NON-LINEAR GROWTH:

ALLOWANCE FOR ALPHA PARTICLE CONTRIBUTION

M. J. Aitken
Research Laboratory for Archaeology
Oxford University
6 Keble Road
Oxford, England

This note points out that when there has been an appreciable alpha particle contribution to the natural TL, techniques for dealing with some types of non-linear growth of TL with dose may need to be more complex than those presently employed. This need arises because the non-linearity observed for beta or gamma dosage does not represent the behaviour for alpha dosage. The familiar supralinearity observed in the low dose region for beta or gamma irradiation is absent for alpha irradiation, and, as regards the non-linearity associated with the onset of saturation it has been reported by Zimmerman (1972, figs. 6-9) that for the five substances investigated the departure from linearity occurred at an order of magnitude lower dose for beta irradiation than for alpha irradiation.

The reason for these differences in response is that whereas beta or gamma irradiation results in an effectively uniform dosage of the whole volume of the sample (except at doses low enough for the particle tracks not to be fully overlapping), for alpha irradiation the dosage is contained in discrete cylinders along the track of the particle and within the central portion of these cylinders the dosage is so high that the TL is in saturation. This is the accepted explanation for the low TL effectiveness of alpha radiation (Aitken, Tite and Fleming, 1967), firmly established by Zimmerman (1972). Thus for alpha particles the observed TL is equal to the product of the (TL per track) x (number of tracks in the sample); since each track is in virgin territory and therefore not influenced by earlier irradiation, the TL is simply proportional to the number of tracks and hence the growth of TL with dose is linear - until the level of irradiation is so heavy that there is appreciable overlap of tracks. With lightly ionizing particles, overlap begins much earlier so that the TL per track changes because of the earlier dosage received by the volume of sample concerned.

Consequences

This difference in behaviour needs to be considered whenever there is non-linear growth and an appreciable alpha contribution to the natural TL (as may be the case not only for the fine-grain technique but also for coarse grains when there is alpha activity within the grain - as in flint and calcite dating). A curve fitted to the growth characteristic obtained for beta or gamma dosage does not then give a valid representation of the growth during antiquity. Similarly when using the regeneration technique in sediment dating (method *a* of Wintle and Huntley, 1980) the paleodose derived by finding the beta (or gamma) dose needed to induce TL equal to the level of the natural TL is liable to lead to an erroneous age - as illustrated shortly by a worked example.

Supralinearity correction

First, however, a recapitulation is given of the basis for correction for supralinearity in the low dose region. As long as the growth characteristic consists of an initial upward curving portion followed by a linear portion, and the level of the natural TL is on the latter, the correction is exact.

The growth of alpha induced TL may be represented by

$$G_\alpha = \chi_\alpha D_\alpha \qquad (1)$$

where χ and D represent (TL per unit dose) and dose respectively. As regards the growth of TL induced by beta, gamma and cosmic radiation the linear portion of the characteristic may be represented by

$$G_\beta = \chi_\beta (D_\beta - I) \tag{2}$$

where the suffix β is to be taken as referring to all these three types of lightly-ionizing radiation, and, I is the intercept of the straight line on the dose axis. Obviously this representation of the characteristic is not correct other than for TL levels which lie on the linear portion.

The natural TL may be written as

$$G_N = \chi_\alpha \dot{D}_\alpha A + \chi_\beta (\dot{D}_\beta A - I) \tag{3}$$

where $\dot{D}$ represents annual dose, and A represents age. If we now introduce Q_β as the beta dose equivalent to G_N, then

$$\chi_\beta Q_\beta = G_N \tag{4}$$

In practice Q_β is obtained as the intercept of the first-glow growth line obtained by measuring the TL from undrained portions which have been given additional beta doses. As so defined Q_β does not have any physical meaning unless there is linearity in this growth. Further we define the relative alpha effectiveness as

$$k = \chi_\alpha / \chi_\beta \tag{5}$$

From (3), (4) and (5), we have

$$A = \frac{Q_\beta + I}{k\dot{D}_\alpha + \dot{D}_\beta} \tag{6}$$

This follows *exactly* and does not involve any approximation in respect of the alpha growth as is sometimes thought. The condition of applicability is that all the TL levels concerned lie on the linear portion of the growth characteristic. We may note incidentally that the analysis applies also in the case of I being negative so that (6) can be used in the case of a growth characteristic consisting of a concave-down initial curve followed by a linear portion such as might be given by two component growth in which one component saturates early.

