_2 \tag{5}$$

The slope $A_1$ and intercept $A_2$ are estimated by least squares regression. At the point where converted ($T_i \rightarrow T_{eq,i}$) geological data are used for estimating the annealing parameters $L_0$, $C_0$, $C_1$, $t_c$ and $T_c$, the calculation becomes self-referring, necessitating an iterative procedure. This is because the conversion of geological data makes use of equation (1) and thus of initial estimates of the annealing parameters. In the present case, first estimates of $A_1$ and $A_2$ were obtained with equation (1) calibrated against the Durango data of Carlson et al. (1999), but excluding all geological data (Figure 3). $A_1$ and $A_2$ were then used to calculate $T_{eq,i}$ for the geological data with equation (5). In this case, the geological dataset included the KTB data of Coyle et al. (1997) and the low-temperature benchmarks discussed in section 2. In a second step, equation (1) was reparameterised by fitting all the available data, including the converted geological data. $A_1$ and $A_2$ were then recalculated for the new parameters in equation (1), giving rise to a modified temperature conversion. This procedure was repeated until $A_1$ and $A_2$ remained constant; their successive values are listed in Table 2. Figure 3 shows the final adjustment of equation (1) to the complete set of laboratory and geological data and Figure 4 the relationship between $A_1$ and $A_2$. It is worth noting that the trend of the conversion curve is approximately independent of the degree of annealing and of the duration of heating in the investigated time span, which includes that of interest for geological applications.

## 5. Discussion and conclusion

### 5.1. Significance and limitations

It has been shown in the previous sections that objective statistical criteria (A and D) show a strong preference for including geological data for the purpose of calibrating annealing equations. It is reasonable to expect that this preference is little affected by the exact form of the model equation. Moreover, the A and D criteria evaluate the overall fit over the entire ($T$, $t$)-domain of laboratory and geological data, without being weighted in favour of geological predictions. The A and D criteria at the same time provide a "ranking" of geological samples, i.e. a measure of the "return" expected from a sample annealed at given ($T$, $t$)-conditions, as opposed to samples annealed at different conditions, and an indication of the overall return expected from an optimal set of size $n$, as opposed to a set of different size. The condition is implied that geological data can be used in the same manner as laboratory data. This is not the case because there is a constant addition of new tracks during the annealing period. A method is proposed to calculate an equivalent temperature, $T_{eq,i}$, so that geological annealing conditions $(T_i, t_i)$ can be translated to laboratory equivalents $(T_{eq,i}, t_i)$, permitting geological data from samples that remained at constant temperatures to be used for calibration. The procedure is iterative with alternate estimates of the parameters $L_0$, $C_0$, $C_1$, $t_c$ and $T_c$ and the relationship between $T_{eq,i}$ and $T_i$. It turns out that this relationship is linear, almost independent of $t_i$ for geological samples and that there is not much difference between $T_{eq,i}$ and $T_i$: $T_{eq,i} \approx T_i + 5°C$.

A related and more serious limitation to the use of geological data for estimating annealing parameters is due to the fact that confined tracks below a certain minimum length are not observed. The causes are not understood in detail but it is probable that anisotropic track length reduction, tracks segmentation and observation effects are involved (Laslett et al., 1984; Galbraith and Laslett, 1988; Carlson, 1993; Green et al., 1993; Galbraith, 2002; Ketcham, 2003). The combined effect of, on the one hand, the addition of long tracks and, on the other, the elimination of short tracks creates a dynamic equilibrium wherein the confined-track length distribution and the mean confined track length become invariant and thus independent of $T_i$. This also happens in the idealized case of pure length-proportional observation probabilities (Wauschkuhn et al., 2006). This effect has the practical consequence that the preferred high-temperature benchmarks cannot be used for calibrating annealing equations. There is no method to overcome this limitation. It depends on several factors at which temperature this effect sets in for given $t_i$. As a first-order estimate, geological samples may be useful down to $L/L_0 \approx 0.6$. It should however be noted that the traditional laboratory-based calibrations are limited to $L/L_0 \approx 0.4$, for related reasons. Figure 3 shows a comparison of calibrations using laboratory (dashed lines) and laboratory plus geological data (solid lines). The first provides a poor fit to the geological data. On the other hand, the inclusion of geological data worsens the fit to the laboratory data. Note also that, although model curve approaches the borehole data, the fit to these data is still rather poor. This is not surprising since equation (1), and other annealing equations, are empirical and reflect rather arbitrary *a priori* assumptions instead of the true physics of the annealing process. This could also indicate that the chosen equation is not the best for describing the annealing process. Ketcham et al. (1999) suggested that better fits could be obtained with a curvilinear equation.

