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High-precision natural dose rate estimates through beta counting

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ABSTRACT

The beta-particle emission from a sediment or rock sample can be measured very precisely using beta-counting instruments. The observed count rate is largely a function of the radionuclide concentration in the sample, so has the potential to provide a precise estimate of the natural radiation dose rate. However, the count rate is also sensitive to the attenuation of beta particles in the sample, and the relative proportions of the different radionuclide sources. Here we devise a correction for the self-attenuation effect using dilution analysis, and show that imprecise prior knowledge of radionuclide activity is sufficient for calculation of an accurate combined beta-plus-gamma dry dose rate. The method is tested on a selection of archive samples, and compared with results from high-resolution gamma-spectrometry. We show that with counting uncertainty ~2%, and calibration uncertainty ~2%, the total random uncertainty of the beta-plus-gamma dry dose rate is less than 3%. For most natural sediments, this level of precision equal to, or better than, that obtainable with other methods.

1. Introduction

Trapped-charge dating methods are in common use in earth science and archaeology for dating sediment, rock surfaces and artefacts. These methods seek to determine the radiation dose absorbed by a natural dosimeter (usually grains of quartz or feldspar) since they were last buried, using some variety of luminescence or electron spin resonance measurement. The age of the sample is estimated by dividing the laboratory estimate of the absorbed dose (the equivalent dose, D_e) by the rate that radiation was absorbed during the burial period (the dose rate, $\mathring{D}$).

Given the importance of $\mathring{D}$ in the age equation, an accurate and precise estimate of the dose rate is essential for all these dating methods. Most estimates of the dose rate are derived from the measured mass or activity concentrations of K, U and Th (and/or progeny). These concentrations are obtained from one or more of several analytical techniques, including Inductively Coupled Plasma Mass Spectrometry (ICP-MS), high-resolution gamma spectrometry (HRGS) or Neutron Activation Analysis (NAA). Concentrations are then converted to the 'infinite-matrix' dose rate (i.e. the dose rate assuming that, within the volume of interest, all energy released is absorbed) using conversion factors which are sums of the total energy released following each parent disintegration (e.g. Guérin et al., 2011).

While it is normal to find that published D_e measurements are provided together with in-lab quality control (e.g. dose recovery test, pre-heat plateau), it is rare to find such validation steps for dose rate measurements. In fact the reproducibility of dose rate estimates between laboratories is poor, as shown in a recent inter-comparison (Murray et al., 2015), and there is little information about the absolute accuracy of such measurements. In that study a homogenised sample of beach-ridge sand was dated by different luminescence laboratories around the world. With each laboratory determining the dose rate independently, using various measurement techniques, the relative standard deviation (RSD) of 20 dry dose rates was 12%. Variability in the radionuclide estimates were much larger, with overall RSD of 15% for potassium, 22% for uranium, and 59% for thorium.

An alternative approach to determining mass or activity concentrations is to use some form of integral particle or photon counting. Beta (and alpha) counting methods do not differentiate the source particles by energy, so cannot easily be used to estimate the individual radionuclide activities. Instead, all particles that produce a pulse that exceeds some threshold are detected, so that the observed count rate is some function of the combined activity concentration of all radionuclide sources. With discrimination between energy sources, such integral counting instruments are capable of extremely high-precision measurements (e.g. Sanderson, 1988). A low-level beta counter has been available from Risø for many years, based on a set of 5 thin-

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window Geiger-Müller counters with a common anti-coincidence guard (Bøtter-Jensen and Mejdahl, 1985). Using this instrument, a typical sample might provide a net beta count rate of ~200 counts per hour. With five sub-samples measured simultaneously for 24 h, the relative standard error on the count rate will be 0.65%.

However, the beta count rate is not a simple function of the sample dose rate. Assuming the decay chains are in equilibrium, the beta count rate will be mainly dependent on:

- The sensitivity of each detector
- The background count rate
- The self-attenuation within each sample (a function of the average stopping power)
- The relative proportions of the radionuclide sources

If all these factors were correctly accounted for during measurement or analysis, then the beta count rate could be used to estimate the present-day dry dose rate in the sample with a random uncertainty equal to the counting precision of the instrumental measurement (i.e. better than 1%). If it were reasonable to assume the decay chains to be in secular equilibrium, this level of precision would be equal to, or better than, that provided by the best alternative method. In this paper, we describe an approach that aims at this target. We describe the rationale behind the study, discuss how to account for the self-attenuation of a sample, and show that the conversion from count rate to dose rate is most precise when combined with some prior information on the radionuclide composition. The protocol is tested on 11 samples which have paired measurements obtained from high resolution gamma spectrometry (HRGS).

