Field gamma dose-rate measurement with a NaI(Tl) detector: re-evaluation of the "threshold" technique

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Abstract
Radiation detectors, like doped NaI, are commonly used in the field for the determination of gamma dose-rates. In most cases, and even though these systems generally allow one to record the full gamma spectrum between 0 to around 3 MeV, this dose-rate is computed from the count rates recorded in a limited number of "windows" (Aitken, 1985). With this technique, only a small part of the spectrum is therefore exploited. Nevertheless, an alternative approach - the "threshold" technique – known for more than 30 years, can easily be used with the same detection system. In this paper, we make a re-evaluation of this technique and discuss its limits and advantages.

Introduction
In a paper published in 1974, Løvborg and Kirkegaard investigated the response of a 3 inch by 3 inch NaI(Tl) detector placed above environments of known radioactivity. According to their experimental and theoretical results for this 2π geometry, the count-rate of their equipment above a chosen threshold (370 keV) was found to be directly proportional to the gamma dose-rate. As the gamma rays come from the radioactive elements of the U- and Th-series and from the ^{40}K, this means that the count-rate was independent of these three sources.

In the next years, Murray, Bowman and Aitken (1978) developed a portable system equipped with a NaI(Tl) detector for gamma dose-rate measurements; it is discussed in Aitken (1985, p. 107-108) as the "gamma scintillometer". In their experiments, they inserted their detector in radioactive doped blocks set up at the Research Laboratory for Archaeology and the History of Art (see Rhodes and Schwenninger, 2007). With these experimental conditions, which are close to field conditions, they concluded that a threshold value could also be defined for their equipment (at 450 keV). However, these authors showed that this threshold occurred in a region of the spectrum where the count-rate changed rapidly with energy, and they then emphasized the necessity to stabilize the system to account for temperature variations.

We recently tested this approach with a 1.5 inch by 1.5 inch NaI(Tl) detector (IPRON-1 connected to a multichannel analyser: Inspector1000 – Canberra) and report here our results and conclusions.

General
In luminescence and ESR dating, the gamma dose-rate generally constitutes a significant component of the total dose-rate and has to be determined precisely. This dose-rate can be measured directly with synthetic dosimeters (e.g. Al_2O_3:C or $CaSO_4$:Dy) buried for relatively long periods of time (weeks or even months), or by inserting in the sediment a portable detector connected to a spectrometer and recording the gamma spectrum. In this case, the gamma dose-rate is calculated from the radioisotopic contents of the sediment (U, Th and K) determined by the "classical" technique, which is based on the definition of three regions of interest ("windows"), each window being centred on a gamma ray energy specific to an isotope (1460 keV for ^{40}K; 1780 keV for ^{214}Bi (U-series) and 2620 keV for ^{208}Tl (Th-series), see for instance Aitken (1985, p.102-105). In each of these two last windows, the counting is due to the detection of gamma rays coming from both the U- and Th-series, whereas in the "K window" the three sources contribute. Mathematically, one can then simply write for each window the following equation:

$$N = n_K \cdot [K] + n_U \cdot [U] + n_{Th} \cdot [Th]$$

where the total count N (per unit of time) is the sum of three factors in which [K], [U] and [Th] are the radioisotopic contents of the sediment and n_K, n_U and n_{Th} represent the detection efficiencies of the detector in the considered window for the three sources. These efficiency factors are then expressed as the number of counts per unit of time for 1 ppm (for the U- and Th-series) and 1% for K.

In considering the threshold approach, these factors have to be interpreted as the number of counts per unit time for 1 µGy/a and [K], [U] and [Th] as the gamma dose-rates in the sediment. Consequently, for N to be proportional to the total dose-rate, n_K, n_U and n_{Th} must have the same value n. In this case, the above equation can be written as :

$$N = n \cdot ([K] + [U] + [Th])$$

where ([K] + [U] + [Th]) is the total gamma dose-rate.

In practice, for using this approach with a given detector, one has simply to define the energy for which the values of n_K, n_U and n_{Th} are identical.

Experiments
One way to answer this question is to get "pure" spectra taken in environments containing only one source of radioelements (either K, U- or Th-series). Such environments do not naturally exist, so we used the doped blocks set up at the Research Laboratory for Archaeology and Art, in Oxford, as reported by Murray et al. (1978). These blocks made of concrete were doped with uranium, thorium and potassium and provide the following gamma dose-rates: U-block: 13.27 Gy/ka; Th-block: 7.10 Gy/ka; K-block: 1.38 Gy/ka. A non-doped block made of the same concrete is also available and is used as a background standard with a gamma dose-rate of 0.53 Gy/ka (Rhodes and Schwenninger, 2007). According to these authors, the Th-block contains a small portion of U with a U/Th ratio of 0.043. Considering this ratio and the dose-rate conversion factors given by Adamiec and Aitken (1998), one can calculate the effective radioisotope concentrations : U-block (117.4 ppm), Th-block (135.2 ppm of Th and 5.8 ppm of U), K-block (5.7 %).