Figure 5 shows the laboratory, standard and borehole data in Arrhenius space, with the iso-retention lines for $L/L_0 = 0.6$ and $0.9$ fitted to the laboratory data (dashed lines) and the laboratory plus geological data (solid lines) superimposed. At geological timescales, the temperature predictions are in good agreement for $L/L_0 = 0.6$ but not for $L/L_0 = 0.9$. The inclusion of geological benchmarks eliminates a problem inherent in laboratory-based models, i.e. that they underestimate the extent of track length reduction at low temperatures. This was pointed out by Kohn et al. (2002) with reference to the annealing equation of Laslett et al. (1987) and by Enkelmann et al. (2005) with reference to later equations. Figure 5 shows that this is also the case if equation (1) is fitted to the laboratory data alone. It is a matter of debate whether a calibration against laboratory data is preferable in principle to one that includes geological benchmarks. However, the annealing equations are all empirical and intended as practical tools for geological applications rather than as quantitative descriptions of a physical process. The general rules of calibration that are also applicable in this case suggest that the calibration data should be as little as possible removed from the area of application. Therefore, for practical applications, it is preferable to use an equation calibrated against geological data on condition that they are unambiguous. In this connection, it is perhaps useful to consider that the reference ages of geological samples are used for the direct calibration of fission-track ages with the Z and ζ-methods. Another argument in favour of geological benchmarks is related to the supposed qualitative differences between the annealing of induced tracks under laboratory conditions and the annealing of fossil tracks in the geological environment (Jonckheere and Wagner, 2000; Barbarand et al., 2002). If such differences exist, geological data gain further in importance. Laboratory data remain important because they constrain the slopes of the iso-retention lines in the Arrhenius plot.

### *5.2. Possible controversial issues*

These results provide *a priori* criteria for sample selection and are thus strategic: the samples that should be targeted are identified without consideration for the relative uncertainties associated with their later measurement. Measurements on samples that exhibit higher degrees of annealing are expected to have higher uncertainties and to be less effective at constraining the annealing parameters than our calculation assumes. The effects of uncertainties on fitting annealing equations are however a matter for *a posteriori* investigation that does not affect the sequence in which the samples should be selected. Error considerations are nevertheless relevant because they suggest practical measures that can be taken. In the case of the more annealed samples, an increased sample size can counteract the lesser number of confined track-length measurements expected for each grain. Practical steps can also be taken to increase the number of measurable confined tracks per grain to a level well above that where the statistical uncertainties cease to matter. This can be achieved through irradiation with fission fragments from an external $^{252}$Cf-source (Donelick and Miller, 1991), step-etching (Ito, 2004) or heavy-ion irradiation (Jonckheere et al., 2007; Min et al., 2007).

The (T,t)-conditions at which geological samples have been annealed are, in general, not so well defined as those at which samples are annealed in experiments. The relevant question is, however, *how* ill or well defined these conditions are or need to be in a specific case. This is determined by independent geological evidence, augmented, in the case of borehole samples, by the internal evidence of the fission-track age *versus* elevation and mean track length *versus* elevation profiles. This question has a reverse side: how relevant are short-duration, high-temperature experiments on induced tracks in pre-annealed apatites for the long-term, low-temperature behaviour of fossil tracks in radiation-damaged geological samples. The differences between lab-based and geological estimates of the closure temperatures of titanite and zircon are considerable. The effect of radiation damage on the track annealing kinetics in apatite is certain to be less because no apatite has been observed to have become metamict from self-irradiation but no evidence has been adduced that it is negligible. Such supporting evidence as there is, is based on geological samples and at least ambivalent. The matter is thus more complex that at first appears. Geological samples are only useful for the calibration of annealing equations on condition that there are minimum assurances that the assumed (T,t)-conditions are valid. On the other hand, the data without doubt describe the process of interest in the material of interest at the timescale of interest for geological applications. The (T,t)-conditions of lab experiments are well defined but it remains to be established that they also describe the geological annealing process. Both approaches therefore have their strengths and weaknesses.

