Conf-760202--2

PREPRINT UCRL- 77106

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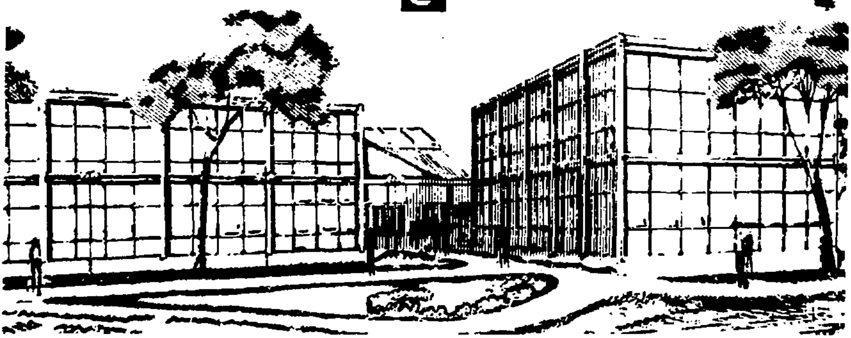
November 21, 1975

This paper was prepared for presentation at the Health Physics Society Ninth Midyear Topical Symposium on Operational Health Physics, Denver, Colorado, February 9-12, 1976.



AMBIENT ENVIRONMENTAL RADIATION MONITORING AT THE LAWRENCE LIVERMORE LABORATORY*

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Abstract

Thermoluminescence dosimetry is the principal means of measuring ambient gamma radiation at the Lawrence Livermore Laboratory. These dosimeters are used at 12 perimeter locations and 41 locations in the off-site vicinity of the Laboratory, and are exchanged quarterly. Control dosimeters are stored in a 75-mm-thick lead shield located out-of-doors to duplicate temperature cycling of field dosimeters. Effect of dosimeter response to radiation in the shield is determined each quarter. Calibration irradiations are made midway through the exposure cycle to compensate for signal fading. Terrestrial exposure rates calculated from the activities of naturally occurring uranium, thorium, and potassium in Livermore Valley soils vary from 3 to 7 μR/hr. Local inferred exposure rates from cosmic radiation are approximately 4 μR/hr. TLD measurements are in good agreement with these data. Off-site and site perimeter data are compared, and differences related to Laboratory operations are discussed.

Introduction

Today's emphasis on environmental monitoring is primarily due to public concern for environmental quality, which gained national attention during the late 1960s. For nuclear installations, ambient gamma radiation measurements are normally an integral part of environmental monitoring. Typically, thermoluminescence dosimetry is the principal means of making these measurements. The Lawrence Livermore Laboratory (LLL) locally operates a number of nuclear facilities having a potential for radiological impact. Our ambient monitoring program assesses any such impact.

At LLL, thermoluminescence dosimeters are used at 12 perimeter locations and 41 locations in the off-site vicinity of the Laboratory. This paper describes the TLD procedures which have been developed over the last six years. In addition to local ambient gamma monitoring, the same basic technique has been applied in a survey of geographical variations in environmental radiation background in the United States,(1) variations in environmental radiation between residences,(2) and radiation surveys both at Enewetak(3) and Bikini atolls.(4)

Dosimetry Procedures

Selection of Phosphors

The environmental TLD package contains two types of phosphors: CaF_2:Dy (TLD-200) and ^{7}LiF (TLD-700). The CaF_2:Dy phosphor was chosen because of its greater sensitivity compared with LiF. The LiF is included in the package to provide a more refined analysis of the field exposure (see Discussion) and ^{7}LiF (TLD-700) was used because of its low response to neutrons compared with naturally abundant lithium.

The CaF_2 and LiF phosphors in the form of 3-mm square chips were purchased in lots of 1000. When received, they were annealed at 425°C for 1-1/2 hr, cooled at ambient temperature for 40 min, and then post annealed at 80°C for a 24 hr period. After cooling and irradiating the phosphors with ^{60}Co, the annealing steps are repeated, followed by re-irradiation. After a 48 hr delay to allow low temperature traps to decay, the phosphors are then read using the LLL developed hot gas reader.(5) The phosphors selected for environmental monitoring have light output signals within ± 5% of the respective sets.

