ENVIRONMENTAL MONITORING WITH THERMOLUMINESCENT DOSIMETERS

An Evaluation of the System and a Comparison to Photographic Methods

by
Chas. K. Fitzsimmons and William H. Horn

Southwestern Radiological Health Laboratory

Department of Health, Education, and Welfare
Public Health Service
Consumer Protection and Environmental Health Service

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This study performed under a Memorandum of
Understanding (No. SF 54 373)
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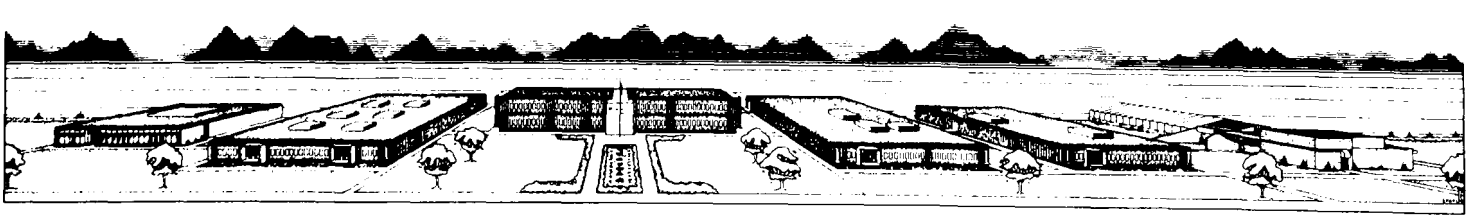
U. S. ATOMIC ENERGY COMMISSION



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ABSTRACT

In August 1965, the Southwestern Radiological Health Laboratory put into operation a thermoluminescent dosimeter (TLD) system for monitoring gamma radiation exposure in the environment surrounding the Nevada Test Site. A description of the equipment and calibration technique is given. Precision of TLD results at the 95% confidence level has been found to be ±3.5%. Accuracy is on the order of ±5%. Actual field data ranging from background levels to about 1 R show TLD's to be more satisfactory than film badges for the particular applications described.

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I. INTRODUCTION

In August 1965, the Southwestern Radiological Health Laboratory (SWRHL) put into operation a thermoluminescent dosimeter (TLD) system for monitoring gamma radiation exposure in the off-site environment. It was soon apparent that a large amount of experimental work was necessary to fully realize the inherent potential of the system. During the first year several questions were answered and techniques developed to bring the system to a full-scale routine monitoring program. The following projects have been completed to a point where the results can be used routinely.

1. Perfection of a reader calibration technique.
2. Establishment of an individual correction factor for each dosimeter.
3. Determination of the internal background due to ^{40}K within the dosimeter.
4. Development of a proper dedosing procedure.
5. Determination of the precision and reliability of the data.

Two other projects are still underway, that of determining the inherent fading rate of the phosphor, and comparing the TLD to the film badge.

II. DESCRIPTION OF EQUIPMENT

The dosimeter in use is a CaF_2:Mn thermoluminescent device, Model TL-12 manufactured by Edgerton, Germeshausen & Grier, Inc. (EG&G). The TL-12 consists of two component parts, a detector (Model TL-32) and an energy compensating shield (Model TL-52). The detector consists of a layer of thermoluminescent CaF_2:Mn bonded to a helical heater element that is contained in an evacuated glass envelope. The aluminum-lead-tin shield that houses the detector is designed to compensate for the detector over-response in the low energy region of the gamma ray spectrum. The energy response is shown in Figure 1, both with and without the shield.

Operation of the dosimeter is based on the thermoluminescent properties of manganese-activated calcium fluoride [1]. When the CaF_2:Mn is exposed to ionizing radiation, valence electrons are raised to higher metastable energy levels, often referred to as "electron traps." This stored energy is released in the form of light (green-orange for Mn^{+} [2]) when the CaF_2:Mn is heated. The released energy, proportional to the cumulative dose received, is converted into an electrical signal by a photomultiplier tube and recorded on a strip chart recorder within the reader. After read-out the dosimeter may be reused. At this writing, 1500 TLD's have been calibrated and are available for use in the monitoring program.

The thermoluminescent dosimeter reader, Model TL-2B, or the newer Model Tl-3B, is an integrated system designed to read-out the TLD's in both standard and micro sizes. The reader accepts the TLD in a light tight chamber, heats it with a regulated current, and converts

the emitted light energy into an electrical signal for display on the built-in strip-chart recorder. Two "read-head" adapters are provided for inserting the different size dosimeters into the reader. One adapter (TL-81) accommodates the vacuum tube-type detector which is used in the routine monitoring program, and the other (TL-81A) the needle micro-thermoluminescent dosimeters.

The reader includes a recording photometer with power supplies and control logic needed to read out the dosimeters. The control logic sequences the recorder operation, dosimeter heating cycle, automatic ranging circuit, and status indicator to effect a chart record of the dosimeter's light emission. The automatic ranging circuit operates when the recorder pen reaches full scale and changes reader sensitivity in decade steps over a total of six decades.

Dosimeter response to exposure is linear over the range from 5mR to 5 kR. The chart can be read to three significant figures by estimating the last digit for values greater than ten on the printed scale.

III. CALIBRATION PROCEDURES

A procedure for insuring the calibration of the Models TL-2B and TL-3B reading instruments has been perfected. Twelve reference dosimeters, type TL-12 devices which have an accurately established correction factor near 1.00, are used as "primary" standards. Two phosphorescent reference lights, one activated by ^{14}C, the other by ^{63}Ni, are used as "secondary" standards.