In the preceeding χ refers to first-glow growth. In order to evaluate the intercept I it is necessary to use second-glow growth measurements. It has to be assumed that I is the same for both but it does not matter if there has been a change in slope between first-glow and second-glow. The validity of evaluating the supralinearity correction by means of a second-glow measurements is a matter of *ad hoc* test for the type of sample concerned; data relevant to this have been reported, for instance by Bowman (1975), by Fleming (1975), and by Huxtable and Murray (1980).

The regenerative technique: a worked example

To illustrate that appreciable error can arise in the case of non-linear beta (or gamma) growth we take the form of that growth to be a saturating exponential, *i.e.*, instead of equation (2) the growth is given by

$$G_\beta = \chi_\beta D_o \ (1 - \exp(-D_\beta / D_o)) \tag{7}$$

where χ_β is now the (TL per unit dose) for $D_\beta << D_o$, and D_o is a constant indicative of the dose level at which saturation effects become serious. The natural TL is now given by

$$G_N = \chi_\alpha \dot{D}_\alpha A + \chi_\beta D_o \ \{1 - \exp(-\dot{D}_\beta A / D_o)\} \tag{8}$$

In the regenerative technique the paleodose is evaluated by finding the beta (or gamma) dose necessary to induce in a bleached sample a level of TL equal to the natural TL, *i.e.*, it is the value of D_β which on substitution into (7) makes $G_\beta = G_N$. We are taking these levels of TL to be the levels above any residual unbleachable component.

For the example we take

$$\dot{D}_\beta A = \frac{1}{2} D_o \tag{9}$$

and

$$\chi_\alpha \dot{D}_\alpha = \frac{1}{2} \chi_\beta \dot{D}_\beta \tag{10}$$

These conditions give the paleodose, P_β, as

$$P_\beta = 2.06 \dot{D}_\beta A \tag{11}$$

If we now derive the age as

$$A_1 = \frac{P_\beta}{k_1 \dot{D}_\alpha + \dot{D}_\beta} \tag{12}$$

where $k_1 = \chi_\alpha/\chi_\beta$, we obtain

$$A_1 = 1.38 A \tag{13}$$

i.e., and overestimate by 38%.

An alternative way of introducing the reduced alpha effectiveness is to use $k_2 = P_\beta/P_\alpha$ where P_α is the value of D_α which when substituted into (1) gives $G_\alpha = G_N$, the latter being as in (8). For the same conditions (9) and (10) as before we find

$$P_\alpha = 1.29 \chi_\beta \dot{D}_\beta A / \chi_\alpha \tag{14}$$

and if the age is derived on the basis of

$$A_2 = \frac{P_\beta}{k_2 \dot{D}_\alpha + \dot{D}_\beta} \tag{15}$$

we obtain

$$A_1 = 1.15 A \tag{16}$$

i.e., the overestimate is less but still significant.

On the other hand if the non-linearity in the beta growth is a form of low-dose supralinearity as described by equation (2) then the age is given correctly by

$$A = \frac{P_\beta}{k \dot{D}_\alpha + \dot{D}_\beta} \tag{17}$$

where P_β is the value of D_β which on substitution into (2) makes G_β equal to the natural TL, and $k = \chi_\alpha/\chi_\beta$. Unlike correction for supralinearity in the additive dose technique it is now necessary that the TL sensitivities, as well as I, for regenerated TL are the same as during antiquity.

Conclusion

When there is appreciable non-linearity in the beta growth, as in the worked example, then except in the simple case described by equation (2), using this beta non-linearity to infer the form of the growth during antiquity is likely to give rise to a significant overestimate of the age if the alpha contribution to the TL is sizeable. At any rate in the case of the regeneration technique used in sediment dating the error is reduced by using the ratio of beta paleodose to the alpha paleodose for the relative alpha effectiveness, as in equation (15).