Recent experiments showed that pressure and stress may affect the fission track annealing rate in apatite (Wendt et al., 2002; 2003). A critical discussion (Kohn et al., 2003; Vidal et al., 2003) and new experiments (Donelick et al., 2003) first contradicted and later confirmed a pressure effect above ~1 kbar (Kohn et al., 2004). These results entail that the annealing equations derived from experiments at atmospheric pressure overestimate annealing rate in geological samples, in particular at high temperatures, assuming a geothermal gradient of 30°C/km and geobaric gradient of 270 bar/km. It is however not improbable that similar problems will arise if higher pressures must be used in laboratory studies aimed at investigating pressure effects, in a similar fashion to high temperatures in thermal experiments (Donelick et al., 2005).

Irrespective of the calibration procedure, the resulting annealing equations must be validated. Such support as there is at present, is based in part on the examination of age standards and in part on volcanoclastic samples from ODP cores that experienced low temperatures throughout their well-documented geological histories (Vrolijk et al., 1992; Spiegel et al., 2007). These studies are important but nevertheless limited because (1) the different samples span a negligible range of (low) temperatures, so that the "initial" track length $L_0$ is the only annealing parameter that can effectively be validated; (2) the fact that the temperature of individual samples was not constant throughout their geological histories necessitates calculations based on the principle of equivalent time (Duddy et al., 1988) that is itself in need of validation. Samples from deep boreholes, in contrast, span annealing conditions from ambient temperatures up to those of complete erasure of the tracks. This makes it possible to validate - or calibrate - the annealing equations over the entire temperature range of geological interest. In addition, samples from rigid, coherent basement blocks allow examining the internal consistency of a set of related thermal histories, with, in the simplest case, a constant temperature offset. Our calculations assume that such samples exist and, in addition, that each sample remained at constant temperature since the onset of track accumulation, which allows validating - or calibrating - annealing equations without appeal to the principle of equivalent time.

Our theoretical sampling scheme stands whether this borehole exists or not. We proposed the *Kontinentale Tiefbohrung* (KTB) as a potential candidate based on circumstantial evidence: (1) the development of a peneplaine and preserved illitic weathering products from the early Palaeocene to Eocene (Peterek et al., 1994) indicates tectonic quiescence since the onset of track accumulation at the end of the Cretaceous; (2) there is no sediment record of post-Cretaceous uplift, in contrast to the preserved testimonies to much older episodes of uplift and burial from the Permian through to the end of the Cretaceous (Schröder et al., 1987; Peterek et al., 1997); (3) there is no indication of differential uplift in the distribution of fission-track ages from the area surrounding the KTB (Hejl et al., 1998); (4) the regional palaeostress remained extensional since the end of the Cretaceous (Schröder et al. 1987; Peterek et al. 1997); (5) post-Cretaceous faults intersecting the KTB borehole are normal with small vertical displacements (Zulauf and Duyster, 1997); (6) neither the apatite (Warnock et al, 1997) nor the titanite (Stockli and Farley, 2004) U/Th-He age *versus* depth profiles show evidence of post-Cretaceous tectonic disturbance. In addition, there is internal evidence from the apatite fission-track age and mean track length *versus* depth profiles (Coyle et al., 1997; Wagner et al. 1998; Wauschkuhn et al., 2006). Their mathematical analysis is not completed but it is evident that, irrespective of the actual track annealing kinetics and therefore of matters of calibration, these profiles are reconcilable with a narrow range of thermo-tectonic histories. The partial annealing zone between 2 and 4 km and "total retention" zone between 0 and 2 km are, in themselves, sufficient to conclude that this narrow range includes the assumed conditions of near-constant temperature.