2. Rationale

If all sources of measurement error have been accounted for (see below), the observed beta count rate is dependent only on the activity concentrations of the radionuclides. The dose rate $\mathring{D}$ (Gy ka^{-1}) is then derived by:

$$\mathring{D} = \frac{b}{q} \tag{1}$$

where b is the sample count rate (ks^{-1}), and q is the count rate conversion factor ks^{-1}/(Gy ka^{-1}). The value of q is unique to each sample, because it depends on the relative proportions of the radionuclides, hence:

$$q = \sum_{i=1}^{3} p_i w_i \tag{2}$$

where p is the proportion of the dose rate derived from the radionuclide source (i = [K, U, Th]), and w is the count-rate conversion factor for each isotope at the specific activity in the given source (ks^{-1}/(Gy ka^{-1})). w can be determined from measurements of standards (see section 3.3 and Table 1). The radionuclide proportions, p, are unknown and must be estimated.

In order to define q, we must use prior information on the radionuclides, i.e. we must already know at least the relative activity concentrations in the sample. If these concentrations are already well-known, then there is, of course, no need for beta counting. But we show below that even poorly known activities can lead to accurate beta count conversion to dose rate, with its associated additional benefit of high precision. Consider first a hypothetical measurement of the Skagen inter-comparison sample (Murray et al., 2015). Suppose that the observed inter-lab mean of the ^{40}K (333 Bq kg^{-1}), ^{238}U (5 Bq kg^{-1}) and ^{232}Th (3.9 Bq kg^{-1}) activity concentrations are accurate. The inferred total beta and gamma dose rate, using the conversion factors of Guérin et al. (2011) is 1.28 Gy ka^{-1}, and the beta count rate we would observe (with the instruments described here, calculated using Table 1) is 37.6 ks^{-1}. To derive the dose rate from the beta count rate, we use the known concentrations to evaluate p and hence q and $\mathring{D}$, and obtain–of course–a perfect dose-rate estimate of 1.28 Gy ka^{-1}. However, suppose instead that the prior estimate of ^{40}K underestimates the true value by 20%, with ^{238}U and ^{232}Th activities still correct. The inferred dose rate derived directly using the conversion factors of Guérin et al. (2011) is now 1.06 Gy ka^{-1}, a 17% underestimate of the true value. However, if the erroneous activities are used instead to derive a revised beta-counter count rate conversion factor q via equation (2), then the dose-rate estimate based on beta counting is 1.30 Gy ka^{-1}, only a 1.3% overestimate of the true value. Remarkably, the beta-counter-derived dose rate has an almost negligible error, even though the count-rate conversion factor was derived using erroneous activity estimates.

Table 1
Calibration count rates by radionuclide source (K, U, Th), for a Riso GM-25-5 low-level gas-flow multicounter, calculated from measured count rates of standards (section 3.3) and the conversion factors of Guérin et al. (2011). Count rates have been corrected for stopping power, and normalised to the sensitivity of a single, specific, detector.

Source	Counts ks^{-1}/(Gy ka^{-1})		
	β	γ	$\beta + \gamma$
K	41.2 ± 0.7	132.1 ± 2.3	31.4 ± 0.5
U	33.8 ± 0.7	44.2 ± 0.9	19.2 ± 0.4
Th	35.1 ± 0.6	20.3 ± 0.4	12.9 ± 0.2

This effect comes from transforming the raw activity concentration into relative proportions, creating an anti-correlation in the calculation of q. If the count rate expected from one source is wrong (e.g. p_1 is underestimated), the effect on q will be partially cancelled out by implicit overestimates of the other sources (p_2 and p_3). The portion of the error that is cancelled out will depend on the similarity of the w_i, the radionuclide-specific calibration factors (Table 1), so depends on whether we are interested in the beta, gamma, or beta and gamma dose rates.