To perform a calibration, the reference dosimeters are exposed to a known amount of ^{137}Cs gamma radiation. The exposure is measured by taking the average reading of three or four condenser ionization chambers (Victoreen or Landverk R-Meters). The recorder pen is adjusted for zero and reference light values. One of the reference dosimeters is read-out and the chart reading multiplied by the particular correction factor. If the result is not the same as the known exposure, the difference is noted in percent and the reference light values are readjusted correspondingly. The process is repeated with each of the other reference dosimeters until the correct read-out is obtained within 2% of the known exposure. The final reference light value is then noted and the reader is ready for use. Readjustment of the gain by means of the reference light may be required when reading a large group of dosimeters, or if there is a time lapse between batches of dosimeters. In order to assure electronic stability the reader is left on at all times, thus no warm up period is required. It is recommended, however, to read-out two or three dosimeters from the shelf before starting a batch.

Once the reader has been calibrated, correction factors for all the dosimeters can be determined. The dosimeters are exposed to 100 and 200 mR determined by the ion chambers, with a 0.100 curie ^{137}Cs source. Up to 200 dosimeters can be exposed at one time on a special circular calibration table. Source-dosimeter distances available are 50 cm and 100 cm. Each dosimeter is exposed three times. The read-out is performed within 24 hours after each exposure. If a reading appears doubtful, the procedure is repeated until three read-outs are obtained within the reproducibility claimed by the manufacturer ($\pm$ 1%). If this is not possible within a reasonable number of attempts, the dosimeter is rejected.

The ion chamber value divided by the mean of the three dosimeter readings is taken as the correction factor. Periodically, correction factors are rechecked to assure dosimeter stability and retention of sensitivity. It has been found that it is best to calibrate all dosimeters even though the manufacturer provides this service. In view of this, uncalibrated dosimeters are usually purchased at a reduced price and calibrated as above. Correction factors range from 0.79 to 1.26.

An inherent property of the TL-12 dosimeter is its "internal background" caused by the presence of ^{40}K in the material (potassium silicate) which bonds the CaF_2:Mn to the heating element. In order to determine the magnitude of the internal background, all other sources of exposure had to be accounted for. A large number of dosimeters were exposed for varying time periods in locations of known background. In one experiment a gun-barrel counting shield was used for which the background was well established at 0.12 mR/day with an ion chamber. A value of 0.7 $\pm$ 10% mR/day was calculated for the internal background exposure rate. Subsequent tests by this laboratory have confirmed this figure and have shown it to be a constant for all the dosimeters.

That thermoluminescent phosphors retain the absorbed energy information for long periods of time is not to say that some spontaneous light emission does not occur. Electron traps of several energy levels are involved and each has its own probability of decay to the ground state. "Shallow" traps decay more rapidly as a rule than deeper ones. Any loss of information by this process is called fading. Our studies have shown that a given dosimeter exposed to 400 mR may fade 5% - 10% over a 30-day period. The bulk of the decay occurs in the first 5 or 6 days and then levels off for the duration of the period. Figure 2 shows the percentage loss of luminescence with time as determined in one experiment. The data points are mean values with $\pm$ one sigma shown by the vertical lines. The degree of fading is not a simple function of time, and thus has not been included as a correction factor in the routine monitoring program. All such plots exhibited a recovery after 5 - 10 days before continuing to decay. Fading may be a function of dose rate as well as time.[2] More experimentation with the TL-12 is necessary to incorporate a correction for fading into the calculations.

The process of reading out the dosimeter virtually erases the dose information and supposedly anneals the CaF_2 for reuse. It has been found, however, that some residual activation remains which can increase readings taken on subsequent usage. Total dedosing is achieved by placing the TL-32 (without shield) in a 350°C oven for at least 12 minutes. Dedosing is accomplished as close to issue time as possible to reduce background.

IV. PRECISION

In the process of determining the fading rate, standard deviations were calculated for each group of dosimeters. The means were assumed to be normally distributed. Thirty-six groups of eleven dosimeters each were exposed to 400 mR. An overall estimate of the variance yielded a coefficient of variation of 1.8%. Thus, at the 95% confidence level, the true mean is expected to be within $\pm$ 3.5% of a given single dosimeter reading.

For comparison it should be noted that controlled exposures of Du Pont 508 film dosimeters yielded a dispersion of values nearly twice that of TLD's. For film, the standard deviation was least at exposures near 500 mR and increased for both lower and higher exposures.[3] A more recent comparison of TLD and film data collected from September 1966 to July 1967 shows an even greater difference between the two types of dosimeters than the controlled experiments (Table 1). Four locations around the perimeter of the Nuclear Engineering radioactive waste disposal site near Beatty, Nevada, were equipped with three TLD's (Type TL-12) and five film badges (Du Pont Type 545). The only unique aspect of these locations compared to the other film-TLD locations in the network is the relatively high monthly exposure (50-1000 mR). As a result, the film badges as well as the TLD's register positive readings and the two can be compared directly. Type 545 film has a single component emulsion and is inclosed by a reusable plastic holder. A lead strip contained in the holder is folded around the film packet to reduce energy dependence. The badge is designed to measure low exposures of the relatively high gamma energies of fission products.

Analyses of variance were performed on the data. For each month and location, the mean of the three TLD or five film badge values was calculated. The differences between the values and their respective means were then calculated as percentages. These calculations normalized the data by removing the effects of monthly and geographical variations. Assuming a normal distribution of percentages, the variance estimates were calculated for the two types of dosimeters. It was found that the dispersion of values about the mean film badge reading was nearly five times that of the TLD's. The standard error for film badges was 20.2%, and for TLD's, 4.2% of the respective means.