In conclusion, borehole data can provide a significant contribution to the calibration of annealing equations. There are certain limitations but so are there to a calibration based on annealing experiments. In our opinion, it is in the interest of fission-track method to allow both calibration procedures to proceed and to examine their interconsistency and to compare their relative performance in practical geological applications.

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Figure captions

Figure 1. D-optimal data structure for estimation of annealing parameters: a) including laboratory data and standards; b) including laboratory data, standards and borehole samples. Black: not available, grey: selected experiments; white: deselected experiments.

Figure 2. (a) Evolution of the A-criterion as a function of the size, n, of a D-optimal set of borehole experiments supplementing the full set of laboratory experiments listed in Figure 1a; (b) ranking of the borehole experiments when standards are included as well as a full set of laboratory data; (c) ranking of the borehole experiments excluding standards.

Figure 3. Plot of $L$ (mean track lengths) *versus* annealing temperature $T$ for the data from laboratory experiments (■ 1 h; □ 10 h; ● 100 h and ○ 1000 h), standards (★ Durango apatite; ☆ Fish Canyon Tuff apatite and ▲ ODP apatite) and borehole samples (▽ KTB) used in the calculations with predictions from equation (1) calibrated against the laboratory data alone (dashed line) and all data (solid line). Laboratory data from Carlson et al. (1999); standards: Durango apatite, Fish Canyon tuff apatite and ODP apatite of Vrolijk et al. (1992); borehole data from the *Kontinentale Tiefbohrung* (Coyle et al., 1997).

Figure 4. Relationship between the temperature $T_i$ of a geological sample in which tracks accumulate in the course of an isothermal annealing episode of duration $t_i$ and the temperature $T_{eq,i}$ of a fictitious laboratory annealing run on pre-existing tracks of the same duration and resulting in the same reduction of the mean track length.

Figure 5. Arrhenius plot of the data from laboratory experiments, standards and borehole samples used in the calculations and a comparison of the iso-annealing lines for equation (1) fitted to the laboratory data (dashed line) and to all data (solid line). Note that the symbols for DUR (black star) and FCT (white star) are almost completely superimposed.

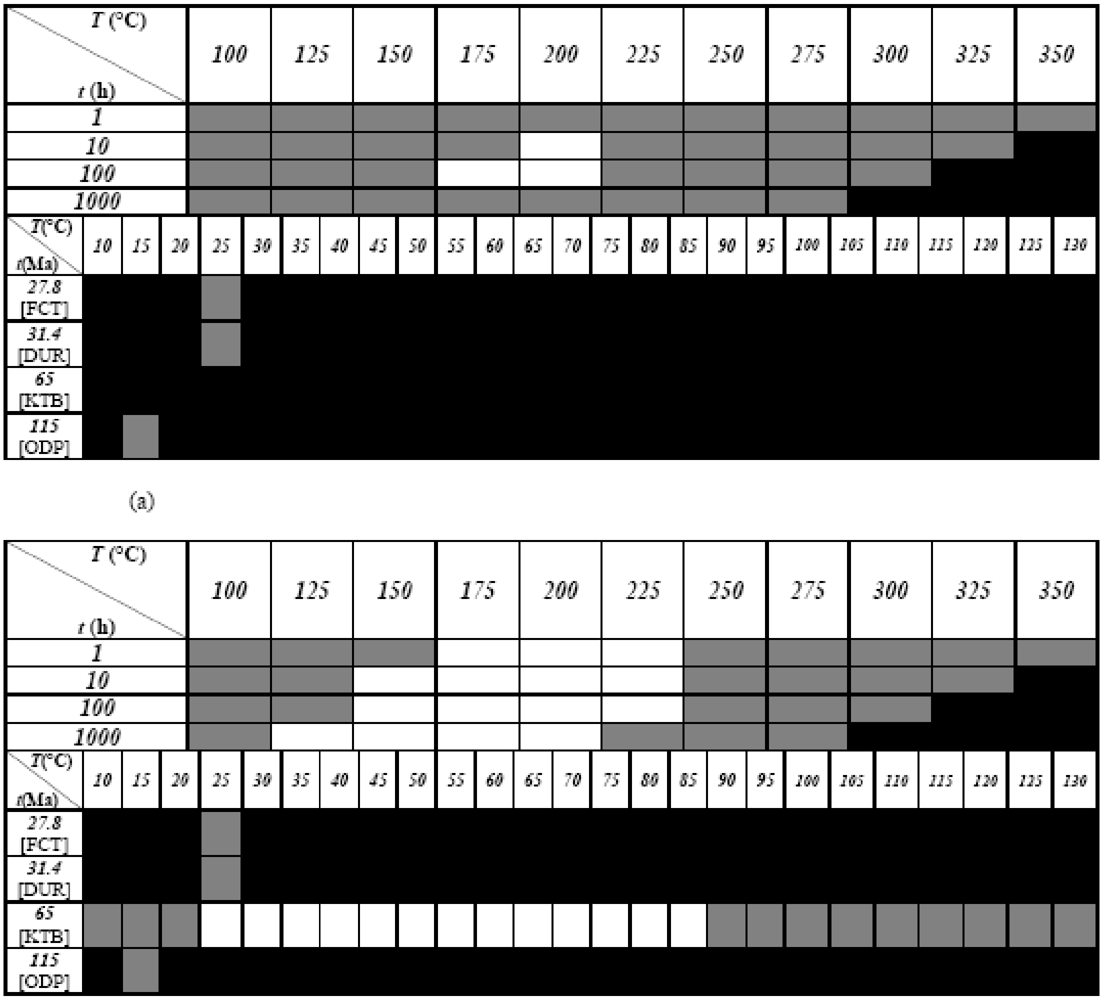
Table 1. Comparison of $\Phi_D$ and $A$ for different calibration approaches

