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Published in:
Radiation Measurements

Link to article, DOI:
10.1016/j.radmeas.2018.04.006

Publication date:
2018

Document Version
Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):

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Measurement of natural radioactivity: Calibration and performance of a high-resolution gamma spectrometry facility

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ABSTRACT

Murray et al. (2015) described an international inter-comparison of dose rate measurements undertaken using a homogenised beach ridge sand from Jutland, Denmark. The measured concentrations for $^{226}$Ra, $^{232}$Th and $^{40}$K from different laboratories varied considerably, with relative standard deviations of 26% (n = 8), 59% (n = 23) and 15% (n = 23), respectively. In contrast, the relative standard deviations observed internally within our laboratory were 9%, 11% and 7%, respectively (n = 20), and in addition our mean values were consistent with the global $^{40}$K mean, but significantly different from the $^{232}$Th mean. These problems in both accuracy and precision have led us to examine both the long term performance of our analytical facility, and its calibration. Our approach to the preparation of new absolute $^{238}$U, $^{232}$Th and $^{40}$K standards is outlined and tested against international standards. We also report analyses of the Volkegem (De Corte et al., 2007) and Nussi (Preusser and Kaiser, 2001) loess samples, for further comparison with other laboratories.

1. Introduction

Dose rate determination in luminescence dating is as important as dose determination, in terms of its contribution to the age. Uncertainties in dose rate are thus of considerable importance, and yet the quality of the input data used to derive these dose rates (such as radionuclide concentrations, or integral count rates) receives little or no systematic attention in the luminescence dating literature. Indeed many laboratories do not determine their own radionuclide concentrations, but rather buy analyses from other analytical facilities; this makes quality control more difficult. In 2006, a laboratory intercomparison study was initiated to investigate the coherence in measurement and analysis of the various parameters that are used to produce a luminescence age. The study was based around a homogenised beach ridge sand from Jutland, Denmark; 23 laboratories returned radionuclide analyses. The results of this intercomparison are now available (Murray et al., 2015) and our present study arises from the dose rate results in particular. As the host laboratory for this study, we had undertaken extensive measurements to test, \textit{inter alia}, homogeneity of radionuclide concentrations in the intercomparison sample; 20 different subsamples were analysed using 4 different gamma spectrometers. These analyses documented the reproducibility of our facility (RSD of 9%, 11%, 7% for $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively) but of course could not give information about the accuracy. The radionuclide concentrations in the intercomparison study were considerably over-dispersed (up to 60%), especially those for $^{232}$Th (Fig. 1). Although our analyses lay within one standard deviation of the respective mean, it was clear that independent testing of our calibration was desirable. This led to the present study.

The calibration standards chosen by high resolution gamma spectrometry facilities for the analysis of natural radionuclides have varied considerably across different laboratories, and unfortunately they are not always described at an appropriate level of detail. Murray et al. (1987) produced multiple independent U and Th standards by mixing, with a low activity sand, known quantities of uraninite ore BL-5 (certified reference material, CRM, produced by the now Natural Resources Canada, NRCAN), or thorium nitrate refined in 1906 (Amersham International). These pulverised mixtures were then in turn mixed with polyester resin to retain Rn and provide a constant counting geometry. For $^{40}$K they mixed either reagent-grade K$_2$SO$_4$ or KCl directly with polyester resin and assumed stoichiometry. Guibert and Schrover (1991) also made their own standards by first diluting and fusing pitchblende (UO$_2$) and ThO$_2$ in di-lithium tetraborate glass; they did not discuss the possible effects of high temperature on volatile daughter radionuclides, or the age of the ThO$_2$ (important for confidence in the degree of secular equilibrium, since all gamma rays are emitted by $^{228}$Ra, $t_{1/2}$, 6.7 years, and its daughters). These glasses were then crushed and mixed with a silica powder to provide the final U and Th standards.
The $^{40}\text{K}$ standard was made from $\text{K}_2\text{SO}_4$. Preusser and Kasper (2001) produced their own standards based on material provided by the International Atomic Energy Authority (IAEA) but no further details were given. Mauz et al. (2002) used a soil material (“BFS ST2.3/Boden/V-98”) for which the concentrations were given by Schkade et al. (1998). De Corte et al. (2005) made use of certified IAEA gamma-spectrometry reference materials RGK-1, RGU-1 and RGTh-1 (IAEA, 1987). The first is based on reagent-grade $\text{K}_2\text{SO}_4$ and the latter two are derived by the IAEA from NRCAN’s CRM BL-5 (uraninite) and CRM OKA-2 (britholite).

