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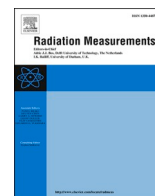
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Calibration quartz: An update on dose calculations for luminescence dating

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ABSTRACT

Previous calculations of the γ dose given to Risø Calibration Quartz (RCQ) were performed using mass absorption and mass attenuation coefficients. In this paper, we update the γ dose given to RCQ using Geant4 simulations and provide a comparison with three other γ sources in Denmark. We also show experimental evidence that the luminescence response to a ^{137}Cs γ irradiation is 4% higher than that resulting from a ^{60}Co γ irradiation for the same dose, confirming the suggestions of Autzen et al. (2021) based on simulations.

1. Introduction

Dating natural sediments using optically or thermally stimulated luminescence depends on accurate and precise calibration of the laboratory dose rate. Laboratory doses are typically given using a β source mounted on the luminescence reader. This source must be cross-calibrated against some independently calibrated source, usually a ^{60}Co or ^{137}Cs γ source, in order to provide an accurate dose rate. Such a comparison is undertaken by matching sensitivity-corrected light-levels, i.e.

$$D_{\gamma}\chi_{\gamma} = t\dot{D}_{\beta}\chi_{\beta}$$

where D_{γ} is the given γ dose, χ_{γ} and χ_{β} are the luminescence efficiencies for γ and β irradiations respectively, \dot{D}_{β} is the β source dose rate and t is the β irradiation time required to match the luminescence produced by the γ dose.

In practice the comparison also relies on the fundamental assumption that the luminescence response for a γ dose (χ_{γ}) is the same as that from a beta dose (χ_{β}) (Hansen et al., 2018), and so

$$\dot{D}_{\beta} = \frac{D_{\gamma}\chi_{\gamma}}{t\chi_{\beta}} \sim \frac{D_{\gamma}}{t} \quad (\text{Eq. 1})$$

The γ source is usually calibrated under certain reference conditions (e.g. at 2 m distance) using an ionisation chamber providing traceability, usually to a national or international accredited standard for air kerma (free in air) or dose to water (in a water phantom). The reference value must be converted to a dose to the medium of interest – in our case,

quartz.

Risø Calibration Quartz (RCQ, Hansen et al., 2015, 2018) and its predecessors have been used for calibrating β sources in laboratories worldwide for the past 30 years. In the past we have used mass absorption coefficient tables to calculate the conversion from air kerma in a ^{137}Cs beam to a given quartz dose, and mass attenuation coefficient tables to calculate the attenuation of the primary photon beam through the glass cell used to provide buildup of secondary electrons. Previous work (Bos et al., 2006) found good agreement between calibrations performed using Fricke dosimetry and RCQ, however, recent work by Tribolo et al. (2019) suggests that there may be a discrepancy in the calibrated dose rates achieved using RCQ and LexCal. This comparison has been investigated in Richter et al. (submitted).

In this paper we will first go through the past calculations of dose to RCQ (Section 2) based on mass-attenuation and mass-absorption coefficients in the two different irradiation geometries used for past and current batches. In section 3 we show that the past calculations have underestimated the actual dose due to neglecting scatter of the primary beam, as well as showing the impact of sample-source distance errors and backscatter on the final dose. Finally, we compare the new dose based on the results of section 3 with irradiations performed in other calibrated sources in order to gain additional confidence in the new dose (Section 4).

2. Past calculations of dose to RCQ

Until 2002, RCQ (Batch 6) was irradiated using a ^{137}Cs source in glass vials (Fig. 1a) taped to a cardboard sheet and packed in black plastic.

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Since 2002, irradiations have been carried out using a different but similar ^{137}Cs source (Nucomat facility, DTU Risø Campus, dose rate ~ 0.1 Gy/h at 2 m). In 2005 (Batch 7 onwards), the irradiation cell was changed to one with a planar geometry, with a wall thickness normal to the γ beam of 1.85 mm, see Fig. 1b. This change was driven by the need for a material suitable for the calibration of systems intended for the measurement of luminescence from individual grains. For this, it is important that each individual grain absorbs the same γ dose, and our preliminary calculations suggested that, using the new irradiation geometry, this would be true to within 1%.