| **Data** | **$\Phi_D$ ($10^{-3}$)** | **$A$ ($10^{3}$)** |
|---|---|---|
| Laboratory | 6.35 | 2.48 |
| Laboratory + standards | 1.25 | 0.41 |
| Laboratory + standards + KTB | 0.51 | 0.27 |

Table 2. Parameters for the recursive procedure leading to Eq. 7

| Step | A1 | A2 | R |
|---|---|---|---|
| 1 | 0.99659<br>(0.00327) | 3.23973 (1.291) | 0.99985 |
| 2 | 1.00005<br>(0.00245) | 5.12899<br>(0.93214) | 0.99992 |
| 3 | 0.99966<br>(0.00254) | 4.99884<br>(0.96631) | 0.99991 |
| 4 | 0.99966<br>(0.00254) | 4.99884<br>(0.96631) | 0.99991 |

Figure 1



| *T* (°C) / *t* (h) | 100 | 125 | 150 | 175 | 200 | 225 | 250 | 275 | 300 | 325 | 350 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ |
| 10 | ▒ | ▒ | ▒ | ▒ | □ | ▒ | ▒ | ▒ | ▒ | ▒ | ■ |
| 100 | ▒ | ▒ | ▒ | □ | □ | ▒ | ▒ | ▒ | ▒ | ■ | ■ |
| 1000 | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ■ | ■ | ■ |

| *T*(°C) / *t*(Ma) | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 | 55 | 60 | 65 | 70 | 75 | 80 | 85 | 90 | 95 | 100 | 105 | 110 | 115 | 120 | 125 | 130 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 27.8 [FCT] | ■ | ■ | ■ | ▒ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ |
| 31.4 [DUR] | ■ | ■ | ■ | ▒ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ |
| 65 [KTB] | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ |
| 115 [ODP] | ■ | ▒ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ |

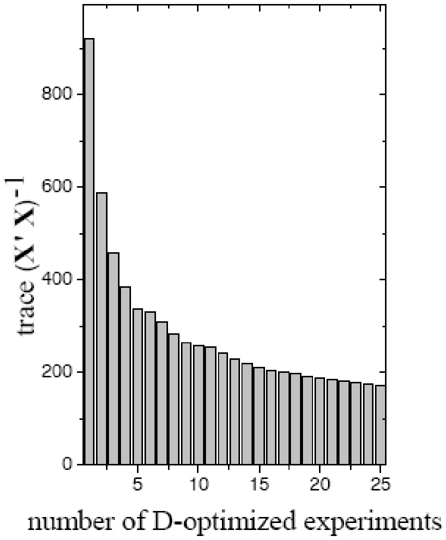
(a)