The preferred procedure is of course to construct a combined alpha and beta growth characteristic, the components being in proportion to the respective annual doses.

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A NEW PROPOSAL FOR THE EXPRESSION OF ALPHA EFFICIENCY IN TL DATING

S. G. E. Bowman
Research Laboratory
British Museum
London, WC1B 3DG
England

D. J. Huntley
Physics Department
Simon Fraser University
Burnaby, B.C.
Canada V5A 1S6

In dating, the first description of the thermoluminescence (TL) efficiency of alpha relative to beta radiation was in terms of the k-value (Zimmerman, 1971) defined as the ratio of TL per unit of absorbed alpha dose to the TL per unit of absorbed beta dose (by beta we mean beta or gamma throughout). For alpha particles the TL per unit dose, and therefore the k-value, varies with the alpha energy. The TL per unit of alpha particle track length, however, is nearly independent of energy. This fact led to the introduction of the presently used a-value system (Aitken and Bowman, 1975). In this note we continue in a similar vein, but rewrite the alpha particle contribution to the dose-rate in a form which we believe to be more easily understood.

For beta radiation we assume that the TL per unit of absorbed energy is a constant independent of the energy of the particle. To begin with we also assume that the TL per unit track length is the same for alpha particles of all energies. With this in mind, the natural parameter to introduce is the beta energy that yields the same amount of TL as one unit length of alpha track. That is the ratio

$$\frac{TL\ per\ unit\ alpha\ track\ length}{TL\ per\ unit\ absorbed\ beta\ energy} \tag{1}$$

which has units of J m^{-1} in the SI system.

This is closely, and very simply, related to what is actually measured in a standard laboratory determination of the relative alpha efficiency. In this, an alpha source, which delivers a known track length per unit volume per unit time, is used to irradiate a sample thinner than the individual alpha track lengths, and the resulting TL is compared to that induced by a known beta dose. This experimental ratio we define now as a new parameter

$$b = \frac{TL\ per\ unit\ alpha\text{-}track\text{-}length\text{-}per\text{-}unit\text{-}volume}{TL\ per\ unit\ of\ absorbed\ beta\ dose} \tag{2}$$

and find it is related to (1) by the sample density,

$$b = \frac{TL\ per\ unit\ track\ length}{TL\ per\ unit\ absorbed\ beta\ energy} \times \frac{1}{density} \tag{3}$$

In the SI system the unit of b is Gy m^2.

We now derive the alpha equivalent dose term in the age equation for the case where thick-source alpha counting measurements are to be used. We define λ to be the total surface alpha emission rate per unit area and time. This is related to the activities (disintegrations per unit mass and time), A_i, of the isotopes, and their corresponding alpha particle ranges, $R_i\rho$ (length x density), by

$$\lambda = \frac{1}{4} \sum_i A_i R_i \rho \tag{4}$$

The rate at which alpha track length is generated per unit volume is $\sum_i A_i R_i \rho = 4\lambda$. Thus for a sample of age T, the beta equivalent dose due to the alpha particles is

$$ED_\alpha = 4\lambda b T \tag{5}$$

To this equation two minor modifications are required. The first is due to the observation that the TL per unit track length may not be constant for alpha particles with energies less than about 2 MeV. To allow for this the right hand side of (5) must be multiplied by a dimensionless factor η (defined by equation 7 of Aitken and Bowman) for which the best estimate is still 0.90 ± 0.05 (see below).

The second modification arises because in practice it is conventional in thick-source alpha counting to count 82% of the alpha particles from the uranium chain and 85% from the thorium chain. Denoting these measured count rates by λ_U and λ_{Th} respectively we obtain finally

$$ED_\alpha = 4\eta \left\{ \frac{\lambda_U}{0.82} + \frac{\lambda_{Th}}{0.85} \right\} bT \qquad (6)$$

In practice then, one uses equation (2) to determine 'b' and equation (6) for the alpha term in the age equation.