Dosimeter Package

To take advantage of automated processing capabilities developed for the Laboratory's personnel dosimetry program,(6) we also use standard LLL dosimeter holders for our environmental package. This injection molded plastic holder shown in Figure 1 is 350 mm in diameter and 3 mm thick. The two sets of holes around the circumference are a binary encoding system to identify the phosphor and monitoring location. Freshly annealed chips are placed in the three central recesses and are held in place by plastic covers. Although the recesses have a slight negative draft and the covers are stressed to make a tight fit, weathering

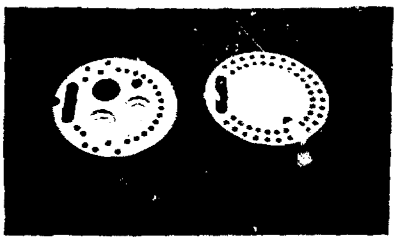

Fig. 1. LLL environmental TLD holder.

*Work performed under the auspices of the U.S. ERDA under contract No. W-7405-Eng-48.

in the field frequently caused the covers to pop out resulting in loss of the phosphor. To eliminate such losses, we now use two facing holders held together with plastic ties as shown in Figure 1. This double badge containing one holder with three CaF_2:Dy phosphors and one holder with three LiF phosphors was first used in an Enewetak radiation survey.(3)

Siting of TLDs

Locations of perimeter and off-site TLD monitoring stations are shown in Figures 2 and 3, respectively. Dosimeters are exposed for a period of three months. Perimeter dosimeters are attached to the woven wire Site boundary fence at a height of approximately 1 metre above the ground. As the vicinity of the Laboratory is essentially rural, the off-site dosimeters are usually secured to roadside wire fencing.

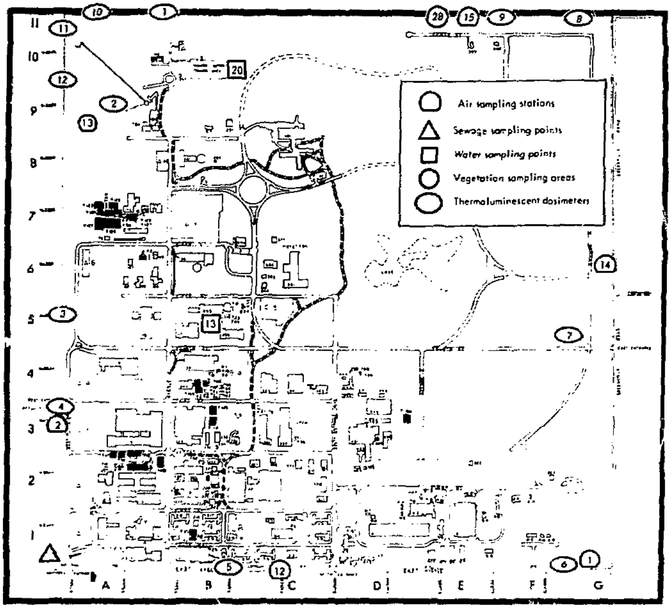

Fig. 2. Lawrence Livermore Laboratory on-site environmental sampling locations.

Storage of Control Dosimeters

Control dosimeters are stored in a 75-mm-thick lead shield equipped with a Cd-Cu "graded" liner. A lead shield of this thickness constitutes an acceptable storage area since the radiation inside is primarily due to the hard component of cosmic radiation plus the contribution of any contaminants in the shield material. The internal exposure rate is therefore relatively low and constant. The shield is shown in Figure 4.

Calibration

A portion of the control dosimeters is removed midway through the field exposure cycle. Using a ^{60}Co source, the individual dosimeters are irradiated at doses from 10 mrad to 100 mrad. Directly following this irradiation procedure the dosimeters are returned to the shield and are read along with the field dosimeters at the end of the exposure cycle.