| *T* (°C) / *t* (h) | 100 | 125 | 150 | 175 | 200 | 225 | 250 | 275 | 300 | 325 | 350 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | ▒ | ▒ | ▒ | □ | □ | □ | ▒ | ▒ | ▒ | ▒ | ▒ |
| 10 | ▒ | ▒ | □ | □ | □ | □ | ▒ | ▒ | ▒ | ▒ | ■ |
| 100 | ▒ | ▒ | □ | □ | □ | □ | ▒ | ▒ | ▒ | ■ | ■ |
| 1000 | ▒ | □ | □ | □ | □ | ▒ | ▒ | ▒ | ■ | ■ | ■ |

| *T*(°C) / *t*(Ma) | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 | 55 | 60 | 65 | 70 | 75 | 80 | 85 | 90 | 95 | 100 | 105 | 110 | 115 | 120 | 125 | 130 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 27.8 [FCT] | ■ | ■ | ■ | ▒ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ |
| 31.4 [DUR] | ■ | ■ | ■ | ▒ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ |
| 65 [KTB] | ▒ | ▒ | ▒ | □ | □ | □ | □ | □ | □ | □ | □ | □ | □ | □ | □ | □ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ | ▒ |
| 115 [ODP] | ■ | ▒ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ | ■ |

(b)

Figure 2.



(a)

| $T$(°C) \ $t$(Ma) | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 | 55 | 60 | 65 | 70 | 75 | 80 | 85 | 90 | 95 | 100 | 105 | 110 | 115 | 120 | 125 | 130 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 65 [KTB] | 17 | 18 | 20 | 22 | 24 | 25 | 23 | 21 | 19 | 16 | 15 | 14 | 13 | 12 | 9 | 8 | 5 | 4 | 2 | 3 | 7 | 10 | 11 | 6 | 1 |

(b)

| $T$(°C) \ $t$(Ma) | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 | 55 | 60 | 65 | 70 | 75 | 80 | 85 | 90 | 95 | 100 | 105 | 110 | 115 | 120 | 125 | 130 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 65 [KTB] | 16 | 18 | 20 | 22 | 24 | 25 | 23 | 21 | 19 | 17 | 15 | 14 | 12 | 10 | 8 | 6 | 4 | 3 | 2 | 5 | 9 | 11 | 13 | 7 | 1 |

(c)

Figure 3.

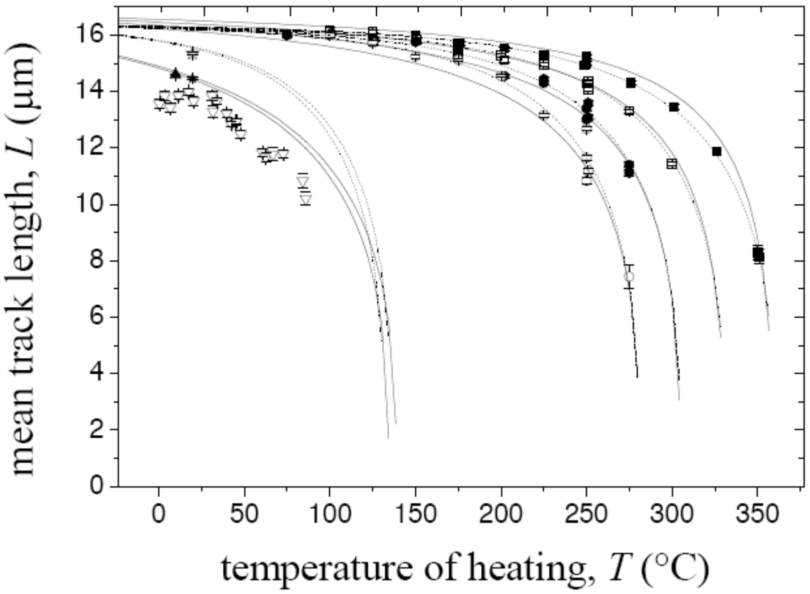

mean track length, $L$ (μm)
temperature of heating, $T$ (°C)
0
2
4
6
8
10
12
14
16
0
50
100
150
200
250
300
350