Note that these values are slightly different from those given in Rhodes and Schwenninger (2007) since they used the conversion factors of Nambi and Aitken (1986).

Fig. 1 shows the spectra recorded with our gamma probe. After time normalisation (1 ksec), "pure" spectra were calculated by subtracting the signal

measured in the non-doped block (thus including the background of the detector and the cosmic contribution) and were normalised to 1 ppm of U or Th, and 1 % of K (Fig. 2), by using the effective radioisotope concentrations cited above. This step was necessary in order to subtract from the Th spectrum the small contribution from U present in the Th-block.
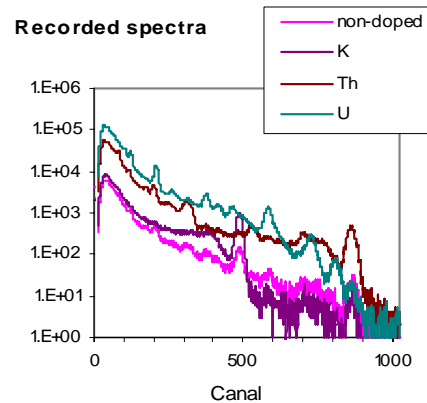

Figure 1: *Spectra recorded with the NaI(Tl) probe in the four Oxford blocks. The multichannel analyser was configured for recording 1024 channels. Measurement times were about 35 minutes for the U, Th and K-doped blocks, and 1.1 hour for the non-doped block. Energy calibration of each spectrum was carried out by identifying specific gamma rays.*
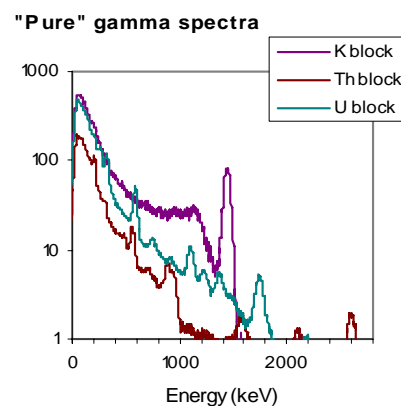

Figure 2: *Spectra given as the number of counts per 1000 seconds and 1 ppm (for U and Th) or 1% (for K) after subtraction of the spectrum recorded in the non-doped block. Notice that for the K spectrum, the contribution of the concrete was limited to 72% due to its dilution by the added potassium salt. A correction was also applied to the Th spectrum for taking into account the uranium contamination (5.8 ppm) present in the Th block. Note that the U, Th and K contents of blocks were deduced from dose measurements performed with dosimeters (Rhodes and Schwenninger, 2007) and differ slightly from the values used by Murray et al. (1978).*
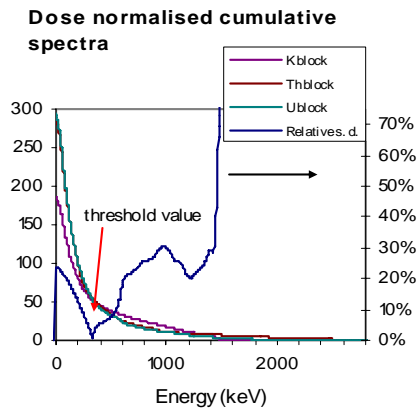
Figure 3: *Cumulative spectra as a function of energy deduced from Fig. 2. For any x-value, the y-axis indicates the number of counts above this energy. Each spectrum was dose normalised according to the gamma conversion factors given by Adamiec and Aitken (1998). The right axis gives the relative standard deviation of the three cumulative spectra; the minimum value is around 320 keV.*

To define the threshold for our detector, these pure spectra were normalised with the dose-rate conversion factors given by Adamiec and Aitken (1998) and thereafter cumulative spectra were computed (Fig. 3). For any given energy, the left y-axis value is the total count recorded by the detector for the corresponding source (either U-series, Th-series or K). The three spectra have a similar shape and are close to each other in the 200-600 keV energy range. By computing the standard deviation of these curves as a function of energy (right y-axis on Fig. 3), the minimum value appears to be located around 320 keV. Hence, for this particular energy value, the n_U, n_{Th} and n_K parameters are almost identical : the n factor of our detector is 53.7 counts/ 1 ksec/ 1 µGy/a.