We now relate the above 'b-value' to the 'a-value' of Aitken and Bowman. We must first note that in all the above equations one can adopt any system of units one wishes. The 'a-value', on the other hand, is defined by

$$a = \frac{x}{1300S} \qquad (7)$$

where x is the number of rads of beta irradiation that produce the same TL as 1 minute of alpha irradiation from a source of strength S μm^{-2} min^{-1}. For the same measurements equation (2) yields b (in rads μm^2) = x/S. Thus we have

$$b \ (in \ \ Gy \ \ \mu m^2) = 13a \quad . \qquad (8)$$

Thus, for example, if a = 0.1 then b = 1.3 Gy μm^2, meaning that a beta dose of 1.3 Gy yields the same TL as one normally incident alpha particle per square micrometre.

Apart from presenting the track length system in a form that we believe to be easier to assimilate, there are two significant advantages to the new proposal. The first is the absence of the arbitrary value of 1300 introduced by Aitken to make the a-value numerically the same as the old K-value at an energy of 3.7 MeV. This avoids any confusion of the track length system with that based on absorbed alpha dose. The second advantage relates to the definition of the a-value. Although the a-value is dimensionless, as defined above its evaluation requires the use of specific units i.e. μm and rads, the latter of which is not an SI unit. Only by changing the definition of the a-value could the use of grays be achieved, even so it would still be necessary to use the units specified in the new definition. The b-value, on the other hand, allows freedom of choice of units and, in particular, consistency with the SI system.

POSTSCRIPT

To this note we would like to add the following points related to the application of the b-value system.

(1) When equal uranium and thorium parent activities and full chain equilibrium are assumed, 52% of the alpha count-rate above the threshold is due to uranium and 48% to thorium. Equation (6) then becomes

$$ED_\alpha = 4.80\eta\lambda bT \qquad (9)$$

and taking $\eta = 0.9$, we have

$$ED_\alpha = 4.32\lambda bT \qquad (10)$$

(2) It has been assumed throughout that η is independent of which series predominates. Strictly equation (6) should be

$$ED_\alpha = 4\left\{\frac{\eta_U \lambda_U}{0.82} + \frac{\eta_{Th} \lambda_{Th}}{0.85}\right\} bT \qquad (11)$$

For full chain equilibrium, however, η_{Th} is only 1.5% higher than η_U and the average value of 0.90 has been used when the two chains are of approximately equal activities (Bowman, 1976).

If one chain predominates, as for example uranium in flint (Bowman et al., 1982), the small spectrum dependence of η which appears to exist, partially balances that of the threshold factor. The equivalent alpha dose for the individual series are then:

	U	*Th*
full chain	4.36λbT	4.28λbT
pre-Rn	4.44λbT	4.44λbT

The dependence on which series is dominant and on gas escape is therefore small, and for all practical purposes can be ignored (see also Aitken, 1983) when the error on η is considered. The value of η recommended for use in dating is 0.90 ± 0.05 (Bowman, 1976). This value is based on measurements on a variety of materials of TL versus alpha energy for energies up to 7.3 MeV. The error on η, considered to be systematic, relates to the variation in results for the different minerals and to the uncertainty regarding the reason for the rapid fall in TL observed in some samples for energies less than 2 MeV. This fall-off could be a genuine effect caused by decrease in energy or be due to agglomeration of grains during deposition which produce a sample thickness that exceeds the alpha particle range.