All the calibrations above used known activity standards presented in the same geometry as the unknown samples to be analysed. Others, such as Oczkowski (2001), Al-Sulaiti et al. (2012) and Rihs et al. (2016, 2015) calibrated their detectors by determining the absolute efficiency as a function of energy and then used emission probabilities from the literature to calculate concentrations in the unknowns.

Two groups have used their calibrations to compare the analyses of loess samples with the results of other analytical techniques (Volkegem et al., 2003). Since 2003 we have repeatedly measured the same sample on each of our detectors (note that this sample, lab. code 950509A, was prepared in 1997 using polyester resin rather than wax). Typical re-producibility for $^{226}\text{Ra}$ only) and one PGT closed-end coaxial ‘p’ type detector (code D, sensitive diameter 49 mm, depth 30 mm, resolution 1.9 keV at 1332 keV, relative efficiency ~10%). Spectra were collected in $4k$ channels usually for $\sim20$ h, and analysed using propriety software based on the procedures described by Murray et al. (1987) to give net peak areas from various daughter nuclides and, after calibration, activity concentrations in Bq$\text{kg}^{-1}$ of $^{238}\text{U}$, $^{226}\text{Ra}$, $^{210}\text{Pb}$ (detectors J and K only), $^{232}\text{Th}$ and $^{40}\text{K}$.

### 3. Analytical reproducibility

Since 2003 we have repeatedly measured the same sample on each of our detectors (note that this sample, lab. code 950509A, was prepared in 1997 using polyester resin rather than wax). Typical re-producibility for $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ analyses are shown in Fig. 2 for detector D.

In general there are no clear signs of systematic changes in calibration when the detector was recalibrated and/or recommissioned after repair (usually after pumping). Only in the case of the $^{226}\text{Ra}$ time series is there a significant (9%) systematic change after 3563 days, associated with a recalibration (but not with repair).

From these radionuclide data, the calculated relative standard deviation in our total dry dose rate is 2.5%. We conclude that our analyses have been acceptably stable over a period of $\sim13$ years.
4. Sample presentation: effects of self-absorption

Before preparing new calibration standards, Monte Carlo modelling (MCNP) was used to investigate the likely variation in self-attenuation as a function of energy resulting from typical variations in the mass of ground sample in a prepared wax/sample mixture (the simulations were performed for a disc geometry, but the controlling factor, the sample thickness, is the same (10 mm) for both the cup and the disc geometry). The results of these calculations are summarised in Fig. 3, normalised to a standard 66% sample by weight. For all mixtures the maximum change in self-attenuation > 100 keV is 6%. For likely mixtures of both calibration standards and material to be analysed (sample/wax ratio > 50%) the change is < 2%. However at 46 keV (210Pb) the range in likely attenuation is considerable and 210Pb activities are correspondingly less certain.

5. Preparation of new standards

Rather than use the low activity standards diluted for the IAEA (IAEA, 1987) by NRCAN using their own high activity CRM's, we have chosen to use NRCAN's high-activity primary material directly; 5 independent dilutions were prepared and cast in wax to provide a measure of the reproducibility of our preparation procedure. We base our calibration on uranium ore (BL-5), thorium ore (OKA-2) (both with well-known parent activities, and both confidently expected to be in secular equilibrium - apart from radon isotopes and short lived daughters) and analytical grade K\textsubscript{2}SO\textsubscript{4} (Table 1). Secular equilibrium in BL-5 has been tested by NRCAN; they found a $^{226}$Ra/$^{238}$U activity ratio of $0.979 \pm 0.022$ (see Table 1). To provide relative gamma ray intensities from $^{226}$Ra and $^{222}$Rn daughters (with negligible contribution from U, especially $^{235}$U at 186 keV) two different $^{226}$Ra rich materials were used: (i) an uncalibrated (i.e. not accurately known) solution of $^{226}$Ra solution containing $\sim 200$ Bq g$^{-1}$, and (ii) an IAEA oil field sediment (IAEA-448) with a $^{238}$U/$^{226}$Ra activity ratio of < 0.3%. The standard ores and IAEA-448 were supplied to us crushed to < 100 μm particle size and homogenised, and the purity of the K\textsubscript{2}SO\textsubscript{4} requires that it is completely homogeneous.

