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ENVIRONMENTAL RADIATION MEASUREMENTS USING LITHIUM FLUORIDE THERMOLUMINESCENCE DOSEMETERS*

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Abstract — The National Radiological Protection Board is at present involved in a large scale environmental survey of radiation levels in homes throughout the United Kingdom. Passive radon and gamma ray dosemeters are posted to a representative sample of households. Lithium fluoride thermoluminescence dosemeters are used to assess natural gamma radiation and are left in the measurement location for a period of six months before being returned to the Board for processing. As a preliminary to the national survey, the Board has been engaged on several limited surveys in regions of igneous and sedimentary geology. Experience gained in these limited surveys and from standardisation studies using environmental dosemeters are reported.

INTRODUCTION

The National Radiological Protection Board is currently conducting a postal survey of natural radiation levels in domestic dwellings. This survey is part of a programme to improve knowledge about the exposure of the population of the United Kingdom to radiation of natural origin(1). The dosemeters used are thermoluminescence devices to measure external radiation from terrestrial and cosmic sources, and passive radon dosemeters based on the etched track technique to measure the radon-222 gas concentration(2). As a preliminary to the main survey, pilot surveys were undertaken to check the adequacy of the measurement techniques(3). The performance of the thermoluminescence devices is considered in this paper. Experience gained in participating in an international standardisation exercise is also reported.

PASSIVE DOSEMETERS FOR GAMMA RAY MEASUREMENTS

The average annual effective dose equivalent to the population of the United Kingdom from external natural radiation has been assessed to be about 700 μSv(4). About 300μSv is due to cosmic radiation, and about 400 μSv to terrestrial radiation from naturally occurring radionuclides in the ground and in building materials. The requirement to measure mean environmental dose equivalents of about 350 μSv over a period of six months can be met by certain types of thermoluminescent material, such as the standard TLD 100 chip.

Each environmental dosemeter package consists of a plastic phial containing three thermoluminescent elements, each sealed in a colour coded plastic sphere of 5 mm wall thickness. One of these elements is given a radiation dose (5 mGy) before issue, and is used to monitor fading of the thermoluminescence signal during the period of issue. The other two elements are used to measure the environmental dose. The encapsulation of the thermoluminescence devices is to ensure mechanical protection, the correct environment for accurate gamma radiation measurement (build-up), and protection against light and moisture. Tests have shown that self-irradiation due to radionuclides in the dosemeter and its holders is negligible.

Dosemeters are initialised by a repeated annealing procedure to stabilise both sensitivity and zero dose response. This procedure involves four cycles of annealing consisting of 1h at 400°C followed by 24h at 80°C. The thermoluminescent chips are given a calibration dose of 5 mGy (^{137}Cs) and read both prior to issue and upon their return. Since the dosemeters are reusable, a history of calibration factors is accumulated on each dosemeter element and used to assess long-term performance. For each batch of dosemeters issued, a set of background controls is kept in the laboratory in an area with a low radiation dose rate. This allows any spontaneous increase in the background signal of the dosemeters to be measured. Two further transit control elements are used with dosemeters issued by post. These are returned promptly by the householder, and are used to detect whether any dose was received or any signal fading occurred during transit. When dosemeters are recalled after their period of measurement, transit controls are again used to monitor doses and fading on the return journey.

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A Vinten 654 (Toledo) thermoluminescence reader was used to integrate the signal from the TLD chips for 24s between 160° and 240°C. A heating rate of 10°C.s^{-1} was used and dosemeter elements were given an anneal in the reader of 16s at 300°C followed by 24 h at 80°C before re-use. Nitrogen gas at a flow rate of 400 ml.s^{-1} was used to reduce background effects. A routine procedure for processing and packaging dosemeters was adopted whereby dosemeters for 25 houses could be processed per week. This procedure involved reading and re-reading dosemeters on day 1 and annealing overnight, exposing dosemeters to a calibration dose on day 2 and reading the dosed material on day 3 to establish calibration factors. After annealing overnight, dosemeters were packaged on day 4 and logged ready for issue on day 5. Dosemeters returning from issue were re-introduced into this processing procedure. The environmental dose was calculated from the measurement parameters by the procedure given in the appendix.