(3) In deriving the equations for the alpha equivalent dose it was implicitly assumed that in the b-value measurement all alpha particles used were sufficiently energetic that the TL was proportional to the alpha track length. The fall in TL per track length for energies below about 2 MeV necessitates the use of a source and geometry such that all alpha particles have at least 2 MeV after passing through the sample. Singhvi and Aitken (1978) showed that their alpha irradiator, in which ^{241}Am sources are used in vacuum, is satisfactory in this respect when used with one of the type AMM.7 10 μCi foil sources from the Radiochemical Centre, Amersham, UK, and provided the source to sample distance was at least 10 mm. The energy of the alpha particles from this source was 4.9 MeV. Another, stronger, source of type AMM.3 from the same company, and as normally used in the Singhvi and Aitken six-seater irradiator, emitted alphas of only 4.2 MeV and Aitken (personal communication) has pointed out that the typical energy shown for ^{241}Am foil sources in the Radiochemical catalogue is only 4 MeV. For these the 2 MeV requirement will not be satisfied by oblique alpha particles from a source at a distance of 10 mm from a sample assumed to be 8 μm thick.

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UNIVERSIDAD NACIONAL DE INGENIERIA (PERU) TL DATES - 1984 (I)

A. Pereyra Parra[1], E. López Carranza[1] and D. Lavallée[2]

The TL measurements to date pottery from Telarmachay, a shelter in the Peruvian Central Andes, were made in 1981-1982. The "fine-grains" technique of Zimmerman (1971) was used. Details about the equipment and procedure to obtain the natural and annual doses were published by López Carranza et al. (1983). First results of these dates without considerations of moisture were reported by Pereyra et al. (1982).

For the annual dose, radioactive analyses were carried out on the pottery and soil by alpha counting (U and Th) and flame photometry (K), and the calculations were made using the conversion factors of Bell (1977). If the ratio between the sealed (α_1) and unsealed (α_o) alpha counts, α_1/α_o, was greater than 1.05, the value $(\alpha_1 + \alpha_o)/2$ was used. In other cases α_o was taken. For the soil, the stone content was not considered and the ratio α_1/α_o was 1.17. An alpha efficiency (1) of 0.15 with an overall uncertainty $\delta a = 20\%$ was assumed for both sherd and soil. 15 mrads/yr was used as the contribution of the cosmic radiation to the dose-rate. To take account of the moisture content, which was very high in the excavation site, saturation water uptake was considered for soil and sherds; soil wt. sat./soil wt. dry was equal to 1.4. Anomalous fading was not studied in any case. The error assessment of Aitken (1976) was used, taking zero for the uncertainties σ_7 (radon emanation) and σ_8 (wetness estimate). The TL and ^{14}C dates are in years before A.D. 1980 and the TL errors appear within parenthesis.

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[1] Departamento de Fisica, Universidad Nacional de Ingenieria, Casilla 1301, Lima, Perú.

[2] URA no. 25, CNRS, 27 rue Paul Bert, 94204 IVRY, France.

ARCHAEOLOGIC SAMPLES

TELARMACHAY (Province of Tarma, Department of Junín, 11 ° 11'S, 75 ° 52'W) Peru

This archaeologic site is located at an altitude of 4,420 m and was occupied between 7000 B.C. (lithic period) and 170 B.C. (formative period). The excavation was realized by D. Lavallée and her archaeologic research group within the Archaeologic Project Junin-Palcamayo of the URA no. 25, CNRS-France (Lavallée et al. 1975; Lavallée 1977). The ^{14}C dates on associated charcoal were measured in the Centre des Faibles Radioactivités, CNRS-CEA, France.

UNI-TL-6: 2570 (---, ±310), 590 B.C.

Pottery: 1468-30

Comments- Natural dose: 1310 rads (I=70), δQ =9%, plateau $\simeq$ 70 ° C.
Annual dose: 0.51 rads/yr with α_1/α_0=1 for the fragment, sherd wt. sat./dry =1.06 and a =0.15, δa = 20%.
^{14}C date for the context is 2220 ± 100 (GIF 3772).

UNI-TL-7 2400 (---, ±260), 420 B.C.

Pottery: 1468-57

Comments- Natural dose: 670 rads (I=50), δQ = 7%, plateau $\simeq$ 60 ° C.
Annual dose: 0.28 rads/yr with α_1/α_0=1 for the fragment sherd wt. sat./dry = 1.06 and a = 0.15, δa = 20%.
^{14}C date for the context is 2220 + 100 (GIF 3772).