Using the same data in a randomized block design (blocked by months), the hypothesis that the mean film badge reading and the mean TLD reading for a given month and location are equal was tested. At the 95% confidence level no significant difference in the means was detectable. There is reason to believe there is some difference in response between the two types of dosimeters, however, especially at low exposures. Further work is being done in pursuit of this problem. For comparison purposes the accuracy (calibration) of both the film badge and the TLD is assumed to be without error. In actuality, the accuracy of the TLD calibration is about $\pm$ 5 to 10% while that of the film is claimed to be $\pm$ 10 to 20%. The range of the film is limited to .05 to 3.0 R.

V. RELIABILITY

By the nature of their construction, the TL-12 dosimeters are quite insensitive to environmental hazards such as heat, light, water, age, handling, and processing. A minimal number of anomalies in the data makes the TLD system very reliable. During summer months, heat damage to film badges is the most common cause of data loss. A plot of data recovered as a percent of dosimeters issued each month shows a striking seasonal variation for film data (Figure 3). Occasional loss of TLD data is attributable to either a lost or destroyed dosimeter, or a reader malfunction.

Earlier studies of the effect on environmental heat on film badges[4] showed that serious errors in readings were produced when the temperature during exposure was 20°F greater than that during processing. Storage at high temperatures before exposure increased the base fog and possibly the emulsion sensitivity which produced errors as great as 75%.

Reliability is a must for an effective monitoring system. During the period from June 16 to July 15, 1966, the following data were collected from the dosimeter station at the Nuclear Engineering facility near Beatty, Nevada:

TLD Readings	Film Badge Readings	
1. 1034 mR	1. Light Damage	No Data
2. 995 mR	2. Light Damage	No Data
3. 1040 mR	3. Light Damage	No Data
	4. Light Damage	Possible Exposure of 3-4 R
	5. Light Damage	Possible Exposure of 3-4 R

If data such as the above were returned from an incident involving possible personnel exposure, it could cause obvious problems if only film badges had been issued.

VI. MEASUREMENT OF BACKGROUND BY THERMOLUMINESCENT DOSIMETRY

Figure 5 shows the present off-site coverage maintained by the dosimetry program. In general the dosimetry stations fall in the same area as other PHS off-site surveillance activities. Most routine milk or air sampling locations have TLD's also. Some TLD locations are in uninhabited areas, but are maintained to provide a tighter network of dosimeters around the Nevada Test Site.

The exposure rates of a number of selected stations are represented in Figure 4. The locations are listed approximately in a counterclockwise order around the test site starting from Las Vegas.

Exposure rates in mR/day were calculated by dividing the net monthly exposures by the number of days the dosimeters were in the field. No known releases of activity occurred during this period so the values obtained are assumed to represent natural backgrounds.

Table 2 lists the yearly mean, the range of monthly values, and the standard error (coefficient of variation) for each of the seventeen locations represented in Figure 4. If the fluctuation in background can be assumed to be random, then some predictions based on statistical tests should be possible. For example, once the dispersion of values around a mean background value at a particular location is established, an upper limit to the expected background exposure for any month can be set. Values above the expected value would be suspected of unnatural origin. The average coefficient of variation for the seventeen stations was 59% (among location monthly means, each month having three replications). An arbitrary limit of 2 or 3 standard

errors above the mean would establish the minimum detectable signal above normal background. The major problem with the statistical approach is how to decide which values are to be included in the background average and which are to be excluded. Deleting marginal values, e.g., three sigma above the current average, might bias the estimate of the true mean.

VII. CONCLUSIONS

The TLD system described above has proved to be well suited to environmental monitoring for gamma radiation. Approximately one year was spent in preparation for the full scale program which promises to provide some very interesting data. Probably some of the best estimates of natural background exposure rates and their fluctuations around the Nevada Test Site have been obtained with TLD's. The TLD's seem to be quite insensitive to environmental hazards, especially heating, which is the greatest hazard to the film badge during the summer months.

The precision, as well as the accuracy, of TLD's is better than that which normally is attained with film badges. By virtue of their reusability, calibration of TLD's does not depend on a representative sample of a batch of dosimeters, but rather on a constantly updated correction factor for each individual dosimeter.

The capability of the dosimeter to measure background levels of exposure has increased the sensitivity of the dosimetry program by a factor of ten.

Table 1. Comparison of film badge (FB) and TLD data from Nuclear Engineering Company disposal site.

Date Issued	Date Collected	Dosimeter	Exposures					Avg. Exp. (mR)
			North Fence					
09/29/66	11/02/66	FB	120	145	125	125	145	132
		TLD	156	156	138			150
11/02/66	11/30/66	FB	0	0	30	0	0	30.0
		TLD	35	37	33			36.6
11/30/66	01/04/67	FB	50	55	45	50	45	49.0
		TLD	35	37	35			35.7
01/04/67	02/02/67	FB	0	30	20	20	0	23.3
		TLD	45	42	43			43.3
02/02/67	03/09/67	FB	120	35	0	125	35	78.8
		TLD	36	37	38			37.0
03/09/67	04/06/67	FB	60	55	60	60	55	58.0
		TLD	58	66	64			62.7
04/06/67	05/03/67	FB	50	60	60	55	55	56.0
		TLD	61	64	62			62.3
05/03/67	06/02/67	FB	85	100	100	90	115	98.0
		TLD	91	95	92			92.7
06/01/67	07/20/67	FB	85	75	90	75	80	81.0
		TLD	51	52	53			52.0
			South Fence					
01/04/67	02/02/67	FB	60	45	30	30	35	40.0
		TLD	64	64	64			64.0
02/02/67	03/09/67	FB	35	160	145	65	160	113
		TLD	70	72	71			71.0

