Dose rates and radioisotope concentrations in the concrete calibration blocks at Oxford

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(Received 21 Nov 2006; in final form 22 May 2007)

Introduction

The "Oxford Blocks" were constructed for the calibration of portable gamma spectrometers, and consist of 4 concrete cubes measuring 50 cm along each edge, each with a central horizontal cylindrical hole of 11.0 cm diameter which goes from one side to the other. Three blocks have material added to increase their radioactivity; the U block includes ground pitchblende, the Th block contains monazite sand and the K block has a potassium-bearing salt (sulphate of potash) mixed with the concrete, to provide calibration standards for U, Th and K respectively. The fourth block is plain concrete, and acts as a background standard. They are suitable for the calibration of portable gamma spectrometers with probes up to 3 inches in diameter. They are separated from each other by additional plain concrete blocks without holes, to reduce crosstalk between blocks during measurements. Each calibration block has additional concrete cylinders which reside within the central holes; two 25 cm lengths each comprise three cylinders which fit inside each other, made of the same concrete mix as their respective block. One set of three cylinders is removed during spectrometer calibration, and the remaining set adjusted to allow the detection crystal to be central; that is the centre of the crystal is positioned 25 cm from both front and back faces. The K block contains only 72% concrete, the rest being sulphate of potash, and the monazite sand in the Th block contains a small amount of U; these must be taken into account during the calibration process.

The blocks were constructed in 1973 (Bowman, 1976) to the rear of the Research Laboratory for Archaeology and the History of Art (RLAHA), 6 Keble Road, Oxford, UK. After construction, neutron activation analysis (NAA) and thick-source alpha counting of small sub-samples were used to derive preliminary radioisotope concentrations. Partly to overcome concerns that the radioisotopes might not be distributed evenly, the gamma dose rates in each block were subsequently measured directly using TL dosimeters; it is these measurements from which the quoted dose rate values, and estimated radioisotope concentrations, are derived (Table 1).

Block dose rate determinations

In 1973, dose rate determinations were made by placing small copper capsules containing heated natural CaF_2 (calcium fluoride) grains in each block for a period of several months, and measuring the acquired dose using TL measurements. This procedure was regularly repeated by RLAHA students (Gaffney, Clark and Rhodes) until around 1989, using a calibrated $^{90}Sr/^{90}Y$ source and a regenerative-dose procedure on equipment in the main glow room in Oxford. Martin Aitken treated the CaF_2 by light exposure prior to its placement in the copper capsules in order to reduce sensitivity change during TL measurement.

The dose rate determination procedure was as follows. TL estimates of dose were made using capsules containing CaF_2 placed in all four blocks, K, U, Th and background (BG) blocks for a known time, typically around 3 months. After heating the capsules in order to zero the TL signal, they were placed at the geometric centre of each block, and the moveable concrete cylinders arranged to minimize voids around each capsule. At the end of the exposure period, the capsules were opened in turn, and the dose determined for each using a simple single aliquot regenerative-dose TL procedure; two or three aliquots were measured for each block and the mean values used in calculations. The total measured dose rate values from the U, Th and K blocks had the measured concrete, and estimated cosmic and self dose rates subtracted. Note that for the K block, only 72% of the BG block concrete component is subtracted since the sulphate of potash used to dope the concrete constitutes 28% of the block. These

	K block	Th block	U block	BG block
Concrete	72%	~100%	~100%	~100%
Gross dose rates (mGy/a)	1.75	7.05	12.66	0.60
Concrete, self dose, cosmic (mGy/a)*	0.50	0.60	0.60	0.60
Net dose rate (mGy/a)	1.25	6.45	12.06	0.00
Wall corrected (x1.1) (mGy/a)	**1.38**	7.10	**13.27**	-
Th dose rate, Th block (92.3%) (mGy/a)	-	**6.55**	-	-
U dose rate, Th block (7.7%) (mGy/a)	-	**0.55**	-	-
Effective concentrations, using Nambi and Aitken (1986)	5.71% K	125.7 ppm Th 4.8 ppm U	116.8 ppm U	

Note :
* The K block is only 72% concrete, and this must be taken into consideration when subtracting the background block dose rate, which comprises contributions from concrete, and estimated self dose (0.1 mGy/a) and cosmic dose rate (0.16 mGy/a). The Th block contains a small amount of U; Th and U dose rates are estimated based on the certified Th:U ratio. Note that these values only represent "excess" dose rates; values in bold represent the current best estimates of dose rates for the Oxford Blocks.

Table 1: *Effective dose rates and concentrations of K, U and Th in the Oxford concrete calibration blocks. These were measured directly using TL of calcium fluoride in copper capsules. The values in this table represent a combination of measurements made by Gaffney (1983), Clark (1984) and Rhodes (1985-89).*

"excess" values (that is, the dose rate contributions only from the added materials in each block) were then corrected for capsule wall attenuation, using a correction factor of 1.1 for the 1 mm copper wall.

As mentioned above, a small contribution to the gamma dose rate in the Th block is from U, which represents an integral part of the natural monazite sand used. The U:Th concentration ratio for this sand was quoted as 0.035 by NBL on the original certification (Martin Aitken, unpublished notes), and this value was used to apportion the measured dose rate to U and Th using dose rate conversion factors. We note that this concentration ratio has more recently been determined using NAA by EJR as 0.043. In principle, this ratio could also be determined using a calibrated NaI spectrometer, or by high resolution Ge spectra of the same monazite sand.