We can explore the issue further using simulations of the error transformation (Fig. 1). We first specify the activity of ^{40}K, ^{238}U, and ^{232}Th for a representative range of samples, which we take from 344 samples measured with HRGS and summarised by Ankjærgaard and Murray (2007); these values are treated as known. For each sample, we create a 'measured' set of activities (^{40}K, ^{238}U, and ^{232}Th) by drawing values from a normal distribution with a mean of the known activity and standard deviation of 10%. The directly converted dose rates (using the Guérin et al. (2011) conversion factors) are shown in Fig. 1a–c, normalised to the known dose rates. The 10% randomisation of each radionuclide leads to ~7.5% uncertainty on the calculated beta-plus-gamma dose rate (Fig. 1c) when derived directly from concentrations, and similar for the beta and gamma dose rates separately (Fig. 1a and b). The lower two rows of Fig. 1 show the simulated beta-counter dose rates. In Fig. 1d–f, these are calculated by using the same count-rate conversion factor q for each sample (i.e. using a single, typical, sample to calibrate all other samples– determined here by the average of all 344 samples). When predicting the beta dose only, the error introduced is very modest (RSD = 2.2%), due to the similarity of the radionuclide conversion factors (Table 1, column 1). When predicting the gamma or beta-plus-gamma dose rate, a much larger error is introduced (~18% and ~8%, respectively). In contrast, Fig. 1g–i shows the effect of using a sample-specific count-rate conversion factor (q), after plugging the erroneous activity estimates into equation (2). The beta dose rate is very precise (RSD = 0.4%), and the RSD in the combined beta-plus-gamma dose rate is just 1.5%.