Table 1. Comparison of film badge (FB) and TLD data from Nuclear Engineering Company disposal site. (continued)

Date Issued	Date Collected	Dosimeter	Exposures					Avg. Exp. (mR)
			South Fence (continued)					
03/09/67	04/06/67	FB	295	285	300	235	220	267
		TLD	270	266	272			269
04/06/67	05/03/67	FB	70	60	70	65	60	65.0
		TLD	72	69	76			72.3
05/03/67	06/02/67	FB	105	90	105	115	105	104
		TLD	89	108	98			98.3
06/01/67	07/20/67	FB	0	0	95	90	105	96.7
		TLD	64	67	61			64.0
			East Fence					
09/29/66	11/02/66	FB	125	135	130	120	135	129
		TLD	134	128	125			129
11/02/66	11/30/66	FB	115	115	110	135	125	120
		TLD	178	177	185			180
11/30/66	01/04/67	FB	85	90	90	90	90	89.0
		TLD	102	98	104			101
01/04/67	02/02/67	FB	30	35	35	35	0	33.8
		TLD	63	65	74			67.3
02/02/67	03/09/67	FB	45	45	0	145	45	70.0
		TLD	60	63	60			61.0
03/09/67	04/06/67	FB	140	305	145	170	150	182
		TLD	150	154	145			150
04/06/67	05/03/67	FB	80	95	105	95	105	96.0
		TLD	75	76	73			74.7

Table 1. Comparison of film badge (FB) and TLD data from Nuclear Engineering Company disposal site. (continued)

Date Issued	Date Collected	Dosimeter	Exposures					Avg. Exp. (mR)
			East Fence (continued)					
05/03/67	06/02/67	FB	100	110	105	110	105	106
		TLD	98	102	94			98.0
06/01/67	07/20/67	FB	0	0	180	0	0	180
		TLD	148	158	146			151
			West Fence					
09/29/66	11/02/66	FB	295	250	260	225	255	258
		TLD	274	295	292			287
11/02/66	11/30/66	FB	560	555	560	560	560	559
		TLD	563	513	541			539
11/30/66	01/04/67	FB	70	75	75	75	85	76.0
		TLD	67	69	61			65.7
01/04/67	02/02/67	FB	140	145	145	120	155	141
		TLD	182	183	190			185
02/02/67	03/09/67	FB	280	260	380	395	290	321
		TLD	294	303	293			297
03/09/67	04/06/67	FB	125	130	125	125	110	123
		TLD	129	108	127			121
04/06/67	05/03/67	FB	780	665	575	600	695	663
		TLD	636	776	626			679
05/03/67	06/02/67	FB	420	530	450	480	645	505
		TLD	577	563	514			551
06/01/67	07/20/67	FB	90	115	125	110	110	110
		TLD	78	75	83			78.7

Note: Zeros indicate nondetectable exposure or no data due to damage. Zero values are not averaged.

Table 2. Average background exposure rates (mR/day) for the period October 1966 to September 1967.

Location	Yearly Mean	Range of Monthly Means	Percent Coefficient of Variation
Las Vegas, Nevada	0.272	0.175-0.619	85
Warm Springs Ranch, Nev.	0.247	0.143-0.490	67
St. George, Utah	0.280	0.152-0.483	54
Pioche, Nevada	0.347	0.206-0.542	43
Alamo, Nevada	0.387	0.243-0.596	45
Hancock Summit, Nevada	0.549	0.357-0.790	37
Garrison, Utah	0.378	0.167-0.780	79
Duckwater, Nevada	0.415	0.184-0.701	58
Warm Springs, Nevada	1.092	0.285-1.529	70
Clark's Station, Nevada	0.478	0.254-0.770	51
Austin, Nevada	0.412	0.222-0.762	73
Tonopah, Nevada	0.419	0.256-0.632	75
Groom Lake, Nevada	0.362	0.194-0.541	50
Lathrop Wells, Nevada	0.406	0.219-0.575	40
Shoshone, California	0.278	0.120-0.417	61
Mammoth Lake, California	0.375	0.198-0.529	80
Barstow, California	0.379	0.262-0.472	34