Different portions of the standards BL-5 and OKA-2 were diluted by mixing with crushed low activity quartz rich sand (M32, Sibelco Belgium; $^{226}$Ra 1.39 ± 0.08; $^{232}$Th 0.85 ± 0.06, $^{40}$K 6.8 ± 0.7 Bq kg$^{-1}$). These mixtures were then ground to < 200 μm and cast in standard cup geometry to give 5 independently diluted standards based on each standard material; the notional activity of the U and Th cups was each $\sim 800$ Bq, sufficiently low so as to ensure that counting dead-time was < 1%. All weighing was to a precision of < 0.1%. Because of the high initial activities of the U and Th standards, it was necessary to undertake dilution and crushing in two steps, introducing an extra weighing uncertainty in the dilution. To provide the relative $^{226}$Ra standards, the pure solution was dried onto a portion of low activity sand, ground, homogenised and cast to give 2 cups each of $\sim 800$ Bq. In addition 3 cups were prepared from IAEA-448. Different portions of K\textsubscript{2}SO\textsubscript{4} were mixed directly with wax to give 20 cups.

The $^{40}$K standards received the least handling, and so should be most reproducible. Fig. 4 shows a histogram of the analyses of the 20 cups; the relative standard deviation (RSD) is 0.8% (n = 20). Using the Shapiro-Wilk test (Shapiro and Wilk, 1965), we cannot reject the hypothesis that the sample is taken from a normally distributed population ($W = 0.962 > W_p = 0.905$ for $p = 0.05$; $n = 20$). In contrast the $^{238}$U and $^{232}$Th standards were manipulated more, and their RSDs are 1.5% (n = 5) and 1.2% (n = 5) respectively.

The uranium and thorium standards were heavily diluted in quartz sand (respectively < 0.5% and < 2.5% of standard in the mixture by weight) to ensure that the attenuation characteristics of the cast mixture are indistinguishable from those of the quartz sand matrix. To confirm that this was also true of the potassium standard (pure K\textsubscript{2}SO\textsubscript{4} mixed only with wax) the mass attenuation coefficients at 1.46 keV (gamma energy from $^{40}$K) of a typical wax and K\textsubscript{2}SO\textsubscript{4} mixture (1:3 by weight) were compared with those of corresponding mixtures of quartz, and typical granite and basalt in wax. For an attenuation depth of 5 mm (i.e. average attenuation through ~ half cup wall thickness) the attenuation loss of 3.4% is indistinguishable for all 4 materials.

Finally three cups of pure wax were prepared for determining background count rates.

6. Accuracy as a function of activity concentration, and minimum detection limits

After initial calibration, it was considered important to investigate the size of any systematic deviation in analysis as a function of activity, and incidentally to determine the minimum detection limits. To this end we first prepared an additional mixed sample, containing $\sim 2700$ Bq $^{40}$K, and $\sim 60$ Bq each of $^{238}$U and $^{232}$Th, using K\textsubscript{2}SO\textsubscript{4}, BLS and OKA-2. The three nuclides are in proportions typical of sediment (see e.g. Ankjærgaard and Murray, 2007). This mixture was then cast and counted for various periods of time, from 30 s to 20 h. At the end of each counting period, the sample was removed and the detector allowed to continue counting background until a cumulative count time
of 20 h was reached. The resulting spectra were then equivalent to those obtained from counting samples of very well-known relative activities, ranging from \(4.2 \times 10^{-2}\%\) to 100% of the mixed sample activity concentrations. The observed deviations from the expected activity concentrations are plotted as a function of expected activity concentrations in Fig. 5a. Any systematic deviations from expected values do not appear until very low concentrations; these probably reflect inaccurate background corrections. The analytical uncertainties also begin to increase significantly below a few Bq.kg\(^{-1}\). Fig. 5b shows these uncertainties plotted against expected activity concentrations for all the measured radionuclides. Minimum Detection Limit (MDL) values are a very useful way of characterising an analytical facility; here we define the MDL as the concentration at which the relative analytical uncertainties exceed 30% - this is shown as a horizontal dashed line in Fig. 5b, and the derived MDLs are summarised in Table 2.