Both ^{137}Cs sources were calibrated using an ionisation chamber to provide the Kinetic Energy Released per unit Mass (Kerma) in air; Kerma is defined as the sum of the initial kinetic energies of all charged particles created by uncharged ionising radiation, per unit mass of sample. The ionisation chambers used for these calibrations were traceable to primary standards at NPL (UK) or PTB (Germany). For more information on how the sources are accredited we direct the reader to the homepage of the Medical Dosimetry group at DTU Healthtech (<https://www.healthtech.dtu.dk/english/services-and-products/hdrl>). To ensure that secondary electron equilibrium was achieved (i.e. each electron entering the volume of interest containing quartz grains is balanced by an electron leaving) some solid material must be placed in front of the quartz grains to be irradiated; we use soda-glass, because its mass attenuation and absorption characteristics are almost identical to those of quartz. Kerma may then be equated to dose inside the sample, and the dose to quartz derived using:

$$D_Q = K_{air} * \left(\frac{\mu_{en}}{\rho}\right)_Q / \left(\frac{\mu_{en}}{\rho}\right)_{air} * A \quad (\text{Eq. 2})$$

where D_Q is the dose to quartz, K_{air} is the air kerma measured at the centre of the position of the quartz volume, $(\mu_{en}/\rho)_Q$ and $(\mu_{en}/\rho)_{air}$ are

the mass energy absorption coefficients for quartz and air respectively, and A is the attenuation through the glass wall and sample calculated using

$$A = \exp\left(-\left(\frac{\mu}{\rho}\right)_{\text{glass}} * \rho_{\text{glass}} * L_{\text{glass}}\right) * \exp\left(-\left(\frac{\mu}{\rho}\right)_{\text{quartz}} * \rho_{\text{quartz}} * L_{\text{quartz}}\right) \quad (\text{Eq. 3})$$

where $(\mu/\rho)_i$ is the mass attenuation coefficient for glass or quartz, ρ_i is the glass or quartz density and L_i is the glass or quartz effective attenuation path length. Mass energy absorption and mass attenuation coefficients for air, glass, Si, and O are shown in Table 1. This simple calculation assumes that end effects, and build-up of scattered radiation

Table 1
Mass energy absorption and mass energy attenuation coefficients.

Mass energy absorption coefficients, $\text{cm}^2 \text{g}^{-1}$					
Energy/ MeV	Si (μ_{en}/ρ_{Si})	O (μ_{en}/ρ_O)	^a Q (μ_{en}/ρ_Q)	Glass (μ_{en}/ρ_{Glass})	Air (μ_{en}/ρ_{Air})
0.600	0.0295	0.0296	0.0296	0.0294	0.0295
0.662	0.0293	0.0294	0.0293	0.0292	0.0293
0.800	0.0288	0.0289	0.0289	0.0287	0.0289
Mass attenuation coefficients					
Energy/ MeV	Si (μ/ρ_{Si})	O (μ/ρ_O)	Q (μ/ρ_Q)	Glass (μ/ρ_{Glass})	
0.600	0.0804	0.0805	0.0805	0.0800	
0.662	0.0774	0.0775	0.0774	0.0770	
0.800	0.0706	0.0707	0.0707	0.0703	

^a Data derived from Berger (1993) and Berger et al. (2005). $(\mu_{en}/\rho)_Q$ derived from $(\mu_{en}/\rho)_{Si}$ and $(\mu_{en}/\rho)_O$ following the recommendations of the National Institute of Standards and Technology, NIST (Berger, 1993; Berger et al., 2005), see text.

