The use of a single aliquot method for intercalibration between radioactive sources.

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Intercalibration between radioactive sources is an onerous, yet essential, part of TL and ESR work. A simple method involving infrared stimulated luminescence (IRSL) is described which can be performed using a single aliquot.

The method is a development of the single aliquot technique described in Duller (1991) for the determination of equivalent dose (ED). Using multiple IRSL measurements on a single disc, with bleaching and dosing of the sample between each measurement, the ED could be determined using a regeneration method. However, using this procedure it was found that if the same dose were repeated several times the IRSL signal progressively increased. This effect was most pronounced when using IR to bleach the sample between each dose, though it was seen to a smaller extent when using a SOL 2 solar simulator. This phenomenon was ascribed to competition for electrons during irradiation. As successive doses were added, part of the trapped charge population was not removed by bleaching, and the build up of this remnant charge increased the sensitivity of the IRSL signal to dose.

To exemplify the problems of accuracy, rather than precision, associated with using IR to bleach a sample between each regeneration step, six discs of potassium feldspar were used to calibrate the ß-source within the first Risø reader at Aberystwyth against itself. In this way the correct answer was known and so any inaccuracies could be easily seen. Potassium feldspar (<2.58 g cm^{-3}) from sample GDNZ 17 (a New Zealand dune sand) with a grain size of 180-211 µm was used throughout the experiments.

The six discs were first heated to 450 °C at 5 °C/s. After cooling they were given a ß-irradiation for 600s. After preheating at 220 °C for 10 minutes the samples had their IRSL measured for 100 seconds at 50 °C (filters used were a BG-39 and 5-60). They were then exposed to IR for 15 minutes (power approx. 40 mW/cm^2) at 50 °C to reduce the IRSL signal to a residual level (<1% of the 'natural' signal due to 600 s of ß-irradiation) before being irradiated by the same Risø ß-source, preheated and their IRSL remeasured. Further bleaching and irradiation was repeated to build up a response curve. Irradiations of 0, 200, 400, 600, 800, 1000, 0, 600 and 1000 s were used. Figure 1(a) shows that the growth curve is supra-linear, and that the ED generated (533 ± 4 s) is significantly lower than the expected 600s (table 1(a)). The precision of the result is excellent (±1%), but there is an overestimation of the true source strength by 13%. The repeat determinations of the I_0, I_0+600 s, and I_0+1000 s points all show an increase in sensitivity of the sample aliquots.

An alternative approach is to heat the aliquots between each regeneration step. To test this, the same procedure as described above was used, calibrating the first Risø ß-source against itself, but the samples were heated to 450 °C between each phase of regeneration. Identical doses were used as before, and a typical growth curve is shown in figure 1(b). The growth curve is linear and the ED generated is 588 ± 13 s (table 1(b)), within 1 sigma of the true figure. Repeat measurements of three of the data points fall on the same growth curve.

To demonstrate the use of this method two intercalibrations between the two Risø reader ß-sources were performed, one using IR, and the other heating between each step to define the residual level. Figure 2 shows the two growth curves generated.

The curve in figure 2(a) was generated using the same procedure as that used for figure 1(a). After heating the discs to 450 °C at 5 °C/s a ß-irradiation of 600s was given in the first Risø reader. Irradiations for 0, 200, 400, 600, 800, 1000, 0, and 600 s were given with the second Risø reader ß-source. The form of the growth curve is supra-linear, and when the I_0 and I_0+600 s measurements were repeated (after all other measurements had been made) they showed a significant increase in IRSL signal. The precision of the results generated using this method is excellent (table 2(a)), but the accuracy poor.

The problem observed above is related to the remnant trapped charge. Heating the sample to 450 °C removes any residual TL or IRSL signal. Figure 2(b) shows a growth curve generated using identical analytical conditions to those for figure 2(a), except that the sample was heated to 450 °C at 5 °C/s between each regeneration phase instead of being exposed to IR. The growth curve is now linear, and when measurements are repeated (after irradiations for 0, 600 and 1000 s) they lie exactly on the growth curve.