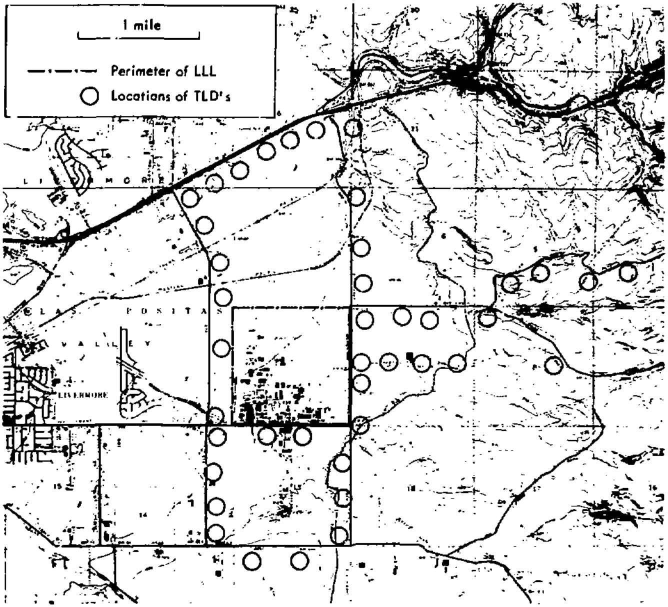

Fig. 3. Location of thermoluminescent dosimeters in the vicinity of the Lawrence Livermore Laboratory.

Dosimeter Readout

After collection from the field, the exposed dosimeters are placed in the lead shield for 48 hrs to permit decay of the low temperature traps. The environmental package is then taken apart and the CaF_2 and LiF holders are loaded into the automatic hot gas reader in separate runs. Calibration phosphors are interspersed within each group. The operating voltage on the photomultiplier tube is increased over that used in personnel dosimetry to provide the increased sensitivity required in measuring environmental background radiation. The reader shown in Figure 5 is then started, and in sequential steps the dosimeter numbers are decoded, the plastic caps are removed, the chips are transferred to the counting chamber and heated, and the light output is recorded. An IBM card is simultaneously prepared identifying each phosphor, its field location, and its light output. These cards are then used in a computer program, which computes mrem doses and confidence limits for these doses.

Fig. 4. Lead shield for control dosimeters.

Results and Discussion

Figure 6 represents distribution plots of annual LLL perimeter and off-site dose rates obtained from TLD data during 1974.(7) The plots show median doses of 69 and 66 mrem for the perimeter and off-site locations, respectively. These are comparable to the doses observed during 1973.(8) An elevated perimeter dose rate (137 mrem) was due to operation of a 14 MeV neutron generator. A single dosimeter that

Fig. 5. Automated TLD reader.

recorded the highest dose (210–215 mrem) is located near an off-site industrial plant where radiography with a ^{192}Ir source is frequently performed.

Thermoluminescence dosimetry measurements do not distinguish between sources of radiation. As we are primarily concerned with the impact of man-made radiation, it is often necessary to be able to subtract the contribution due to the natural radiation background. Terrestrial radiation from naturally occurring uranium, thorium, and potassium in the soil plus cosmic radiation are the principal sources of natural radiation. The terrestrial exposure rate can be approximated from the specific activities of naturally occurring gamma emitters in the soil,[9] and the cosmic radiation can be approximated from the elevation and geomagnetic latitude of the area using the techniques of Lowder and Beck.[illegible] Gamma spectral measurements were made at 37 of the off-site locations using the LLL developed field Ge(Li) spectrometer.[11] Terrestrial exposure rates from uranium, thorium, and potassium calculated from these spectra showed a range of 3.0 to 6.8 μR/hr, with a median of 5.0 μR/hr.[12] The inferred local exposure rate from cosmic rays is 3.8 μR/hr, based on an average site elevation of [illegible] m. These estimates then indicate that the natural ambient gamma radiation background in the vicinity of the Laboratory varies from 6.8 to 10.6 μR/hr with a median of 8.8 μR/hr. This median corresponds to an annual dose of 62 mrem in good agreement with the TLD data. High pressure ion chamber measurements are periodically made at Laboratory perimeter monitoring locations, and the agreement between these measurements and those using TLD is generally good. The advantage of the ion chamber is that it provides real time data, which is useful in investigating possible transitory fluctuations in background radiation. However, our reporting requirements imply data reflecting annual averages. Consequently, integrating devices, such as TLD, are perhaps better suited for routine environmental monitoring.