COSMIC RADIATION AND SURVEY SITES

The dose rate from cosmic rays varies only slightly throughout the United Kingdom. However, the dose rate from terrestrial gamma radiation can differ significantly with location. The contribution from cosmic radiation can be measured in an environment (such as over water) which shields the measurement devices from the terrestrial gamma radiation. This contribution can be subtracted from the total response of the measurement device to assess the terrestrial component. Thus, the response of the external radiation dosemeters to cosmic radiation was measured on the water surface of a reservoir 11.5 m above the reservoir base and 100 m from the nearest shoreline. The reservoir is raised above the surrounding countryside and is at a height of approximately 70 m above sea level. The dosemeters were sealed inside a desiccated container and placed inside a buoy which was moored on the surface of the reservoir. All components of the buoy and its contents were checked for inherent radioactivity, which was negligible. Land controls were placed close to the reservoir. The results of these measurements compared with instantaneous measurements for the international standardisation exercise are shown in Table 1. Instantaneous measurements were made using an energy compensated Geiger-Müller tube specifically developed for environmental measurements[5].

The pilot survey of external radiation in dwellings was carried out over a period of one year in about 140 homes of Board staff. The majority of homes were brick built two storey houses concentrated in predominantly chalk or clay regions.

Participants living in accommodation of two or more storeys were issued with two identical sets of dosemeters, one for the main living area and one for the main bedroom. The average values of absorbed dose rates in aior due to gamma rays measured in living areas and bedrooms were 0.036 and 0.035 μGy.h^{-1}, respectively. In two different areas of the United Kingdom with different geological characteristics, both passive and instantaneous measurements were taken inside dwellings. In a predominantly granite region, the mean values of absorbed dose rates in air for 18 houses due to gamma rays were measured as 0.095 amd 0.105 μGy.h^{-1} based on passive and instantaneous measurements respectively. In a predominantly sedimentary rock region, the equivalent mean values for 20 houses were 0.035 and 0.045 μGy.h^{-1} respectively.

The main lesson learnt from the pilot surveys was the need to simplify and, wherever possible, to automate laboratory procedures. To assist in this for larger scale surveys, an automatic sample changer has been fitted to the thermoluminescence reader and the processing cycle is controlled automatically by a microprocessor unit. This unit also collects data and stores glow curve information. In general, there were no major problems in the performance of the thermoluminescent chips. Significant changes in sensitivity over the issue period were observed with a few dosemeters, but in the majority of cases, the pre-calibration and post-calibration factors agreed to better than 5%.

INTERNATIONAL STANDARDISATION

The international standardisation exercise was organised by the Board and involved six organisations within European Community countries (Denmark, Germany, Italy, the Netherlands and the United Kingdom). Each organisation provided two measurement sites where environmental dosemeters from all of the participants were placed. One type of site was a relatively low activity site with an absorbed dose rate in the range of 0.04 - 0.09 μGy.h^{-1} in air. The other site was higher in activity with absorbed dose rates in the range 0.11 - 0.6 μGy.h^{-1} in air. The sites included domestic buildings, nuclear and laboratory sites, as well as open field and moor environments. Each participant also had a low background environment for storage of control dosemeters with absorbed dose rates generally below 0.025 μGy.h^{-1} in air. Each laboratory provided sets of transit control dosemeters for each measurement site and these controls were stored in the low background environment provided by each organisation during the measurement period. The parameter used for the comparison of the results