UNI-TL-8 1820 (---, ±240), A.D. 160

Pottery: 1478-117

Comments- Natural dose: 1110 rads (I=40), δQ = 8%, plateau $\simeq$ 70 ° C.
Annual dose: 0.61 rads/yr with α_1/α_0=1 for the fragment, sherd wt. sat./dry = 1.06 and a = 0.15, δa = 20%.
^{14}C date for the context is 2310 ± 100 (GIF 3773).

UNI-TL9 2890 (---, ±430), 910 B.C.

Pottery: 1643

Comments- Natural dose: 1010 rads (I=10), δQ = 9%, plateau $\simeq$ 80 ° C.
Annual dose: 0.35 rads/yr with α_1/α_0 =1 for the fragment, sherd wt. sat./dry = 1.13 and a = 0.15, δa = 20%.
^{14}C date for the context is 2630 ± 90 (GIF 4187).

UNI-TL-10 3670 (---, ±620), 1690 B.C.

Pottery: 1614-41

Comments- Natural dose: 1580 rads (I=30), δQ = 6%, plateau $\simeq$ 60 ° C.
Annual dose: 0.43 rads/yr with α_1/α_o = 1.03 for the fragment, sherd wt. sat./dry = 1.13 and a = 0.15, δa = 20%.
^{14}C date for the context is 3440 ± 100 (GIF 4188).

UNI-TL-11 3240 (---, ±420), 1260 B.C.

Pottery: 1721

Comments- Natural dose: 940 rads (I=110), δQ =11%, plateau $\simeq$ 70 ° C.
Annual dose: 0.29 rads/yr with α_1/α_o = 1.09 for the fragment, sherd wt. sat./dry = 1.11 and a = 0.15, δa = 20%.
^{14}C date for the context is 3660 ± 100 (GIF 4833).

POLONIUM LOSS FROM GLASSES PREPARED FOR ALPHA COUNTING

R. A. Akber, J. T. Hutton and J. R. Prescott

Physics Department, University, Adelaide, Australia 5001

Solution of the Age Equation in thermoluminescence dating technique requires estimates of uranium and thorium in sherd or sediment samples. We have previously reported on thick source alpha counting using fused glass discs for this purpose (Prescott and Jensen, 1980; Jensen and Prescott, 1983). A 1:1 or 1:2 proportion by weight of finely ground sample and anhydrous lithium borate is heated to above 1000 ° C for 15 to 20 minutes in a platinum-gold crucible. The melt is then poured to form a disc which is slowly cooled to room temperature. We follow the procedure described by Norrish and Hutton (1969) for the preparation of discs for XRF analyses. Further details of the recipe and the merits of the method can be seen in the references cited. Our experience in the use of these discs now extends over several years and a critique of the over-all success and limitations will be compiled in the near future. However it would be beneficial to publish, albeit briefly, some of our latest observations in advance. These concern the loss of activity during disc preparation which, if not accounted for, may lead to error in U/Th content calculations or wrong estimates of disequilibrium in the decay series.

At the elevated temperatures achieved during disc fusion, the diffusion out of radon gas is quite well understood and can be corrected for (Jensen and Prescott, 1983). Now we have data which suggests the loss of polonium activity as well. Polonium in its metallic form boils at 962 ° C while all its known compounds either boil or decompose well below this temperature (Handbook of Chemistry and Physics, 1982). Of all the Polonium isotopes in the three natural radioactive decay series, Po-210 in the U-238 series is the only one of significance. It has a half life of 138 days and is a 5.3 MeV alpha emitter. We were able to pick up 5.3 MeV alpha activity on the lid of the crucible in which one of the uranium standard glass discs had been fused. The activity decayed with the half life of Po-210. We also observed the build-up Po-210 activity in a uranium standard disc with the passage of time. These observations explain our previously-reported findings of a consistent difference in the (activity-in-glass/activity-in-sample) ratios for uranium and thorium standards (Jensen and Prescott, 1983).

Further interesting results on the polonium activity loss due to sample heating and the corresponding time-dependent corrections based on alpha range-energy relationships are presently being compiled and will be reported shortly.

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