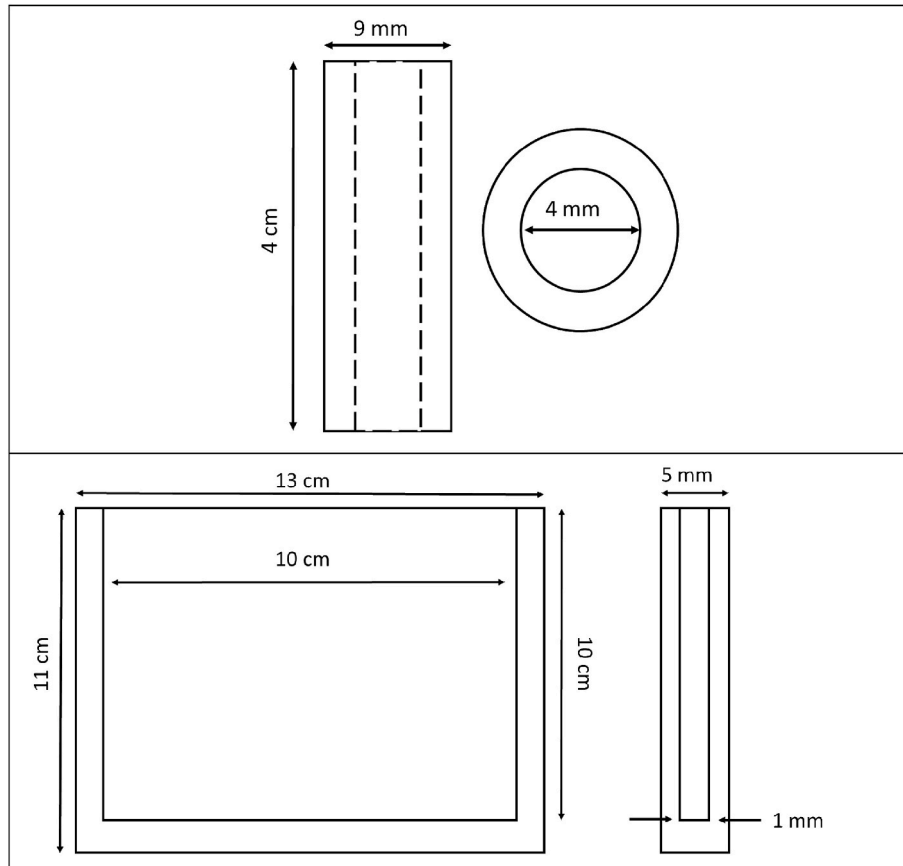


Fig. 1. a) Glass tubes used for irradiating Batch 1–6, b) Glass cell used for irradiating Batch 7 – present.

within the cell, are negligible; the resulting attenuation is small, as is described below.

In Table 1, we have calculated the mass absorption and mass attenuation coefficients for quartz by summing the relevant coefficients weighted by their relative mass fractions (Berger, 1993; Berger et al., 2005)

$$\left(\frac{\mu_{en}}{\rho}\right)_Q = \frac{m_{Si}}{m_Q} \left(\frac{\mu_{en}}{\rho}\right)_{Si} + \frac{2^*m_{O}}{m_Q} \left(\frac{\mu_{en}}{\rho}\right)_O \quad (\text{Eq. 4})$$

We also interpolate linearly between 0.6 and 0.8 MeV in order to calculate the mass absorption and mass attenuation coefficients for the primary γ energy of 0.662 MeV; no significant difference is observed if logarithmic interpolation is used. From Table 1, $(\mu_{en}/\rho)_Q/(\mu_{en}/\rho)_{air} = 1.0008$ at 662 keV; the mass energy absorption coefficients are indistinguishable, and so any difference between air kerma and quartz dose will arise from attenuation of the primary beam through the cell wall and half of the sample thickness.