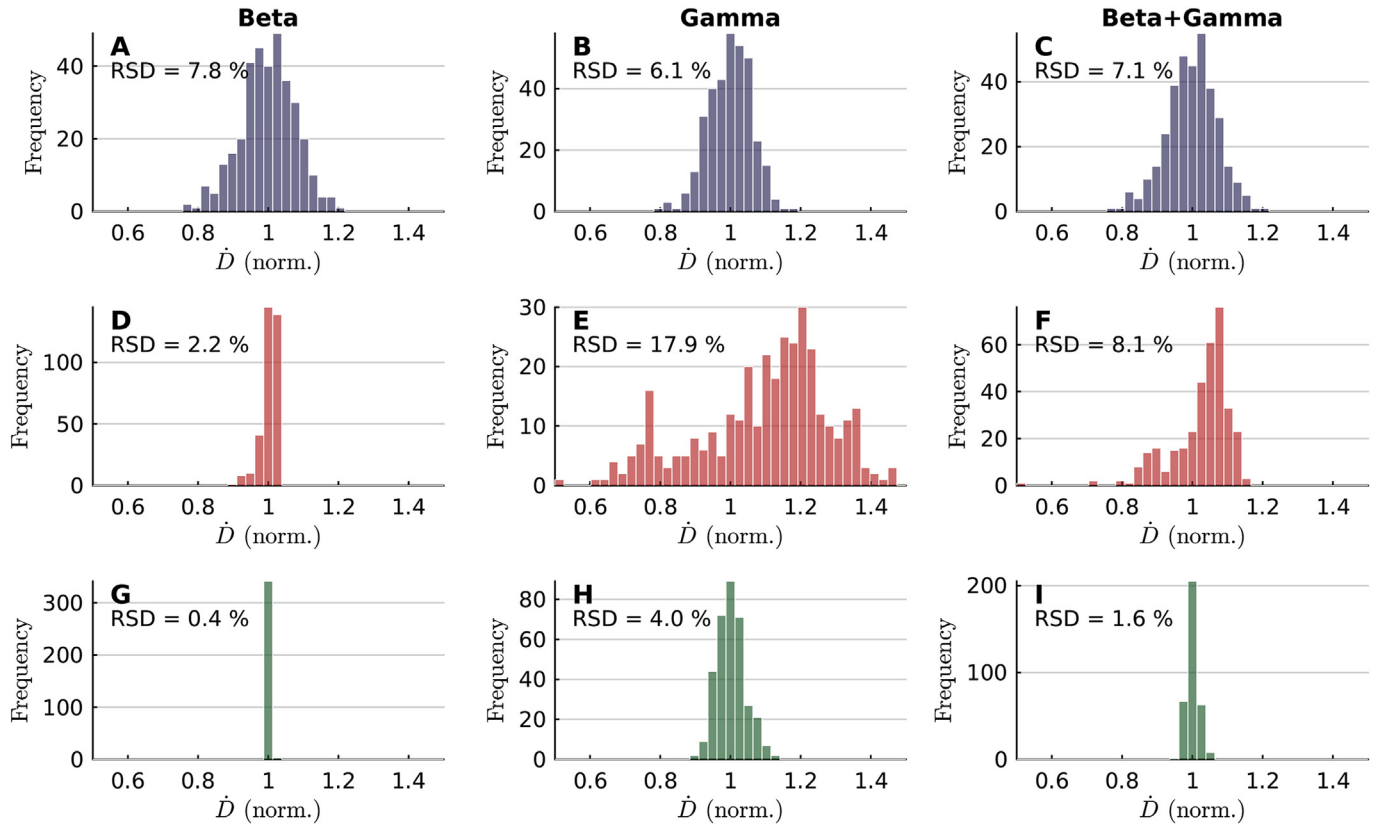

Fig. 1. Simulation showing the effect of prior information on the beta-counter derived dose rates. Top row: known radionuclide activities have been randomised by 10% each; the beta, gamma and beta-plus-gamma dry dose rates are calculated directly via conversion factors, and normalised to the known dose rate. Middle row: simulated beta-counter derived dose rates using a single count-rate conversion factor for all samples (i.e. calibrating against a 'typical' natural sediment). Bottom row: beta-counter derived dose rates calculated using a sample-specific count rate conversion factor, determined from the imprecise radionuclide estimates. RSD = relative standard deviation. Simulations do not include any other sources of error (e.g. counting statistics).

3. Measurement

3.1. Instrumentation

The measurement apparatus is a Risø GM-25-5 low-level gas-flow multicounter (Bøtter-Jensen and Mejdahl, 1985). Most measurements were carried out on two instruments, although several instruments were used in total. Each instrument has five GM detectors, which may have different sensitivities and background count rates. The background count rates were measured using samples of pure wax (see below), over ~65 h, and range from 2.2 to 4.2 ks^{-1}. The relative sensitivity of each detector was assessed using (relatively) high activity samples – a set of ^{99}Tc sources, and some of the standards described below. Detector sensitivities vary by ~40%; the oldest instrument (detectors 1–5) has detectors of the lowest and most variable sensitivity (Fig. 2). For all subsequent measurements, the observed count rates are corrected for the detector background and sensitivity.

Sub-samples are prepared by mixing powdered sample with a high-viscosity wax (Bottle wax, blend 1944, British Wax Refining Company). The mixing takes place on a hot plate; the hot mixture is pasted into a cool mould and the excess is removed. Embedding the samples in wax has several purposes: it prevents radon gas from escaping, provides identifiable and re-measureable sub-samples with an indefinite storage lifetime, and permits the stopping-power correction described below.

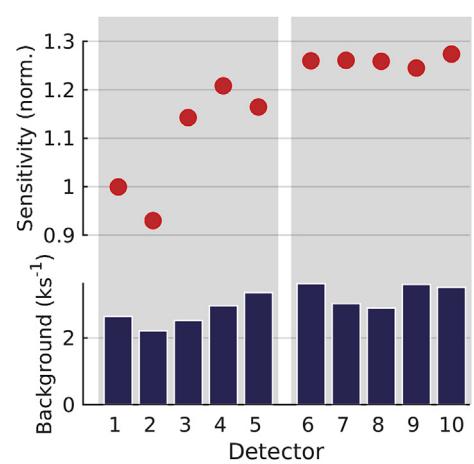

Fig. 2. Sensitivity and background measurements on two instruments, each with five detectors. From this data, detector-specific corrections are applied to subsequent measurements.

3.2. Stopping power

The number of beta particles reaching the detector is affected by the self-attenuation of the sample. By design, the sample depth (5 mm) is infinitely thick with respect to the range of beta particles in the sample material (~2 mm). In consequence, for a given atomic number, the sample density has no effect on the count rate: for a given activity (Bq kg^{-1}), a denser sample will stop more beta particles from reaching the detector, but will also contain more sources within the sample volume–effects which cancel out exactly. However, the attenuation of beta particles is also dependent on the atomic number of the material, and this can be illustrated with a Monte Carlo simulation (Fig. 3). Using Geant4 (Agostinelli et al., 2003; Allison et al., 2006), we simulated ^{40}K disintegrations in a source region identical to the sample geometry

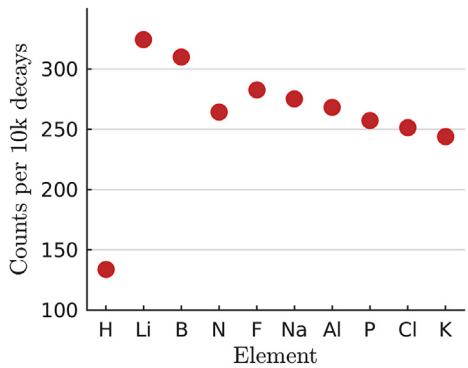

Fig. 3. Z-dependence of the beta count rate for a ^{40}K source, simulated using Geant4. Sample material is the pure element, with fixed, arbitrary density.