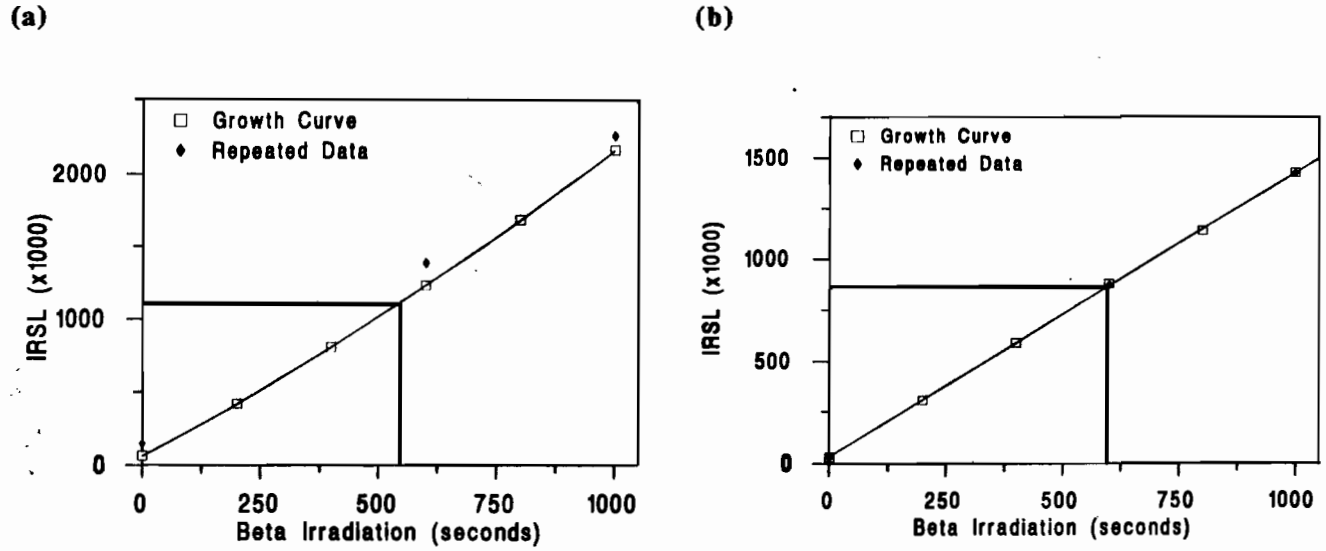

Figure 1.
Intercalibration of the Risø I β-source against itself (a) exposing the samples to 15 minutes of IR and (b) heating the samples to 450 °C at 5 °C/s, between each phase of regeneration. The IRSL signal has been integrated over the 100 s of measurement. The data points at I_0, I_0+600 s and I_0+1000 s were repeated after all other measurements were made.

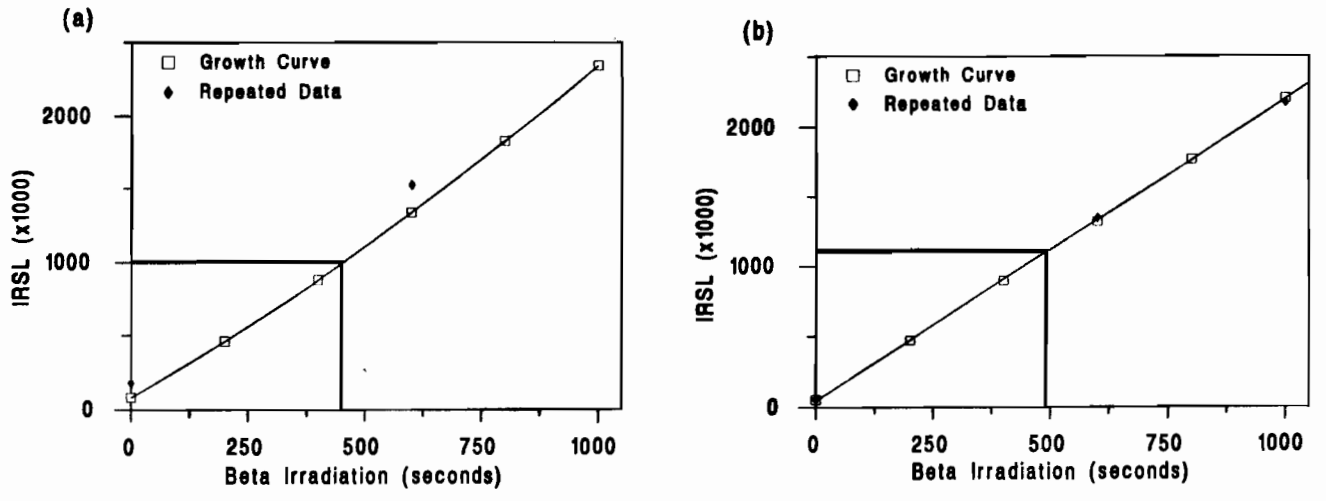

Figure 2.
Intercalibration between the Risø I and Risø II beta sources (a) exposing the samples to 15 minutes of IR and (b) heating the samples to 450 °C at 5 °C/s, between each phase of regeneration. The IRSL signal has been integrated over the 100s of measurement. The data points at I_0 and I_0+600 s (also I_0+1000 s for fig. 2(b)) were repeated after all other measurements were made.

Table 1(a).
Intercalibration of the first Risø ß-source against itself using IR to reduce the IRSL signal to a residual level.

Disc No.	ED (s) [a]	Source ratio[b]
1	535	1.12
5	533	1.13
9	527	1.14
13	535	1.12
17	539	1.11
21	529	1.13

Average = 1.13 ± 0.01

Table 1(b).
Intercalibration of the first Risø ß-source against itself heating samples to 450 °C to reduce the IRSL signal to a residual level.