With the possible exception of \(^{210}\)Pb, these MDLs are much lower than all likely concentrations in real sediment samples. At low concentrations, our analyses are much more likely to be inaccurate (Fig. 5a) than imprecise (Fig. 5b). The likely systematic uncertainty at the MDL (taken from Fig. 5a) is listed in Table 2, column 3, together with the more useful likely uncertainty at 1 Bq.kg\(^{-1}\) (U series and Th series) and 20 Bq.kg\(^{-1}\) K (column 4).

7. Comparison with known and potential standards

To test our new calibration, various reference materials from IAEA and NRCAN were identified for measurement, together with two analytical-grade K salts. These are summarised in Table 3. Note that only the values labelled R (for Recommended) and C (for Certified) are considered reliable by the suppliers. The ratios of our analyses to the given values are also given in Table 3. In the discussion below, we do not consider sample preparation as a likely source of discrepancy, because we have shown that we are able to grind, mix and cast samples with a standard deviation of < 1.5% (see section 5 above). Note also that Nussi and Volkegem results are not included in average ratio calculations because the ‘given’ values are not considered sufficiently well known.

**Uranium** – Given the uncertainties, all our U analyses are considered acceptable, and the weighted average of the ratios of our analyses to the expected values is 1.029 ± 0.013 (n = 5).

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### Table 1
Summary of absolute calibration materials.

<table>
<thead>
<tr>
<th></th>
<th>(^{238})U, Bq.kg(^{-1})</th>
<th>(^{226})Ra, Bq.kg(^{-1})</th>
<th>(^{210})Pb, Bq.kg(^{-1})</th>
<th>(^{232})Th, Bq.kg(^{-1})</th>
<th>(^{40})K, Bq.kg(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>BL-5</td>
<td>875.4 ± 1.9 C</td>
<td>857 ± 19 C</td>
<td>866 ± 10 C</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>OKA-2</td>
<td>2.70 ± 0.05 C</td>
<td>–</td>
<td>–</td>
<td>117.4 ± 1.2 C</td>
<td>0.108 ± 0.003 P</td>
</tr>
<tr>
<td>K(_2)SO(_4)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>14.23 ± 0.03 S</td>
<td></td>
</tr>
</tbody>
</table>

* Natural Resources Canada (NRCAN) concentration given as 7.09 ± 0.03 %U (95% confidence).
* NRCAN concentration given as 2.893 ± 0.058 %Th, 218.6 ± 8.2 μg g\(^{-1}\) U, 0.34 ± 0.02 %K (95% confidence).
* BDH Analyt NORMAPUR, purity given as 100.4%.
* Weight concentrations converted to activity concentrations using data given in Guérin et al. (2011).
* C – certified, P - Provisional, S – stoichiometry.

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### Table 2
Minimum detection limits (MDL) and likely systematic uncertainty, for 250 g sample in wax/sample mix, 20 h count time.

<table>
<thead>
<tr>
<th></th>
<th>MDL, Bq.kg(^{-1})</th>
<th>Sys. uncert. (%) at MDL</th>
<th>Sys. uncert. (%) for U, Th: 1 Bq.kg(^{-1})</th>
<th>K: 20 Bq.kg(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{238})U</td>
<td>0.9</td>
<td>19</td>
<td>19</td>
<td>19</td>
</tr>
<tr>
<td>(^{226})Ra</td>
<td>0.04</td>
<td>140</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>(^{210})Pb</td>
<td>2</td>
<td>15</td>
<td>15</td>
<td>15</td>
</tr>
<tr>
<td>(^{232})Th</td>
<td>0.03</td>
<td>100</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>(^{40})K</td>
<td>0.6</td>
<td>16</td>
<td>4</td>
<td>4</td>
</tr>
</tbody>
</table>
of engineering activity (226Ra/238U activity ratio > 380). It also accu-
directly. This sediment is known to have accumulated 226Ra as a result
from the IAEA information value for 228Ac concentration (short lived
daughter of 228Ra) of 1166 ± 55 Bq.kg

The primary source of our calibration (NRCAN BL-5) was derived by
Preusser and Kasper (2001). The new analyses are also plotted inFig. 1
ed Reference Material
RG-Th-1, and this is probably at least in part the result of a normally
weak Th series interference (1459 keV, 228Ac) in the 1461 keV line from
40K. The 232Th to 40K ratio in RG-Th-1 is very far from what is typical in
natural sediments and non ore-grade rocks (see e.g. Ankjærgaard and
Murray, 2007), and so this result was omitted from the average ratio in
Table 3. The weighted average of our analyses to the expected values is
1.017 ± 0.010 (n = 5).