Table 1. Summary of the results of the international standardisation

Country	Site	Mean Total Dose Rate (in air) (E-T) (μGy h^{-1})* ± % standard deviation		
		All Dosemeters	NRPB dosemeter	Instantaneous**
Denmark	Moor	0.037 ± 3%	0.038 ± 2%	0.04
	Field (Nat. U)	0.119 ± 4%	0.123 ± 1%	0.120
Germany	Concrete building (12th floor)	0.035 ± 5%	0.038 ± 1%	0.046
	Brick House	0.100 ± 1.2%	0.099 ± 1%	0.069
Italy	Laboratory	0.061 ± 5%	0.066 ± 1%	0.068
	House	0.259 ± 1.3%	0.263 ± 2%	0.259
Netherlands	House	0.034 ± 4%	0.034 ± 2%	0.024
	Nuclear site	0.221 ± 3%	0.225 ± 3%	0.282
United Kingdom	Field	0.034 ± 5%	0.035 ± 1%	0.019
	Nuclear site	0.559 ± 4%	0.585 ± 2%	0.479
United Kingdom	Office	0.035 ± 5%	0.036 ± 1%	0.029
	Laboratory	0.262 ± 7%	0.259 ± 3%	0.236
	Reservoir	0.009 ± 11%	0.010 ± 1%	0.017

* Includes terrestrial + cosmic components (TLD measurements based on a γ calibration).
E is the absorbed dose rate (in air) measured by the TLD at the environmental site and T is the absorbed dose rate (in air) measured by the TLD at the low background storage facility.

** Difference between instantaneous measurements of absorbed dose rate at the environmental site and at the low background storage facility.

obtained for each thermoluminescence dosemeter was (E-T) where E is the absorbed dose measured by the thermoluminescence dosemeter at the environmental site and T is the absorbed dose measured by the transit dosemeter stored in the low background facility at the site. A summary of the results of the standardisation exercise which covered a period of six months is given in Table 1. Instantaneous measurements of exposure rate were taken at each of the measurement sites. Two organisations used LiF TLD-100 chips; two used LiF TLD-700 chips; one used CaF_2:Mn powder in glass bulbs; and one used CaF_2 TLD-200 chips. Plastic and/or aluminium were used to provide build-up around the dosemeters. Calibration facilities using ^{137}Cs and ^{60}Co γ radiation sources were offered to all participants by the Board. For a calibration absorbed dose of 8.73 mGy in air (± 1%) the mean assessment from the participants dosemeters for ^{137}Cs was 8.83 mGy in air (± 4%) and for ^{60}Co, 8.56 mGy in air (± 2%).

SUMMARY

The results from field trials and a standardisation exercise of the Board's environmental gamma radiation dosemeter are encouraging. The dosemeter, developed specifically for long-term measurement of natural radiation in dwellings, incorporates several TLD 100 chips. The agreement among these chips at a given measurement site is generally to within ± 3%. In dwellings, where both passive and instantaneous measurements were recorded, results agree to within ± 15%. In the standardisation exercise, where a wider variety of environmental sites were available, agreement between the results of all thermoluminescence dosemeters is generally better than 5%. In some cases, the passive measurements agree closely (better than a few %) with the instantaneous measurements (particularly, where high pressure ionisation chambers were used). The results of the environmental survey and the standardisation exercise will appear in fuller reports at a later date.

APPENDIX

The rate of increase of environmental signal

$$B = \frac{dx}{dt} + f\,x$$

where x = measurement counts
f = fading factor
t = time

multiplying through by exp ft and integrating gives

$$\frac{d\,(x \exp ft)}{dt} = B \exp ft$$

$$\text{thus } x \exp ft = \frac{B}{f} \exp ft + C \qquad \ldots (1)$$

$$\text{when } t = O,\ x = O\ \therefore C = -\frac{B}{f}$$

Substituting back in (1) gives

$$x \exp ft = \frac{B}{f}(\exp ft - 1)$$

when $t = T$ (the time of measurement), $x = X$

$$\therefore\ X = \frac{B}{f}(1 - \exp - fT)$$

$$\text{or } B = Xf\,(1 - \exp - fT)^{-1}$$

The rate of fading

$$\frac{dy}{dt} = -fy$$

The solution of this equation is $y = k \exp - ft$
for $t = 0$, $y = k$ $\quad\therefore y = k \exp - ft$
for $t = T$, $y = j$, $\quad\therefore j = k \exp(-fT)$

$$f = -\frac{1}{T}\log_n (j/k)$$

where k = calibration counts on fading controls
j = measurement counts on fading controls after $t = T$

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