Most users of CaF_2 incorporate energy filters in their dosimeter package to flatten the phosphor's enhanced response below 100 keV. At LLL, we have elected to utilize this characteristic of CaF_2 to detect the presence of low energy photons. To do this our dosimeters also contain LiF whose response is comparatively energy independent. An increase in the CaF_2/LiF response is then an indication of low energy

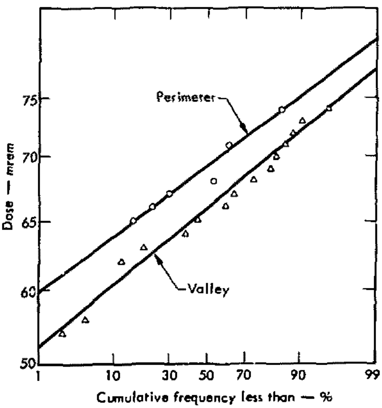

Fig. 6. Laboratory perimeter and Livermore Valley environmental radiation dose rates – 1974.

radiation. In addition, since LiF is approximately tissue equivalent, dividing the CaF_2 mrad values by the CaF_2/LiF ratio converts those doses to mrem. This ratio, which averages 1.3 is obtained by averaging the da . from the off-site dosimeters. Because this ratio is based on a large number of measurements each exposure period, the dose values so derived are usually more precise than individual LiF values. Similarly this manipulation permits us to obtain mrem data with better statistics due to the greater sensitivity of CaF_2.

Appreciable signal fading of CaF_2:Dy — approximately 30% over a period of three months — is a problem that is not encountered with LiF. Rather than apply fading corrections, we have compensated for fading by irradiating the calibration dosimeters midway through the field exposure cycle. This method assumes a linear fading and a constant radiation field during the exposure period.

When the monitoring program began, the 75-mm-thick lead storage shield was located within a laboratory maintained at approximately constant temperature (23°C). During the fourth quarter of 1972 an anomalously high response was observed in all field CaF_2:Dy dosimeters. This increased response was attributed to reduced signal fading of the field dosimeters at the unseasonably low temperatures during that quarter. In order that the control dosimeters might more closely duplicate the temperature cycling of the field dosimeters, the shield was moved out-of-doors. The shield in its present location is shown in Figure 4.

To determine the effect of seasonal variations on dosimeter response in the storage shield, a freshly annealed dosimeter is placed in the shield each week of the field exposure cycle. These dosimeters are read out along with the others at the end of the exposure cycle. An internal dose rate is then calculated from the calibration and the data from these weekly dosimeters.

In the siting of our thermoluminescent dosimeters, the height above ground is maintained at approximately 1 metre at the perimeter locations. As the vicinity of the Laboratory is essentially rural, dosimeters are secured to roadside fencing, variations in which make it difficult to maintain the same height as at the perimeter locations. Concern over these differences in height, which varied about ± 0.30 metres, prompted a test exposure consisting of dosimeters suspended at heights from 25 to 305 centimetres. Results of this three month test, which are shown in Table 1, confirm that dosimeter height is not a critical parameter.

Table 1. Effect of Dosimeter Height

Height, cm	CaF_2 mrad/90 days	mrem/90 days
25	17.9	13.5
45	19.0	14.3
65	19.7	14.8
[illegible]	17.9	13.5
105	18.9	14.2
145	19.2	14.4
165	18.8	14.1
185	18.2	13.7
205	19.5	14.7
225	18.7	14.1
245	20.4	15.3
265	19.7	14.8
285	20.1	15.1
305	21.2	15.9
Av	19.2	14.5

In addition to detecting non-environmental doses at a particular dosimeter site, groups of dosimeter sites may be compared to see if the means of those groups are statistically different. Considering the dose to TLDs in a group to be lognormally distributed and applying a priori tests as described by Toy and Lindeken[13] and Sokal and Rohlf,[14] we have determined that our program is more than adequate to detect a difference of 5 mrem per year between the mean at our perimeter and the mean in the Livermore Valley.

Summary

Although supplemented by other techniques, thermoluminescence is the principal means of measuring ambient gamma radiation at the Lawrence Livermore Laboratory. The procedures employed for environmental monitoring with TLD are designed to take advantage of the automated processing capabilities developed for the Laboratory's personnel dosimetry program. The environmental dosimeter contains both CaF_2:Dy (TLD 200) and ^{7}LiF (TLD 700). The CaF_2/LiF mrad response ratio is used to detect low energy photons in the radiation field, as well as convert the more sensitive CaF_2 data to equivalent mrems. Using dosimeters at 12 locations on the site perimeter and 41 locations in the off-site vicinity, provides adequate sampling to detect a difference of 5 mrem between these two populations.

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