Disc No.	ED (s) [a]	Source ratio[b]
4	604	0.99
8	565	1.06
12	596	1.01
16	587	1.02
20	583	1.03
24	592	1.01

Average = 1.02 ± 0.02

Notes
(a) 'ED' is the irradiation time interpolated from the growth curve to be necessary to match the signal produced by the initial 600 s of irradiation with the Risø ß-source. A linear fit was used.
(b) By definition this ratio should be 1.00 since the ß-source is being calibrated against itself.

Table 2(a).
Intercalibration between the two Risø ß-sources using IR to reduce the IRSL signal to a residual level.

Disc No.	ED (s) [a]	Source ratio[b]
14	440	1.36
16	442	1.36
18	428	1.40
20	429	1.40
22	430	1.40
24	429	1.40

Average = 1.39 ± 0.02

Table 2(b).
Intercalibration between the two Risø ß-sources heating samples to 450 °C to reduce the IRSL signal to a residual level.

Disc No.	ED (s) [a]	Source ratio[b]
1	505	1.19
5	515	1.17
9	494	1.22
13	493	1.22
17	483	1.24
21	488	1.23

Average = 1.21 ± 0.03

Notes
(a) 'ED' is the irradiation time using the second Risø source necessary to match the signal produced by 600 s of irradiation with the first Risø source. A linear fit was used.
(b) The source ratio is the ratio of Risø II/Risø I source strengths.

All samples gave a flat plot of 'ED' against IRSL exposure time for the 100 s period analyzed. The signal from the irradiation with the first Risø ß-source could be compared to the growth curve generated by the second ß-source using the regeneration analysis program written by R.Grün. The precision here is also excellent (table 2(b)), but the result is significantly different from that in table 2(a). This second method is considered to give a reliable ratio between the two sources.

Any sensitivity changes that may result from heating the sample occur before the 'test' dose is given to the sample. Subsequent heating of the sample causes no further changes in sensitivity but removes the residual trapped charge population within the crystal. This method has two major advantages over other methods of intercalibration, first that it is very quick and simple to perform, and hence one may reasonably perform many determinations to get a precise value. Secondly, the calibration is determined for the actual material being dated, not for calcium fluoride or some other phosphor. Hence there is no need for further adjustment of the data. This approach is appropriate for the basic calibration using a gamma source.

Reference

Duller G.A.T., (1991) Equivalent dose determination using single aliquots. *Nuclear Tracks and Radiation Measurements* **18**, 371-378.

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PI Vagn Mejdahl

I was glad to see this note on intercalibration of sources and hope it will mark the beginning of a thorough discussion on this important problem.

The extreme precision that can be achieved with the single aliquot-regeneration technique introduced by Duller (1991) makes it a very attractive method. However, source calibration using IR stimulation appears to be even more complex than with TL.

The paper shows convincingly that part of the problem is the occurrence of a sensitivity change when only IR irradiation is used as "anneal" between each measurement step. The fact that the change can be eliminated by heating the samples to 450 °C. corroborates the suggestion made earlier by Duller (1991) that the sensitivity change may be caused by the change in distribution of trapped charges resulting from the gradual build up of charges in traps not affected by the IR irradiation.

I agree that the procedure described should ensure a correct intercomparison of two beta sources. The next, closely related question is then how should one proceed in actual dating using the single aliquot-regeneration method. Because there is the same problem with sensitivity change it seems necessary to apply the same heating anneal between each measurement step. However, because no initial heating is possible, one is faced with the problem of a sensitivity change as a result of the first heating, especially for sediments.

Perhaps even more important, it seems to me that the main problem, namely the intercomparison of beta and gamma sources remains unsolved. At the 1991 UK meeting in Aberystwyth I reported that when we used the single aliquot-regeneration technique with IR stimulation for intercomparing our beta and gamma sources, we obtained an apparent beta dose rate of 3.60 Gy/min as compared with 2.40 Gy/min when using TL. This has turned out to be a very consistent feature. However, when using IR stimulation with multiple samples as with TL, I obtain precisely the same figure as with TL. The results of the experiments described by Duller suggest that perhaps 13% of this 50% difference may be ascribed to sensitivity changes, but this still leaves a puzzling 37% unexplained.

Applying IR-OSL and TL for dating some Late-Glacial samples of well-known ages we obtained correct results when using the respective calibration values. Perhaps the lesson to be learned is that one should use the same procedure for calibration and dating.

Author's Reply

I attempted in this short note to restrict myself to a discussion of the use of a single aliquot regeneration method for intercalibration between radioactive sources. I deliberately avoided the far more complex issues concerning the use of single aliquots for equivalent dose determination of sedimentary materials since I do not see the approach used here being applicable to these materials (unless it can be shown that heating a sample to 450 °C causes no change in its sensitivity to irradiation!). Furthermore I would suggest that using the same, inaccurate, procedure for calibration and dating, as suggested by Dr Mejdahl, may fortuitously produce a correct result, but should not be relied upon for routine dating.

I agree entirely with Dr Mejdahl that it is now vital to show whether this method provides results consistent with those from TL measurements for the calibration between a gamma source and a beta source.