In our calculations we assumed that the maximum path length of a 0.662 MeV electron in glass is 0.3 mm; this was subtracted from the total wall thickness (because the dose absorbed at a point is derived from photons that interact upstream of that point). Until 2002, attenuation path lengths of 1.7 mm in glass ($\rho = 2.66 \text{ g cm}^{-3}$) and 1.675 mm in quartz ($\rho = 2.00 \text{ g cm}^{-3}$) (to the middle of the quartz sample) were assumed. Using Eq. (3), and an assumed (but realistic) packing density of 2 g cm^{-3} , this gave attenuation factors of 0.966 and 0.975, respectively, and a total attenuation of $0.966 \times 0.975 = 0.941 \pm 0.010$, where we have estimated the uncertainty on the attenuation; all uncertainties in this paper are presented at a coverage factor $k = 1$.

After the change to the planar geometry (in 2005), these calculations were revised with an effective glass wall thickness of 1.6 mm and a density of 2.66 g cm^{-3} and a half mean path length through the sample of 0.5 mm with an effective density of 2.00 g cm^{-3} . Using the same calculations as before gives a total attenuation of 0.965 ± 0.010 , where we have again estimated the uncertainty on the attenuation. From these data, a standard exposure equivalent to an air Kerma of 5.00 Gy leads to a dose of $4.71 \pm 0.07 \text{ Gy}$ until 2002 and $4.82 \pm 0.07 \text{ Gy}$ after 2002.

3. Dose calculations using Geant4

To test the validity of the explicit calculations summarised above, we have simulated the flat irradiation geometry in Geant4 (Agostinelli et al., 2003; Allison et al., 2006) based on Fig. 1b. The planar geometry was simulated by dividing the 5 mm thick cell (Fig. 1b) into five 1 mm thick, $10 \times 10 \text{ cm}$ slices of quartz ($\rho = 2.65 \text{ g cm}^{-3}$) or air ($\rho = 0.00120479 \text{ g cm}^{-3}$) stacked against each other facing the beam. As the geometry is scatter-free we can assume that the γ beam is parallel in the position of the sample (sample located 2 m from the origin) and the source is simulated as a square emission located at the origin with parallel emission in the direction of the sample. The kerma in the quartz sample was calculated as the initial energy of electrons created by photons in each 1 mm slice. The dose in each slice was recorded in the same simulation.

The results of these simulations are summarised for the various depths in the container in Table 2, using a normalised number of emitted photons emitted as a parallel beam of cross section identical to that of the sample ($10 \times 10 \text{ cm}$). A simulation was also run with a point source and a radial emission of photons, but no difference between the simulations was observed.

The simulations (Table 2) show that, as expected, the quartz-to-air kerma ratios are close to unity through the length of the sample holder. However, the dose-to-kerma ratio for quartz is only significantly below 1 in the first millimetre; further into the cell the ratio of dose to kerma increases as the secondary electron flux builds up until it is again indistinguishable from unity.

We further tested a more accurate simulation of the sample holder, created to the same specifications as seen in Fig. 1b, with the glass (the

Table 2

Results of simulating kerma and dose to air and quartz in the irradiation post-2005 geometry. Uncertainties given at $k = 1$ level.

Planar geometry (2005 – Present)					
Depth/ mm	K _{Air} (pGy)	K _Q (pGy)	D _Q (pGy)	K _Q /K _{Air}	D _Q /K _Q
0.5	304.8 ± 3.4	314.5 ± 0.4	269.1 ± 0.3	1.03 ± 0.01	0.86 ± 0.01
	302.8 ± 3.4	312.9 ± 0.3	312.4 ± 0.3	1.03 ± 0.01	1.00 ± 0.02
1.5	313.9 ± 3.5	311.5 ± 0.3	310.1 ± 0.3	0.99 ± 0.01	1.00 ± 0.02
	309.1 ± 3.4	309.1 ± 0.3	308.8 ± 0.3	1.00 ± 0.01	1.00 ± 0.02
2.5	309.4 ± 3.5	306.9 ± 0.3	298.1 ± 0.3	0.99 ± 0.01	0.97 ± 0.02
	309.4 ± 3.5	306.9 ± 0.3	298.1 ± 0.3	0.99 ± 0.01	0.97 ± 0.02