(cylinder of 25 mm diameter and 5 mm depth). The sample material was specified as a pure element from hydrogen to potassium, and sample density was fixed at 2.66 g cm^{-3} to maintain the infinitely thick source. The count rate was defined by the number of electrons leaving the upper surface of the cylinder. Two main trends can be seen; an increase in the count rate for sample materials beyond hydrogen, caused by the increase in relative beta scattering; second, a decrease in the count rate with atomic number, due to the Z-dependency of in the beta stopping power equation.

The Z-dependence of stopping power means that the count rate from a sample is sensitive to its elemental composition. Although clastic sediment and rocks are composed chiefly of O and Si, there can be large differences in the proportions of other major elements—e.g. Al, Ca, Na, Fe, K. Note, however, that the wax composition is about 15% H by mass, with the rest C, and so the count rate given a pure wax stopping power will be less than for the pure sample for most clastic sediment or rock. Moreover, the wax composition is consistent between samples. To remove the effects of the stopping-power differences between samples, we need only normalise to pure wax, i.e. estimate the sample count rate that would be observed if the sample composition was the same as wax. This can be achieved by varying the proportion of the wax-sediment mixture; the observed count rates are then corrected for activity (divided by the proportion of sediment, x), and plotted against x (Fig. 4). If a dependence on x can be seen, then the stopping power of the sediment differs from that of the wax. When the data is fitted with a linear function: (3)

$$y = ax + b \tag{3}$$

then b is the inferred count rate (ks^{-1}) from the sample, normalised to a pure wax matrix, and is independent of the sediment stopping power; b can then be used to calculate the dose rate via equation (1). In addition, the relative gradient of the trend can be used to investigate differences in stopping power between samples. A useful statistic for this purpose is given by $y_{(x=1)}/y_{(x=0)}$, i.e. the inferred count rate for the pure sample compared to the inferred count rate for wax of the same activity. When x is expressed as a proportion, this statistic equates to $(a+b)/b$. Here, we evaluate the fit with a Bayesian routine written with *Stan* (Carpenter et al., 2017) that allows for overdispersion in the data, provides an estimate of uncertainty for b, and accounts for the correlated uncertainties in the calculation of the stopping-power statistic.

3.3. Radionuclide calibration

Source calibration can be performed using recognised standards or isometric substances. We used pure K_2SO_4, a uranium ore (NRCAN BL-5) mixed with quartz sand, and $Th(NO_3)_4$ .4H_20 mixed with quartz sand. The uranium ore has a well-defined U activity, and is known to be in secular equilibrium down to ^{226}Ra (https://www.nrcan.gc.ca/mining-materials/certified-reference-materials/certificate-price-list/8115); mixing with wax guarantees full ^{222}Rn retention. The thorium nitrate was synthesised several decades ago and so equilibrium is assured. Count rates for the standards were determined using the same measurement procedure as the samples, i.e. wax-embedded and stopping power corrected. The known activities and the conversion factors of Guérin et al. (2011) were used to derive the count rate per Gy ka^{-1} (Table 1).

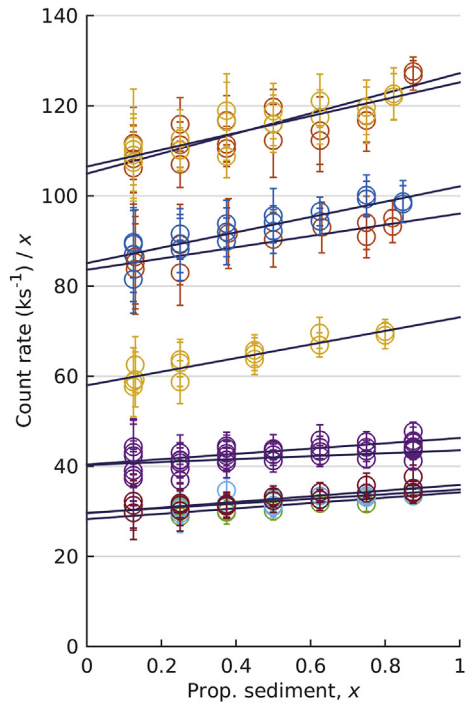

Fig. 4. Count rates for 10 samples measured by varying the sediment-wax proportions between sub-samples, with count rates corrected for the proportion of sediment in each. The extrapolated y-intercept is independent of stopping power, and used to define the count rate b (eq. (2)).