Radium – Our result underestimates the IAEA-448 certified value by 8%
but, as previously noted, the IAEA RGU-1 by 3% (this material is
manufactured by IAEA from the NRCAN Certified Reference Material
(CRM) BL-5 used for our primary calibration). Either the given value for
IAEA RGU-1 is wrong, or that for IAEA-448. Because these analyses are
significantly over-dispersed, the unweighted average is used to give the
ratio of our analyses to the expected values of 1.01 ± 0.04 (n = 3). The
primary source of our calibration (NRCAN BL-5) was derived by
pulverising a solid rock sample, and so we can be confident that the
238U series in this material is in equilibrium down to and including
226Ra. This assumption is confirmed by the certified values listed in
Table 1, and it is also consistent with the average of the NLL given
values of 1.029 ± 0.013 (n = 5) and 1.01 ± 0.04 (n = 3) for
238U and 226Ra analyses, respectively, (see Table 3). These data support
the suggestion that our calibration is accurate, but that there is true
variability in the ‘known’ materials.

Thorium – Again we probably overestimate the IAEA-314 value
(1.23 ± 0.08), and also the IAEA RGTh-1 (1.06 ± 0.02). The latter is
surprising, because both our standards and the IAEA material are based
on the same NRCAN CRM Th OKA-2. On the other hand, the over-
estimate of the IAEA-448 oil field sediment is not surprising; our ana-
lysis is based on 226Ra and daughters, whereas the IAEA reports 234Th
directly. This sediment is known to have accumulated 226Ra as a result of
engineering activity (223Ra/238U activity ratio > 380). It also accumu-
lated 226Ra (half life 6.7 years) in a similar manner – this is clear from the
IAEA information value for 241Am concentration (short lived daughter of 223Ra) of 1166 ± 55 Bq.kg

(a) Natural Resources Canada (NRCAN).
(b) International Atomic Energy Agency (IAEA).
(c) Preusser and Kasper (2001).
(d) De Corte et al. (2007).
(e) 1, R, C labels indicate Information, Recommended and Certified, according to the IAEA. S indicates calculated from stoichiometry.
(f) Analytical grade potassium salts with purity > 99.0%.
(g) Not included in average ratio.

Table 3

<table>
<thead>
<tr>
<th>Material</th>
<th>238U, Bq.kg⁻¹ (NLL given)</th>
<th>226Ra, Bq.kg⁻¹ (NLL given)</th>
<th>232Th, Bq.kg⁻¹ (NLL given)</th>
<th>40K, Bq.kg⁻¹ (NLL given)</th>
</tr>
</thead>
<tbody>
<tr>
<td>WGB-1 Gabro</td>
<td>9.8 ± 0.9</td>
<td>9.3 ± 0.6</td>
<td>4.58 ± 0.04</td>
<td>4.1 ± 0.2</td>
</tr>
<tr>
<td>IAEA-314</td>
<td>610 ± 53</td>
<td>701 ± 24</td>
<td>89 ± 2</td>
<td>72 ± 2</td>
</tr>
<tr>
<td>IAEA-448</td>
<td>25 ± 17</td>
<td>49 ± 0.5</td>
<td>19050 ± 260</td>
<td>134 ± 0.6</td>
</tr>
<tr>
<td>IAEA-RGU-1</td>
<td>5162 ± 69</td>
<td>4940 ± 15</td>
<td>n.a.</td>
<td>n.a.</td>
</tr>
<tr>
<td>IAEA-RGTh-1</td>
<td>63 ± 3</td>
<td>78 ± 3</td>
<td>n.a.</td>
<td>n.a.</td>
</tr>
<tr>
<td>KCl</td>
<td>n.a.</td>
<td>n.a.</td>
<td>n.a.</td>
<td>n.a.</td>
</tr>
<tr>
<td>K2HPO4</td>
<td>n.a.</td>
<td>n.a.</td>
<td>n.a.</td>
<td>n.a.</td>
</tr>
<tr>
<td>Nussi loess</td>
<td>37 ± 2</td>
<td>31.5 ± 12</td>
<td>35.5 ± 0.5</td>
<td>32.0 ± 1.4</td>
</tr>
<tr>
<td>Volkegem loess</td>
<td>37.8 ± 0.7</td>
<td>34.5 ± 1.5</td>
<td>42.77 ± 0.16</td>
<td>42.2 ± 2.5</td>
</tr>
<tr>
<td>Average ratio</td>
<td>1.029 ± 0.013</td>
<td>1.01 ± 0.04</td>
<td>1.099 ± 0.019</td>
<td>1.017 ± 0.010</td>
</tr>
</tbody>
</table>