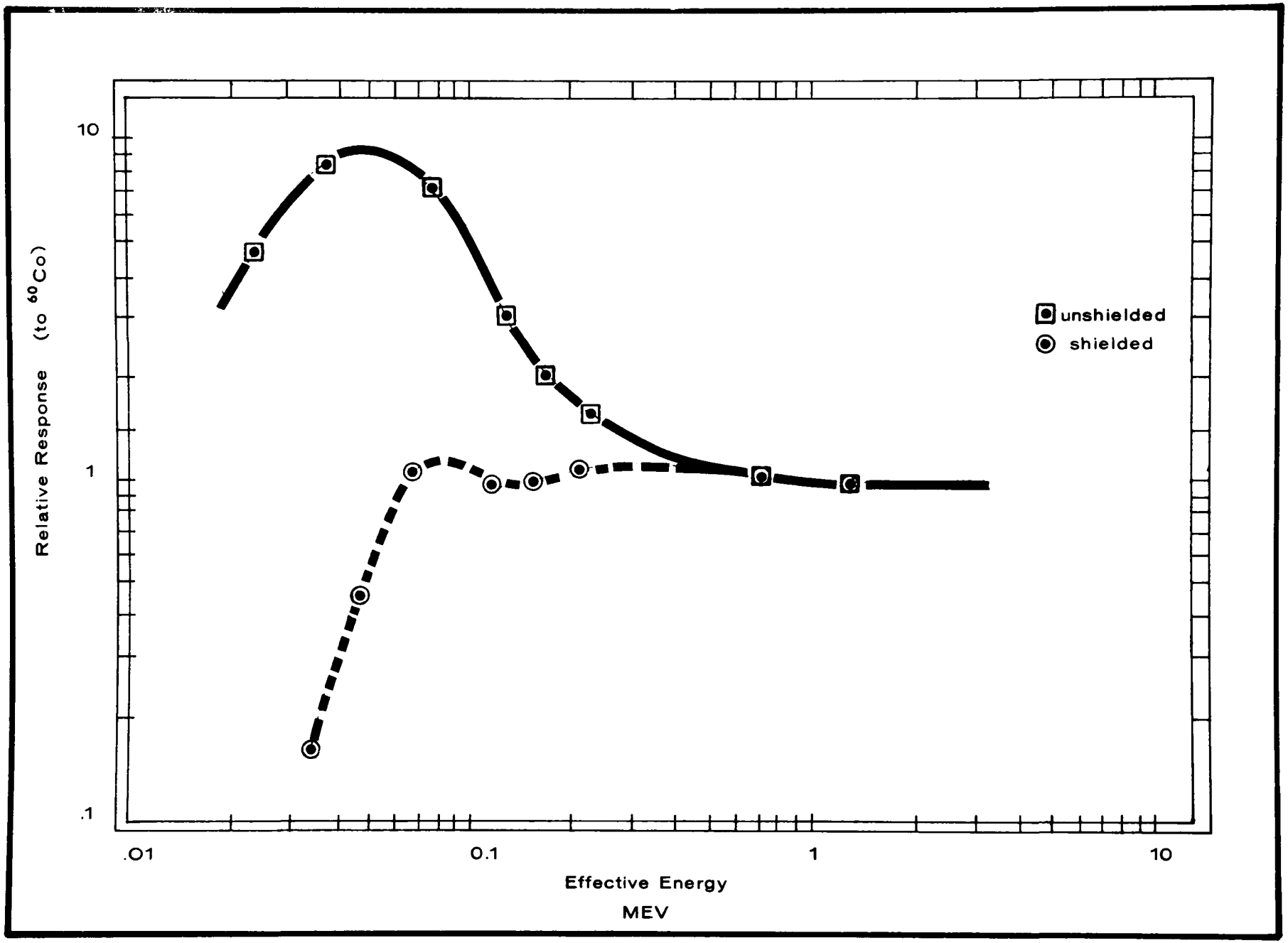

Figure 1. Energy response of thermoluminescent CaF_2:Mn.
(Data provided by manufacturer)