material was chosen as SiO₂) having a density of 2.38 g cm^{-3} and filling the sample space with 0.1 mm thick slices of reduced density quartz ($\rho = 1.78 \text{ g cm}^{-3}$), which is more comparable to loosely-packed grains, and were unable to detect a difference in the relationship between air kerma and quartz dose compared to that of Table 1. Similarly, we also tested the effect of the density of the glass by varying the density between 2.00 and 2.65 g cm^{-3} and found no difference between the results.

3.1. Impact of scattering on initial calculations

The simulations in the previous section showed that the ratio between air kerma and quartz dose was indistinguishable from unity at the points in the sample volume. This is in contrast to the calculations described above in section 2, where we predicted an attenuation of 3.5% at the sample position. In the earlier calculations we only considered the behaviour of the primary 662 keV photon beam; the build-up of secondary (scattered) photons within the irradiation cell was neglected. In the Geant4 simulations, if we disregard interactions with scattered photons (i.e. only consider interactions with the primary beam) the dose-to-kerma ratio at 2.5 mm into the cell decreases to 0.97 ± 0.01 , consistent with our simple calculations.

3.2. Source-sample distance, backscatter and build-up in air

The ¹³⁷Cs source was calibrated with the centre of the ionisation chamber at a distance of 0.6 m from the source, but quartz irradiations were performed at a distance of 2.0 m. In these simulations, we assumed that the sample volume is centred on 2.0 m in air, with no support material behind the irradiation cell. In reality, the glass cell was mounted on the front of a 5 mm thick perspex plate (simulated using included NIST Material in GEANT4, $\rho = 1.19 \text{ g cm}^{-3}$), and the front of this plate was mounted at 2.0 m distance from the source. This moved the centre of the sample volume forward by approx. 3 mm, leading to an increase in dose of 0.2%. However, at some point this 5 mm plate was replaced by a 10 mm plate, with the back edge at 2.0 m. This led to an undetected positioning error, with the centre of the sample lying 1.0–1.5 cm closer to the source and gave rise to a 1.0–1.5% increase in dose given to the quartz. Modelling suggests that the presence of the perspex plate will itself lead to an increase in absorbed dose of 1.6% (because of secondary photons backscattered towards the sample). The calibration of the source at 0.6 m was extrapolated to 2.0 m using the inverse square law, and build-up in the intervening 1.4 m of air was neglected. A new calibration with the ionisation chamber at 2.0 m was undertaken, and this showed that the previous dose had been underestimated by 1.3% due to the neglect of air build-up. These factors are all summarised in Table 3.

Only for very few sub-batches used to make a single batch would we expect the error in the positioning of the cell to be 1.5 cm in total. In the calculation below we have assumed a distance of 1.25 cm (leading to 1.25% increase in dose) to be the most likely. Since these correction

Table 3
Sources of error for dose estimate.

Error component	Increase in dose compared to original calculation (with estimated uncertainty, $k = 1$)
Contribution from build up of scattered photons in cell	$3.5 \pm 0.5\%$
Error in positioning	$1.25 \pm 0.25\%$
Backscattered photons from support material	$1.6 \pm 0.5\%$
Contribution from build-up in air	$1.3 \pm 0.5\%$
Relative combined dose change	$8.25 \pm 1.00\%$

factors are ratio quantities close to unity, other non-statistical components of the uncertainty will cancel.

4. Comparing RCQ with material irradiated at other sources

Having identified a number of errors in previous batches of RCQ, batch 122 was irradiated in the Nucomat facility at Risø and the dose calculated making allowance for all the factors listed in Table 3. To gain additional confidence in the dose given, we compare with irradiations performed in three other γ sources. Unirradiated calibration quartz (Batch #122) was used for the comparison.