4. Results

Eleven samples were selected from the NLL archives, covering a range of dose rate, location and depositional setting. For one sample, the measurement procedure was repeated six times (Fig. 5). The mean stopping-power corrected count rate for this samples is 107 ks^{-1}, with a relative standard deviation of 1.9%. The overdispersion in the detrended sub-sample count rates is 2.4%. For the 6 replicates, the estimated uncertainty on b ranges from 1.6% to 2.2%. We can be confident, therefore, that the extrapolated count rate estimate is very precise (1.9% measured RSD), and that the estimate of the uncertainty is also accurate.

For each of the 11 samples, the beta-plus-gamma dry dose rates have been derived from the beta count rates using three choices of prior information, then compared to HRGS-derived dose rates (Fig. 6).

- Method 1: Prior information from the relative proportions of the K, U, Th sources, as determined by HRGS (Fig. 6a and b). This represents the maximum amount of prior information. The uncertainties plotted in Fig. 6 are derived purely from the beta count rate. The ratios have an overdispersion of 2.8%, part of which must be accounted for by random uncertainty in the HRGS measurements.
- Method 2: Prior information uses only the ^{40}K activity concentrations, taken from the HRGS data, from which the relative proportion of K, U, Th are estimated (Fig. 6c and d). We assume equal contributions of U and Th to the beta-plus-gamma dose rate, which is reasonable in most cases. The ratio of method 2 to method 1 has

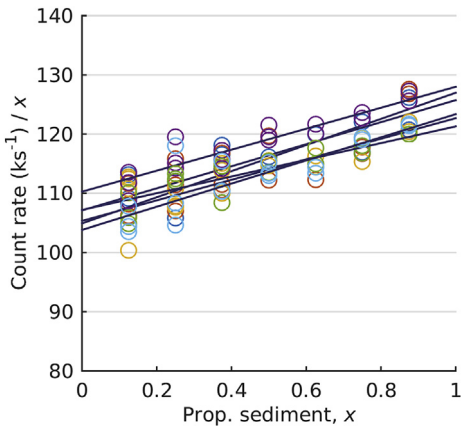

Fig. 5. Reproducibility estimate of the stopping-power correction procedure. Six batches of varying wax/sample mixture were prepared for one sample, each plotted here in different colours. The relative standard deviation in the corrected count rate (i.e. the y-intercept) is 1.9%. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

relative standard deviation (RSD) of 2.1%; i.e. the cost of using method 2 is a 2.1% increase in uncertainty, compared to the ideal method 1.

- Method 3: Prior information uses approximate ^{238}U and ^{232}Th activities measured using an alpha counter, from which the relative proportions of the three sources are estimated (Fig. 6e and f). The ratio of method 3 to method 1 has RSD = 2.3%; i.e. it results in a 2.3% increase in uncertainty above the ideal method 1.

Note that for all methods, the prior activities are expressed as the proportion of the dose rate that can be expected from each source, and then plugged into equation (2).

5. Discussion

Random errors in the estimated dose rate represent one of the major sources of uncertainty in luminescence dating. The uncertainty estimates of mass concentrations are rarely published, but we can guess that the random uncertainty on the dose rate exceeds 5%. High-resolution gamma spectrometry is probably the most precise method of analysis. Murray et al. (2015) obtained a dose rate RSD of 6.2% for 20 separate subsamples of the inter-comparison sample, measured over three detectors. However this was a relatively low radioactivity quartz rich-sand; using a single detector on a higher activity loessic material, the variance can be lower, with RSD around 3% (e.g. Hossain et al., 2002). However, HRGS instruments are not typically available in OSL laboratories, and if activity concentration estimates are obtained externally (from ICP-MS, NAA etc.) then the opportunities to validate the results, or quantify the uncertainties, may be limited.

Using the procedure described here, beta counting instruments provide a means of obtaining high-precision dose rates at relatively low cost. The maximum obtainable random precision on the dry dose rate is limited by the counting uncertainty. Given a practical measurement time of 24–48 h, this will typically be ~2%. The conversion of count rate into dose rate involves some additional uncertainty, but using prior information this can be kept very low. The simulations suggest it can be reduced to ~1.5% (Fig. 1i); measurements here on 11 samples show the additional uncertainty is ~2%. Added in quadrature, the total random uncertainty should then be less than 3% – comparable to, or better than, the best alternative (HRGS). Imprecise prior knowledge of either the ^{40}K (method 2) or ^{238}U and ^{232}Th (method 3) activities is sufficient, and can be obtained cheaply—e.g. flame photometry for K, or alpha counting for U and Th.