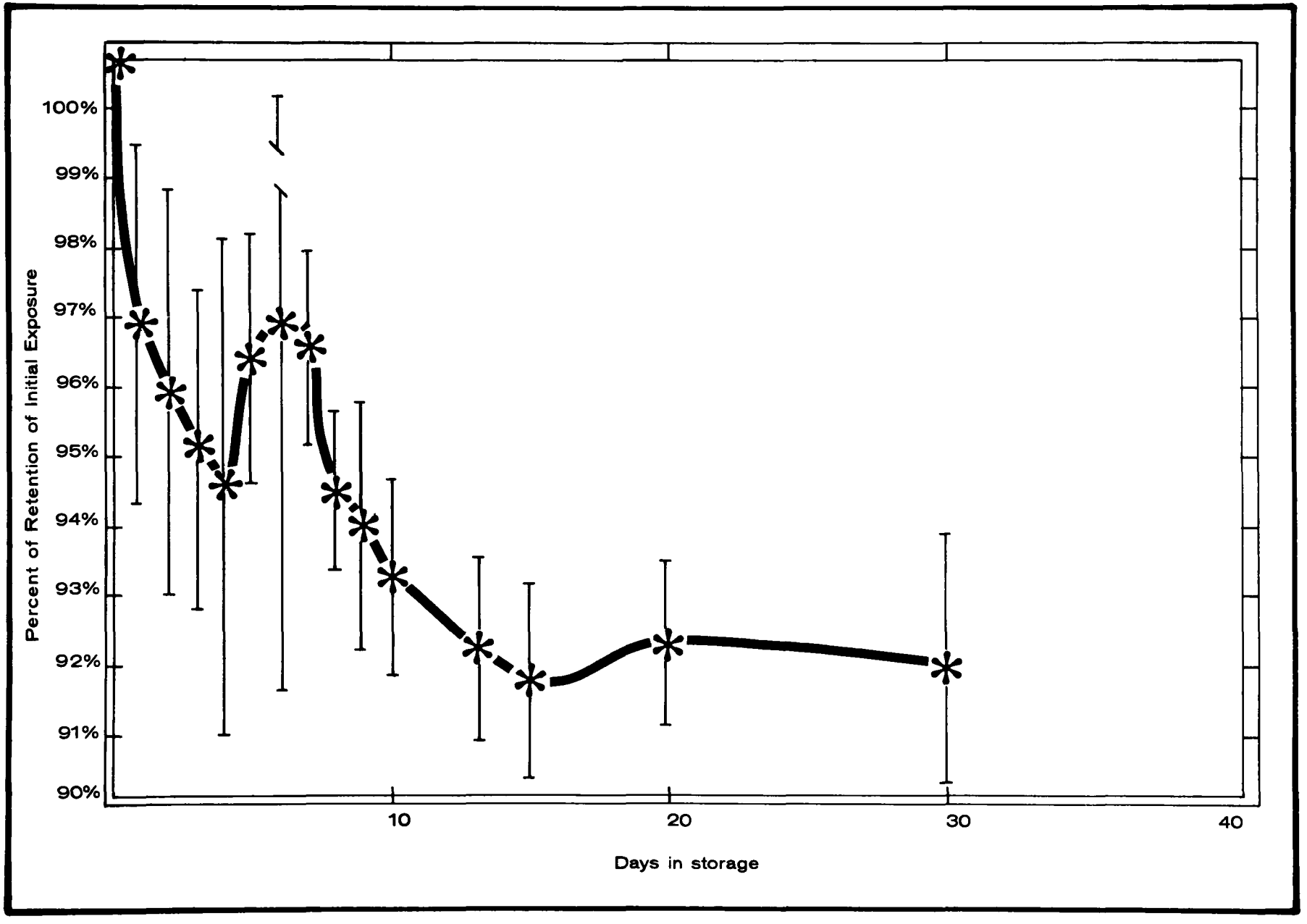

Figure 2. Fading of CaF_2:Mn.

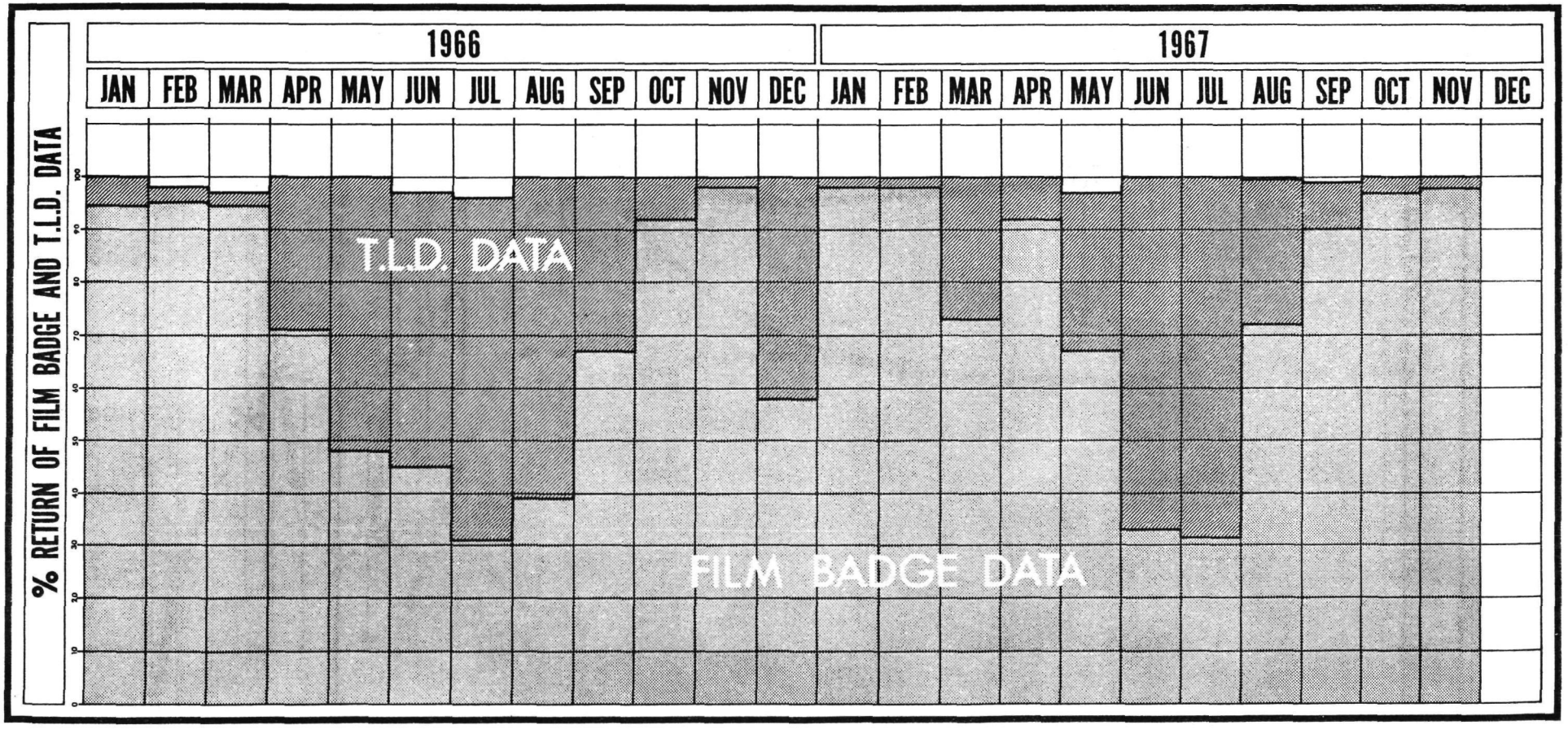

Figure 3. Percent return of film badge and TLD data during 1966 and 1967.