(a) Danish Health Authority – Radiation Protection (SIS)

A ^{137}Cs point source in a scatter-free geometry at the Radiation Protection division (SIS) of the Danish Health Authority with air kerma calibration traceable to PTB, Germany. We used our planar geometry and a new sample holder with no rear support to avoid backscatter and ensure that the sample was centred on the desired distance. The sample holder was centred on a distance of 1 m from the source and given 2.00 Gy air kerma, with an estimated total calibration uncertainty of 1.4% ($k = 1$). This air kerma is, according to the modelling presented in Table 2, equivalent to a dose to quartz of 2.00 ± 0.03 Gy.

(b) Therapy Cobalt source (LFMD)

A ^{60}Co point source is located at the Laboratory for Fundamental Medical Dosimetry (LFMD) at DTU Health, Risø Campus, with traceability to PTB for absorbed dose to water. The sample was irradiated in a plastic housing designed for ionisation chambers at a distance of 1 m and given a dose to water of 4.956 ± 0.017 Gy ($k = 1$) at 5 cm depth centred in a 30 cm \times 30 cm \times 30 cm water phantom. The sides of the water phantom were simulated as 5 mm thick perspex with a 0.5 cm perspex window. A further irradiation was carried out with the sample suspended in water inside the plastic housing at a depth of 5 cm, at the same location.

The source geometry was simulated according to the specifications provided. The ionisation chamber sleeve was modelled as a perspex tube ($\rho = 1.19$ g cm^{-3}) with an outer diameter of 6 mm and an inner diameter of 5 mm. The detector was simulated as a water ($\rho = 1.00$ g cm^{-3}) or quartz cylinder with a height of 26 mm, centred on the plane of the beam. The ^{60}Co source was simulated as a point source at 1 m distance from the centre of the sample with a radial emission due to the water phantom. Using these parameters we obtained a ratio of dose to quartz to dose to water of 0.944 ± 0.02 (at $k = 1$).

(c) Gamma Cell 1 (GC1)

This is a shielded ^{60}Co laboratory irradiator with cobalt source pencils arranged in a ring around the irradiation position (inner diameter of the source ring ~ 150 mm) located at the High Dose Reference at DTU Health, Risø Campus, and traceable to the National Physical Laboratory (NPL), UK; this is a national standard for Denmark. The sample was irradiated in a thin plastic tube surrounded by build-up material and

given a dose to water of 5 Gy. These laboratory irradiators have been simulated using the EGSnrc Monte Carlo code (Kawrakow et al., 2017), and a similar conversion factor was found between dose to water and dose to quartz.

These various known-dose samples were used to calibrate the beta source on two OSL readers (laboratory code ‘D’ and ‘E’) and the resulting calibrations are listed in Table 4. The aliquots were measured in pairs on each reader, so for each aliquot of the different known-dose samples, an aliquot of RCQ (Batch #122) was also measured on the same wheel. We have taken the average and standard uncertainty of the pair ratios to avoid variability resulting from run to run variations.

We observe agreement between the two ^{137}Cs sources (Risø and SIS) and between the two ^{60}Co sources, however we observe a consistent 4% difference between ^{137}Cs and ^{60}Co . Autzen et al. (2021) recently suggested that the luminescence generated by a ^{137}Cs source should be 4% higher than that generated by a ^{60}Co for the same dose because of the slight differences in ionisation density produced by the two different gamma energies. Guérin et al. (2018) observed an energy dependence on the luminescence response of natural quartz when irradiating with low energy x-rays compared to ^{137}Cs . Here, we observe this predicted 4% difference between our two types of sources. Correcting for this difference in luminescence generation we get Table 5.