Without prior information on the relative strength of the sources (proportions of K, U, Th), a common calibration factor must be applied to all samples. The common calibration is reasonably accurate when predicting the beta dose rate (Fig. 1d), because the source calibration

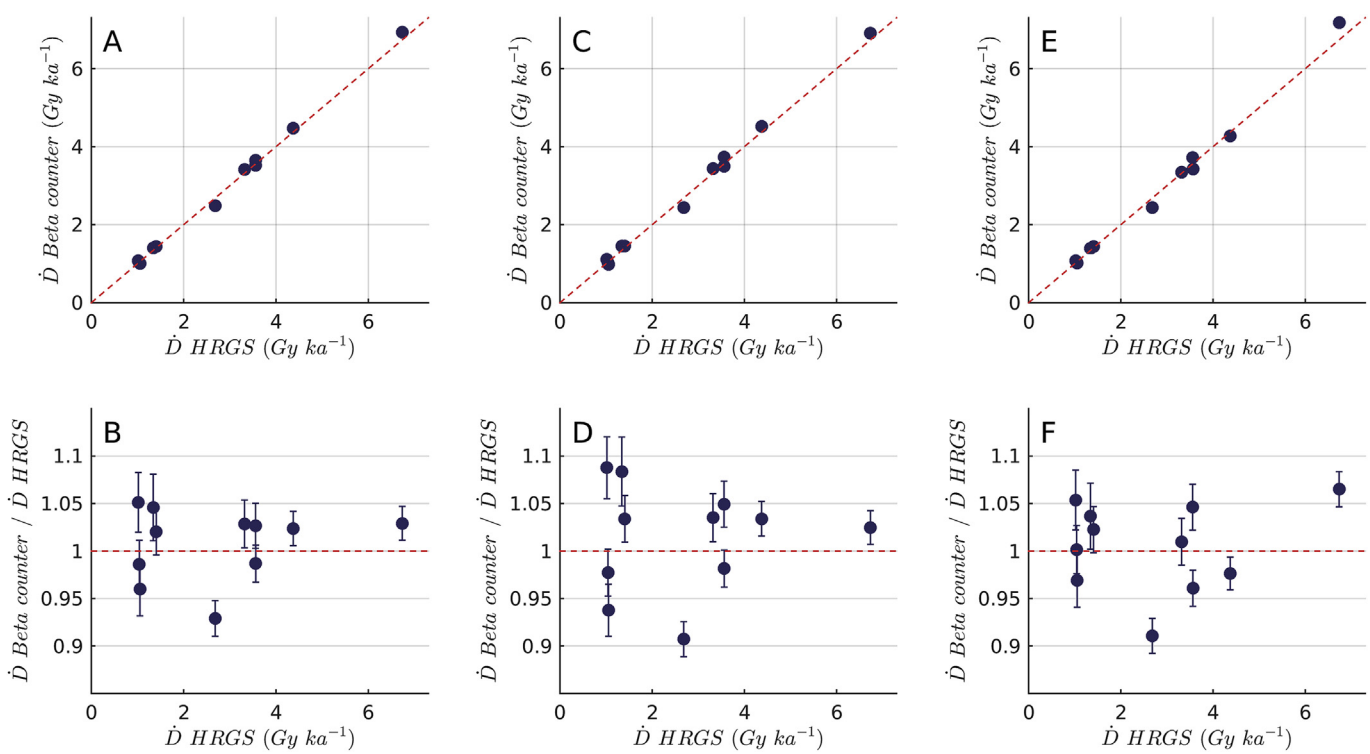

Fig. 6. Beta-plus-gamma dose rates estimated for 11 natural sediment samples, compared with HRGS measurements (top row), and plotted as a ratio (bottom row). (a–b) Prior proportions of K, U, Th sources determined by the HRGS measurements. (c–d) Prior proportions estimated using only the HRGS ^{40}K activity. (e–f) Prior proportions estimated from approximate ^{238}U and ^{232}Th activities from alpha counting. Error bars display only the random uncertainty in the beta count rate.