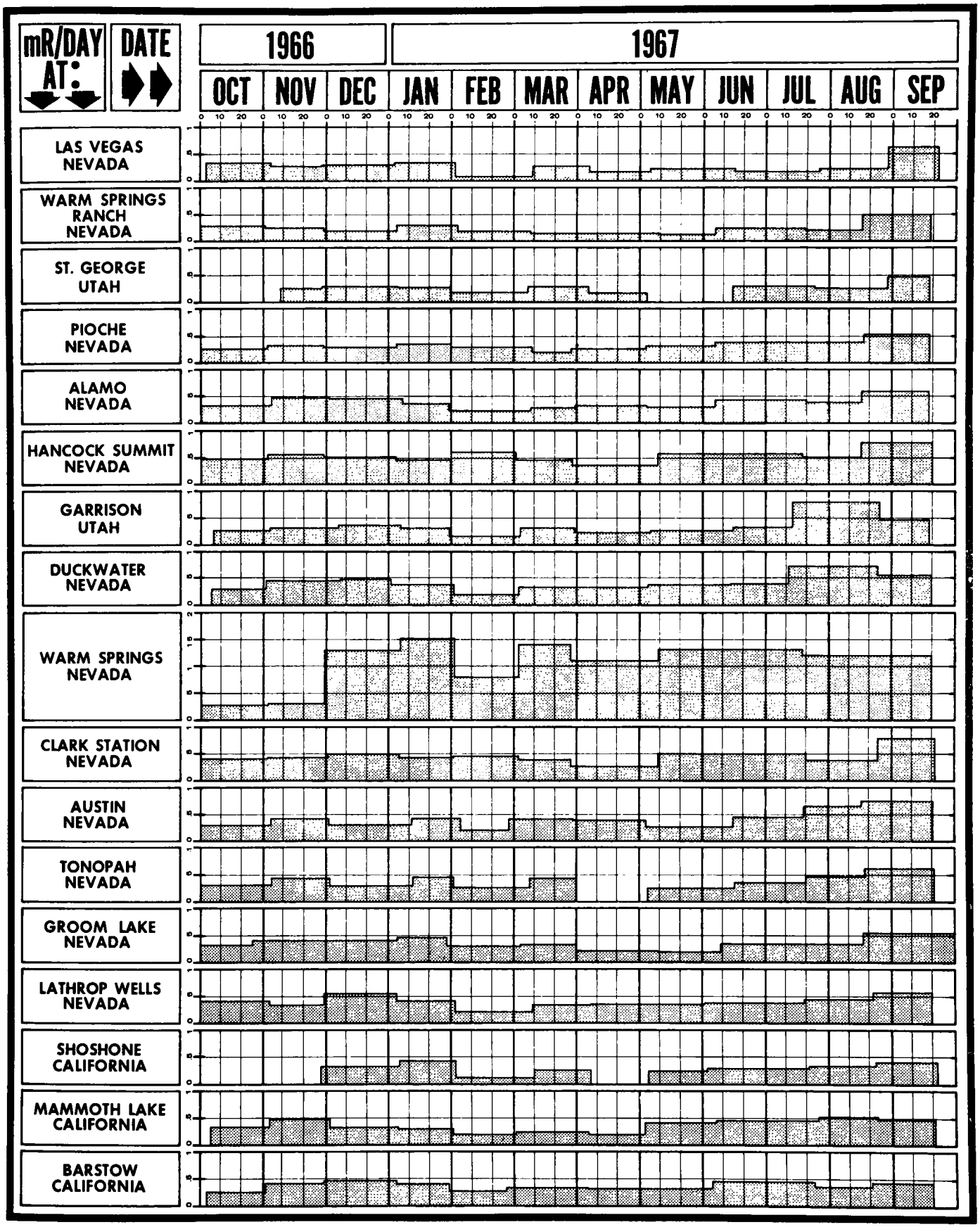

Figure 4. Background exposure rates at selected locations around the Nevada Test Site as determined by TLD's.