Following the correction to the expected luminescence from ^{60}Co we observe complete agreement between all five sources used in the experiments. Table 4 supports the modelling hypothesis that the dose given to RCQ should be increased by 8% to agree with the other irradiation sources. It is interesting to note that the dry and wet irradiations using the LFMD ^{60}Co source does not result in a difference in dose rate. This suggests that the medium between the grains does not played a significant role in the final dose absorbed by the grains.

5. Discussion and conclusion

Using Geant4 modelling we have tested the validity of the simple calculations of γ dose delivered to RCQ used over the past 30 years. Modelling shows that while these calculations were indeed correct, they were also incomplete. Because the contributions from photons scattered within and from behind the irradiation cell were neglected, and because the cell was inadvertently placed 1–1.5 cm closer to the source than intended, the γ dose given to RCQ was underestimated by 8% prior to Batch #122. Using the new estimate of given dose gives results consistent with those obtained using the same source material irradiated in by different sources traceable to both PTB (the same primary standard used for RCQ), NPL and Bureau International des Poids et Mesures (BIPM, France).

The exact correction factor needed for each batch of RCQ (see Table 6) is not well-known, because of small variations in the distance to the source and the amount of backscatter through time. We estimate the average increase to be 8.25%, with an uncertainty of 2%. This correction should be applied to RCQ calibrations from Batch 1 to Batch 121.

Table 4

Comparison of dose rates from the same source material irradiated with different sources. The irradiated sample was divided into multiple aliquots for luminescence readout as indicated.

Gamma irradiation facility	N (# of paired aliquots)	Dose given	Dose Rate Ratio to Standard Irradiation
^{137}Cs , Nucomat (RCQ standard)	–	5.22 ± 0.05 Gy	1
^{137}Cs , SIS	76	2.00 ± 0.03 Gy	1.001 ± 0.011
^{60}Co , GC1	76	4.53 ± 0.04 Gy	1.043 ± 0.014
^{60}Co , LFMD (Dry)	36	4.49 ± 0.01 Gy	1.042 ± 0.013
^{60}Co , LFMD (Wet)	24	4.49 ± 0.01 Gy	1.043 ± 0.013

Table 5

Comparison of dose rates from the same source material irradiated with different sources, corrected for the modelled difference in ionisation density of ^{137}Cs and ^{60}Co .

Gamma irradiation facility	N (# of paired aliquots)	Dose given	Dose Rate Ratio to Standard Irradiation
^{137}Cs , Nucomat (RCQ standard)	–	5.22 ± 0.05 Gy	1
^{137}Cs , SIS	76	2.00 ± 0.03 Gy	1.001 ± 0.011
^{60}Co , GC1	76	4.53 ± 0.04 Gy	1.003 ± 0.014
^{60}Co , LFMD (Dry)	36	4.49 ± 0.01 Gy	1.002 ± 0.013
^{60}Co , LFMD (Wet)	24	4.49 ± 0.01 Gy	1.003 ± 0.013

Table 6

Average batch specific dose correction.

Batch No.	Dose on package	Correction needed
1–121	4.82 Gy (Some variation in earlier batches)	8.25%
122–125	5.00 Gy	4.4%
126 -	6.00 Gy	0.0%

Starting with batch 122 in October 2019, the dose was corrected to 5.00 Gy in response to our updated simulations, however, this correction did not take into account the distance and backscatter corrections, and so calibrations performed with batch 122 to 125 should be corrected by an average of 4.4%. Newer batches of RCQ (Batch 126 and onwards) now use the fully updated correction factors and can be identified by the given dose of 6.00 ± 0.12 Gy.

We observe that the luminescence generated from a ^{137}Cs source was 4% higher than the luminescence generated from a ^{60}Co source for the same dose, in agreement with predictions from the modelling of Autzen et al. (2021). This has significant implications for luminescence dosimetry, because the luminescence generated by different γ sources is usually regarded as energy independent. Further investigations are necessary to determine how far these differences extend but preliminary modelling suggests that identical gamma doses from different isotopes or spectra may not give rise to an identical luminescence response.

Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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