factors are fairly similar (Table 1; and Bøtter-Jensen and Mejdahl, 1988; Ankjærgaard and Murray, 2007). For this reason, current use of the beta counter in OSL dating is restricted to beta dose rate estimation only. While measurement details are rarely provided, we presume that most laboratories evaluate the beta dose rate of a sample by comparing its count rate with a reference material (Mauz et al., 2002; Jacobs and Roberts, 2015). This method assumes, in addition to a common calibration factor, that the stopping power of the sample is the same as that of the reference material. Of the 11 samples measured here, the range of calibration count rates is 38.3–40.6 $ks^{-1}/(Gy\ ka^{-1})$ of beta dose, and the range in the stopping-power statistic is 1.09–1.24. Use of a reference material could then lead to an error of up to 6% due to relative source-strength differences, and up to 15% due to stopping power differences; an error in the beta dose rate of over 20% is then possible. This error might be largely systematic within any one site—because elemental composition may be similar— leading to a precise but inaccurate chronology.

The use of a combination of alpha counting and beta counting is now very compelling, provided that the alpha count rates are used as prior information to calibrate the beta count rate. In principle, alpha counting alone is a very precise means of estimating the combined beta and gamma dose rate from U and Th sources, thanks to some fortuitous anti-correlations in the uncertainties. Using the pairs method (see Aitken, 1985), the estimated ^{232}Th activity is rather imprecise, because the slow-pairs count rate is very low and random pairs must be considered. However, the ^{238}U activity is estimated by subtracting the inferred ^{232}Th alpha count rate from the total alpha count rate, so any error in the ^{232}Th activity is largely cancelled out by an opposite error in the estimated ^{238}U activity. A second anti-correlation occurs through the conversion of ^{238}U and ^{232}Th activities into dose rate. The dose rate from ^{238}U is dominated by the beta component, while that from ^{232}Th is dominated by the gamma component; the beta and gamma dose-rate uncertainties are therefore anti-correlated, and the combined beta-plus-gamma dose rate is more accurate than might otherwise be expected. Finally, the use of ^{238}U and ^{232}Th activities as prior information exploits a further anti-correlation in the beta-counter calibration: by expressing the priors as a proportion, an error in the U plus Th dose rate estimate is partly cancelled by an opposite error in the inferred ^{40}K activity.

The anti-correlations in alpha and beta counting contrast with the correlated uncertainties when deriving the dose rate from activity estimates (via HRGS, ICP-MS. etc). In that case, an error in an activity estimate causes an error in both the beta and gamma dose rates, in the same direction. For simplicity, standard procedures for estimating the dose rate uncertainty from such concentration measurements usually presume that these two components (beta and gamma dose rates) are independent (i.e. uncorrelated), and so can be added in quadrature. The random dose rate uncertainty is then slightly underestimated (typically, by about 20%). Unlike spectroscopic methods, however, beta counting cannot provide information on the present-day state of secular equilibrium. When using beta counting methods for dose-rate estimation, it is implicitly assumed that the state of equilibrium in the sample is the same as the radionuclide standards, and that this condition has prevailed throughout the burial period. For sediments, this is most relevant for the U-series, in which secular disequilibria may sometimes occur. The radionuclide standard for the U-series employed here is known to be in secular equilibrium, so the dose-rate estimates also presume secular equilibrium in the samples. It should also be noted that beta-counting methods offer no direct information on the alpha dose rate, which may contribute a significant component of the total dose rate to fine or non-etched grains.

Random errors also occur in the measurement of water content, and in the measurement of equivalent dose. Under favourable conditions, both sources should be expected to add 2–3% to the age uncertainty. There are also a number of systematic errors in luminescence dating, mostly related to dose rate estimation. These are more difficult to quantify than random uncertainties, but a summary of their probable magnitude was made by Murray and Funder (2003). They are chiefly related to the measurement of saturation water content, the calibration of instruments, and conversion of mass-concentrations to dose rate. Added in quadrature, the systematic uncertainties sum to ~5.5%. It is clear that using the beta counter to reduce the random uncertainties to < 3% is highly beneficial, and would leave the systematic errors and the water content as the major sources of uncertainty in luminescence ages.

6. Conclusion

Beta counting instruments are simple to use and relatively cheap, and provide a very high precision beta count rate for a sediment sample. Using the procedure described here, the count rate can be used to estimate the dry beta and gamma dose rate in natural sediment, with random uncertainty < 3%. This procedure includes a means of correcting for self-attenuation via dilution analysis, and requires prior information on the relative strength of the radionuclide sources (K, U, Th). The prior knowledge need not be well known, and so can be obtained very easily by a number of methods.

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