- 1.029 ± 0.013 (n = 5)
- 1.01 ± 0.04 (n = 3)
- 1.099 ± 0.019 (n = 3)
- 1.017 ± 0.010 (n = 5)

Potassium – We are in good agreement with the two independent
K-salts, and the low activity Gabbo. We overestimate the 46K in IAEA
RG-Th-1, and this is probably at least in part the result of a normally
weak Th series interference (1459 keV, 228Ac) in the 1461 keV line from
46K. The 232Th to 40K ratio in RG-Th-1 is very far from what is typical in
natural sediments and non ore-grade rocks (see e.g. Ankjærgaard and
Murray, 2007), and so this result was omitted from the average ratio in
Table 3. The weighted average of our analyses to the expected values is
1.017 ± 0.010 (n = 5).

We conclude that our calibrations for the uranium series and 46K are
consistent with the expected concentrations in the independent mate-
rials, but that this is not true for our Th analyses which are consistently
higher than expected. In our view this deviation is most likely to ori-
ginate with discrepancies between the independent reference materials,
rather than in our sample preparation and counting procedures. The
reproducibility of the independently prepared U, Th and K calibration
cups (n < 1.5%) and the satisfactory U series and K analyses all indicate
that the uncertainties in our preparation procedures are very much
smaller than 10% (the size of the Th discrepancy). In addition, the IAEA
RGTh-1 is prepared from the high level OKA-2 NRCAN Th ore, which is
also the source of our Th calibration. And yet, as pointed out above, our
analyses of RGTh-1 is 6 ± 2% high.

It is interesting to compare our new calibrations with those for the
same detectors (J and K) using the previous set of calibration standards.
The effect of the new standards described in this paper on U and Ra
analyses is to decrease the concentrations by 1–2%. Thorium con-
centrations would be increased by 4% and K would decrease by 3%.
Given that the previous calibration materials had been made over the
period 1987-2008 by at least three different individuals we consider
this agreement very satisfactory.

All 20 of the inter-comparison samples were realanalysed on detectors
J and K after recalibration. The new analyses are also plotted in Fig. 1
and 40K) measured on one gamma detector, we conclude that our dose rate analyses have remained satisfactorily reproducible over at least 13 years. In order to test the accuracy of these measurements, we have prepared new calibration standards based on NRCAN U and Th ores, and analytical grade K₂SO₄. By independently preparing between 5 and 20 samples of each calibration material, it has been shown that uncertainties arising from preparation lead to an overall standard deviation of < 1.5%.

By counting a single mixed sample for different durations and then continuing to count background until all net measurement times are the same, the dependence of accuracy and precision on activity concentration has been measured on one of the three detectors. We observe that there are no significant systematic uncertainties in our analyses as the activity concentration decreases to well below those typical of unknown samples, and it is concluded that the minimum detection limits are not likely to be a limiting factor in the majority of these samples.

It has proved surprisingly difficult to obtain high quality samples with which to test our calibration, and even in the few we have used, there are significant inconsistencies in measured-to-known ratios between samples; these inconsistencies cannot be explained by our calibration procedure, unless they are attributed to sample preparation. However, since sample preparation has been shown to introduce uncertainties of < 1.5%, this is not considered a likely cause of these discrepancies.

Although the accepted ratios of our analyses to the given values are considered satisfactory for 238U, 226Ra and 40K, we overestimate the expected 232Th activity concentrations by ~10%, probably because of discrepancies in the values of independent materials. In general, however, this is not of major concern in most dose rate estimations, because, in our experience, 40K contributes around ~50% to the total dry dose rate in most coarse-grained samples, and the typical Th-series contribution is ~25%. In such a sample, a 10% uncertainty in a Th analysis resulting from a poor calibration corresponds to a dose rate uncertainty of 2.5%.

Acknowledgements

J.-P. Buylaert and M. Autzen received funding from the European Research Council (ERC) under the European Union Horizon 2020 research and innovation programme ERC-2014-StG 639904 – RELOS. We thank Dimitri Vandenberghe for the Volkegem loess sample and Andreas Lang and Barbara Mauz for a sample of Nussi loess.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.radmeas.2018.04.006.

References