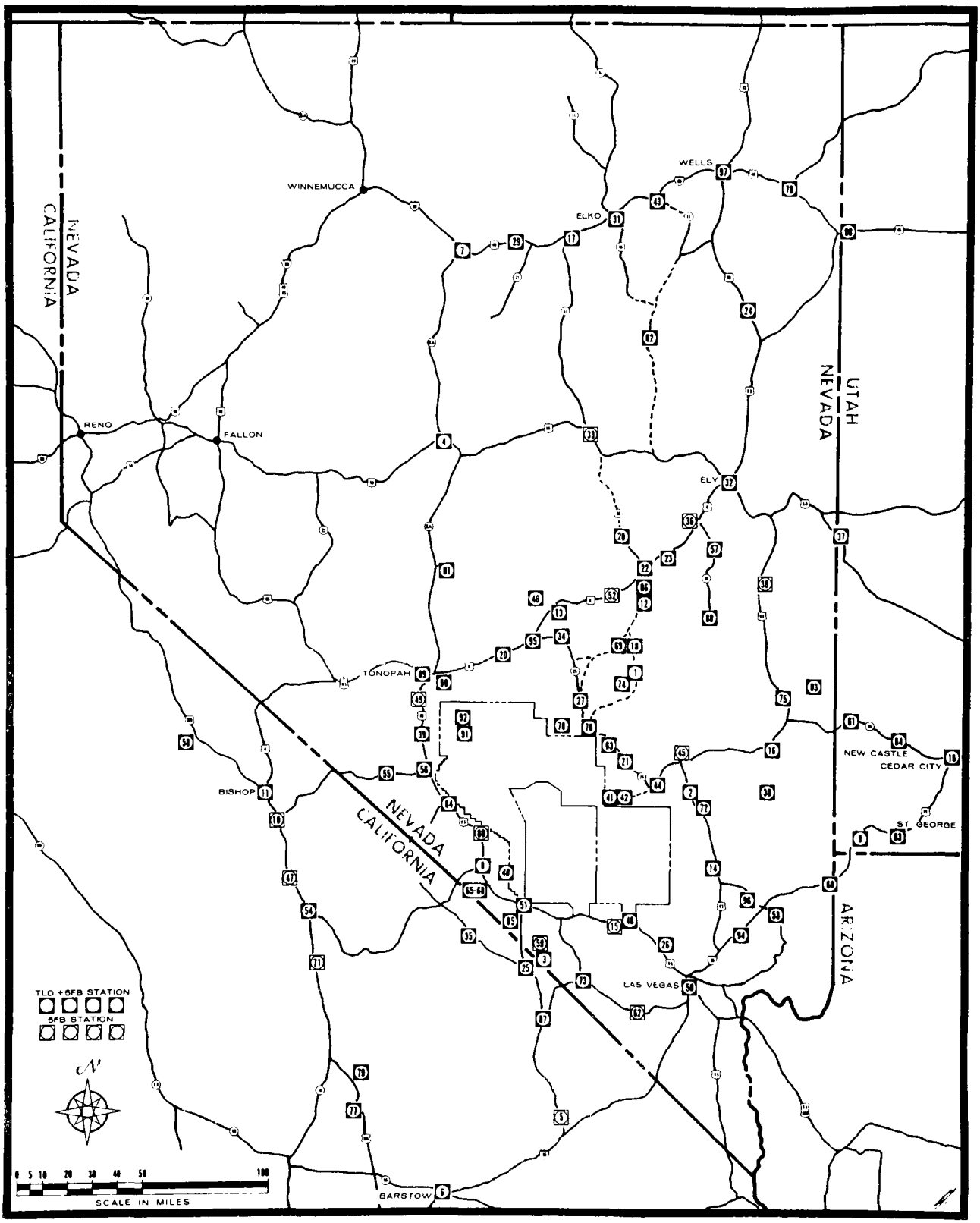

Figure 5. Film badge and TLD Stations, Environmental Surveillance Program.

Table 3. Film badge and TLD stations.

1.	Adaven, Nevada	30.	Elgin, Nevada
2.	Alamo, Nevada	31.	Elko, Nevada
3.	Ash Meadows, Nevada	(32)	Ely, Nevada (Airport)
4.	Austin, Nevada	(33)	Eureka Maint. Sta., Nev.
(5)	Baker, California	34.	Fallini's Ranch, Nevada
6.	Barstow, California	35.	Furnace Creek, California
7.	Battle Mountain, Nevada	(36)	Gardner Ranch, White River Valley, Nevada
8.	Beatty, Nevada		
9.	Beaver Dam Summit, Utah	37.	Garrison, Utah
(10)	Big Pine, California	(38)	Geyser, Nevada
11.	Bishop, California	39.	Goldfield, Nevada
12.	Blue Eagle Ranch, Nevada	40.	Goldspar Mine, Nevada
13.	Blue Jay, Nevada	41.	Groom Lake, Station A, Nev.
14.	Butler Ranch, Nevada	42.	Groom Lake, Station B, Nev.
(15)	Cactus Springs, Nevada	43.	Halleck, Nevada
16.	Caliente, Nevada	44.	Hancock Summit, Nevada
17.	Carlin, Nevada	(45)	Hiko, Nevada
18.	Casey's Ranch, Nevada	46.	Hot Creek Ranch, Nevada
19.	Cedar City, Utah	(47)	Independence, California
20.	Clark Station, Nevada	48.	Indian Springs, Nevada
21.	Coyote Summit, Nevada	(49)	Klondike, Nevada
22.	Currant, Nevada	50.	Las Vegas, Nevada
23.	Currant Maint. Sta., Nevada	51.	Lathrop Wells, Nevada
24.	Currie, Nevada	(52)	Lockes Ranch, Nevada
25.	Death Valley Junction, Cal.	53.	Logandale, Nevada
26.	Desert Game Range, Nev.	54.	Lone Pine, California
27.	Diablo, Nevada	55.	Lida, Nevada
28.	Duckwater, Nevada	56.	Lida Junction, Nevada
29.	Dunphy, Nevada	57.	Lund, Nevada

Table 3. Film badge and TLD stations. (continued)

58. Mammoth Lake, California
(59) McGillivary Ranch, Nevada
60. Mesquite, Nevada
61. Modena, Utah
(62) Mt. Springs Summit, Nevada
63. Nevada Farms, Nevada
64. New Castle, Utah
65. Nuclear Engineering Co. (North), Nevada
66. Nuclear Engineering Co. (South), Nevada
67. Nuclear Engineering Co. (East), Nevada
68. Nuclear Engineering Co. (West), Nevada
69. Nyala Ranch, Nevada
70. Oasis, Nevada
(71) Olancha, California
72. Pahranagat Lake, Nevada
73. Pahrump, Nevada
74. Pinecreek, Nevada
75. Pioche, Nevada
76. Queen City Summit, Nevada
(77) Randsburg, California
78. Reed Ranch, Nevada
79. Ridgecrest, California
(80) Road "D" and Hwy. 95, Nevada
81. Round Mountain, Nevada
82. Ruby Valley, Nevada
83. St. George, Utah
84. Scottys Junction, Nevada
85. Selback-Strickland Ranch (Amargosa Desert), Nevada
86. Shell Oil Site, R. R. Valley, Nevada
87. Shoshone, California
88. Sunnyside, Nevada
89. Tonopah, Nevada
(90) Tonopah (Airport), Nevada
91. Tonopah Test Range, CPI, Nevada
92. Tonopah Test Range, Main Gate (Point Able), Nevada
93. Ursine, Nevada
94. Valley of Fire, Nevada
95. Warm Springs, Nevada
96. Warm Springs Ranch, Nevada
97. Wells, Nevada
98. Wendover, Utah

0. = TLD & 5 FB (0) = 5 FB St.

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