Testing the "threshold" approach
We tested this approach by recording a gamma spectrum in a block made of building bricks available at the Gif laboratory (Mercier et al., 1994). The radioisotopic contents of these bricks produced in the Auvergne region (France) were determined by NAA and high resolution gamma spectrometry and are, on average : U 6.40±0.68 ppm; Th 22.89±2.43 ppm; K 3.61±0.23 %. These analyses indicated no disequilibrium in the U- or Th-series. A CaSO$_4$:Dy dosimeter placed at the centre of this block recorded a dose-rate to quartz of 2578±80 µGy/a – corrected for a small cosmic contribution estimated at 160 µGy/a after Prescott and Hutton (1994). The gamma dose-rate deduced from the "threshold" approach

using the NaI detector (2475 µGy/a) was found to be in good agreement with this value.

Here, an important point is to estimate the error associated with the calculated value. At least three sources have to be studied: counting statistics, disequilibrium in the radioelement series, and energy calibration.

1) The gamma spectrum recorded in the Gif block was taken using a counting time of 1342 seconds and the number of counts above the threshold was 186,980; the counting statistics were then sufficient to ensure that they do not constitute the main source of error. Notice that the threshold factor (53.7 counts/ 1 ksec/ 1 µGy/a) allows one to estimate easily the contribution of the statistics to the total error or, for an accepted error, to evaluate the minimum time of counting (if one has a rough estimation of the dose-rate in the sediment). For instance, for a sediment with a gamma dose-rate of about 500 µGy/a, a counting time of 5 minutes is sufficient to get a counting precision around 1%.

2) As noted above, the U- and Th-series are in secular equilibrium in the Gif block. It was assumed that this situation prevails also in the Oxford blocks, and this is indirectly confirmed by the test done in the Gif block. However, if one of the doped blocks, for instance the U block, exhibited strong radioactive disequilibrium because of radon loss, one would expect that the U cumulative spectrum of Fig. 3 would have a slightly different shape since post-radon emitters give high energy gamma rays: such disequilibrium would therefore have a small impact on the calibration as the value of the threshold would stay virtually unchanged. Moreover, it is important to recall here that the threshold approach takes account of most of the emitted gamma rays and should be a little bit less sensitive to radioactive disequilibrium than the classical windows technique, from which the U content is derived from the [214]Bi gamma ray (1780 keV), detected in the U window, i.e. a post [222]Rn emitter.

3) The main source of error is probably related to the energy calibration of a recorded spectrum, and consequently, to the setting of the threshold for this spectrum. For this purpose, one can generally use the [40]K peak at 1460 keV but it can also be useful to have at hand, especially if the counting time is reduced to a few minutes, a short-lived radioactive element (such as [133]Ba which has multiple gamma ray lines under 500 keV, or [241]Am as discussed in Aitken (1985, p. 323-324)). Such a source can be used just before and after recording the spectrum in the sediment. It should be noted that it is illegal to carry such a source

on an aeroplane. In some gamma spectrometers (e.g. Harwell Nutmaq) such a source was built in to the detection head in order to overcome energy drift with changing temperature.

To estimate the influence of the calibration on the deduced dose-rate, we computed this dose-rate in the interval 300-360 keV (as we estimate that the energy calibration can be performed with a maximum error of ±30 keV). The deduced values would vary from 2422 to 2481 μGy/a with a minimum of 2414 μGy/a at 310 keV. According to this example, one can therefore estimate that the error introduced by the energy calibration of the spectrum is less than 3% for our detector.

Conclusions

By using a large part of the spectrum (all the counts above a fixed energy), the "threshold" approach allows one to reduce considerably the recording time and this limits the undesirable effects of temperature changes during measurement. In considering the different sources of error, including the accuracy of the gamma dose-rates measured in the Oxford blocks, one can estimate the overall error associated with a dose determination to be around 5 %. In spite of these encouraging results, it seems important to make systematic comparisons with doses recorded with dosimeters, especially by choosing different environments (dominated by silicates, carbonates, clays, or others) in order to study their influence on the threshold value (see Liritzis and Galloway, 1980).

Furthermore, as this approach makes use of a large part of the spectrum (in contrast with the "windows" technique), it is tempting to reduce the size of the detector for use in sediments rich in stones or gravels and also in archaeological layers. Also, the resolution of the detector is not a critical parameter with the threshold approach, neither is the light output efficiency relative to NaI, and other crystals could then be tested.

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