Figure 4

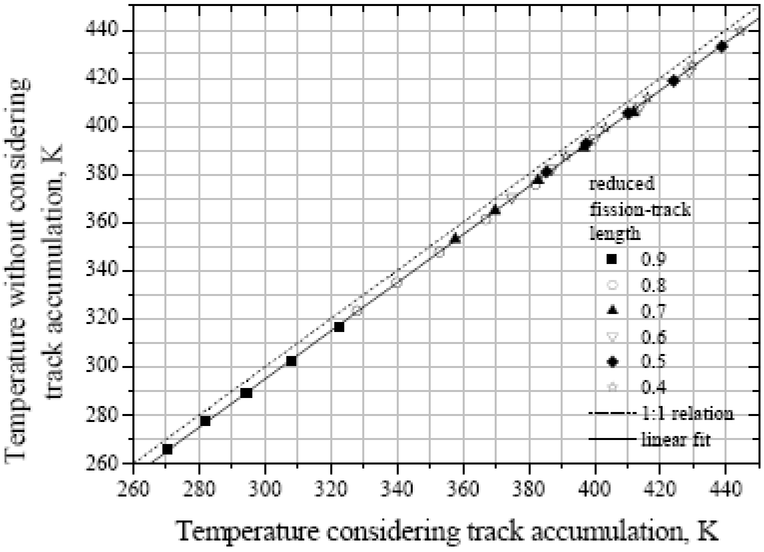
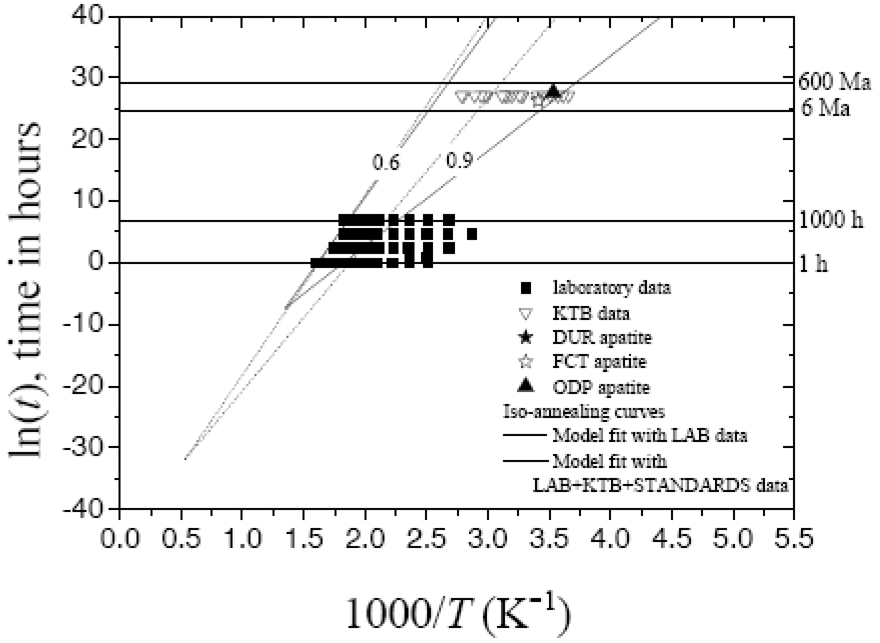

Temperature without considering track accumulation, K
440
420
400
380
360
340
320
300
280
260
260
280
300
320
340
360
380
400
420
440
Temperature considering track accumulation, K
reduced fission-track length
■ 0.9
○ 0.8
▲ 0.7
▽ 0.6
◆ 0.5
☆ 0.4
1:1 relation
linear fit

Figure 5.


ln(t), time in hours
40
30
20
10
0
-10
-20
-30
-40
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
4.5
5.0
5.5
1000/T (K-1)
600 Ma
6 Ma
1000 h
1 h
0.6
0.9
laboratory data
KTB data
DUR apatite
FCT apatite
ODP apatite
Iso-annealing curves
Model fit with LAB data
Model fit with LAB+KTB+STANDARDS data