This procedure yields net dose rate values for each block which are used in the calibration of gamma spectrometers to derive individual U, Th and K dose rates from subsequent field measurements, and also for the calculation of total dose rate using the threshold method. For this reason, these effective "excess" dose rates are of primary importance, rather than the U, Th and K concentrations calculated using conversion factors. However, the values that have previously been widely quoted for the blocks are expressed as "effective concentrations" of K, U and Th, estimated using either Nambi and Aitken (1986) or with earlier conversion factors.

The dose rate values presented in Table 1 represent estimates based on combined measurements by Chris Gaffney (in 1983), Peter Clark (in 1984) and Ed Rhodes (1985 to 1989), and represent an update on the values given by Aitken (1985; appendix L). Also shown are the widely circulated radioisotope concentration values (note that the potassium concentration quoted here as 5.71% K is equivalent to 6.88% K_2O).

Discussion
The process described above for characterising the blocks for use as calibration standards relies on several assumptions, namely:

1. CaF_2 provides an accurate estimate of dose-rate on timescales of months,
2. the background dose-rate subtraction does not introduce error,
3. the correction for copper capsule wall attenuation is accurate,
4. the laboratory beta source calibration is accurate,
5. the size of blocks is sufficient for good replication of an infinite matrix,

and for accurate radioisotope concentrations,

6. the dose-concentration conversion factors used are accurate.

The use of CaF_2 in copper capsules to determine small environmental doses has been standard practice for many years (e.g. Aitken 1985), and is not discussed further here. As the dose rate measured in the concrete background block is subtracted from the gross dose rates for the U and Th blocks, it is not important what the relative magnitude of the concrete, self dose and cosmic dose rates are. However, this does matter a little for the K block which is only 72% concrete; we consider that the cosmic dose rate is probably underestimated, and we have no information regarding the CaF_2 self dose rate. For the derivation of the dose rates presented in Table 1, a single wall correction factor of 1.1 was used irrespective of the gamma energies present, for the 1mm thick copper capsule. We consider that there may be room to improve this value in the future using modern Monte-Carlo modelling (e.g. Nathan et al., 2003), and Al_2O_3:C may represent an improved material for future dose rate assessments (e.g. Burbidge and Duller 2003).

The RLAHA beta source calibration was undertaken on several occasions, notably by Bert Roberts and Nigel Spooner in 1993. It was re-assessed by Phil Toms using a SAR OSL protocol in 1998, who demonstrated agreement within 1% for calibrations based on irradiations at GSF (Munich, Germany), NIST (Gaithersburg, USA) and the NPL (UK). Note that the K, U and Th concentration values which are widely circulated and presented below in Table 1 rely on the energy conversion values of Nambi and Aitken (1986). The updated values of Adamiec and Aitken (1998) provide improved estimates and these are given in Mercier and Falguères (2007).

Whether the blocks are of sufficient size to replicate the geometry used for in-situ gamma measurements has been a matter of debate. Jean Fain expressed concern, in particular for the Th block, for which the ^{208}Tl emission at 2614.6 keV represents the line used in NaI spectrometry for Th concentration estimation. If this block is too small to reproduce the full spectrum of infinite matrix Th emissions accurately, the estimated effective concentration will differ slightly from the true concentration. The effect will be greater for higher energy gamma emissions, which travel further through the concrete matrix. However, the estimation of total dose rate will not be severely affected, as this relies on the direct measurement of a luminescence dose using CaF_2, which includes contributions across the full energy spectrum. In summary, the calibration of the gamma spectrometry estimation of Th content using the 2614.6 keV ^{208}Tl emission will be the parameter most influenced by non-infinite matrix effects caused by the limited block size. The estimated Th content using spectrometers calibrated in these blocks, should consequently be treated with some caution, while the total Th series gamma dose rate contribution remains relatively well determined. The same effect, though to a reduced magnitude, is expected for the K and U blocks.

Gamma dose rate data collected with an EG&G MicroNomad NaI portable spectrometer, which was calibrated in these blocks using the values shown in Table 1, were used in the relatively high precision OSL age estimates of archaeological sediments presented by Rhodes et al. (2003). Very good agreement with radiocarbon dating was observed, with no indication of any systematic error; beta dose rates were calculated using NAA determination of K, U and Th content. The ratio of the mean sediment K and U contents determined by NaI and NAA was within 5% of unity, while for Th the mean of the NaI to NAA ratio was 0.90 ± 0.06. These observations are consistent with the expectations discussed above, namely that the gamma dose rate contributions should be well estimated, but that individual concentrations of K, U and Th are probably less reliable, in particular the Th value.

The blocks were recently relocated from the Keble Road site to the rear of the Dyson Perrins Laboratory in specially constructed steel cradles, where their original relative geometry is retained. It now seems sensible to update the dose rate estimates with new measurements based on Al_2O_3:C.

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Reviewer
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Editor's Comments
I am very grateful to the authors for putting this paper together, and think that it is very important that the basis for these calibration values have been described. Rhodes and Schwenninger refer in this paper to the DPhil thesis of Sheridan Bowman that was completed in 1976. With the permission of the Sheridan Bowman, a PDF of Appendix A5 of Bowman (1976) is available via the Ancient TL website (http://www.aber.ac.uk/ancient-tl/issue25_1/bowman.pdf). This appendix describes the original construction of the doped concrete blocks in Oxford, and a suite of CaF_2 dosimetry measurements that were made to assess the gamma dose rates in each of them. The gamma dose rates presented in this appendix are different to those given above by Rhodes and Schwenninger, and this perhaps serves to reinforce the final comment in the paper that the gamma doses should be reassessed. Additional information about the original characterisation of these blocks and about construction of a larger U doped block can be found in Murray (1981).

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