RECEIVED BY DTIC 12 27 1971

UCLA - 12 - 8 51

MASTER

UNIVERSITY OF CALIFORNIA, LOS ANGELES

LABORATORY OF NUCLEAR MEDICINE AND RADIATION BIOLOGY

CONTRACT NO. AT (04-1) GEN-12

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Short Title:

SOIL THERMOLUMINESCENCE

Complete Title:

SOIL THERMOLUMINESCENCE IN RELATION TO RADIATION
EXPOSURE UNDER FIELD CONDITIONS

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

Author's name and affiliation:

H. NISHITA AND M. HAMILTON

University of California. Los Angeles¹

Table 1

Ranges in which the minimum radiation exposure level needed to obtain measurable soil thermoluminescence might occur.

Method of Measurement	Hanford s.l. R	Soil No. 4FF R
Peak height at 200°C	20 - 35	45 - 70
Area below 200°C	40 - 75	35 - 85
Total area	20 - 55	--

Footnotes:

- ¹ From the Laboratory of Nuclear Medicine and Radiation Biology, University of California, Los Angeles, California 90024. These studies were supported by Contract AT(04-1) GEN-12 between the Atomic Energy Commission and the University of California.

Figure Legends:

- Fig. 1. Glow curves for Hanford sandy loam and soil No. 4FF before and after exposure to Cs137 photons (Experiment I).
- Fig. 2. Lithium fluoride (TLD-100) dosimeter readings in relation to soil depth for different lengths of irradiation time.
- Fig. 3. Soil thermoluminescence as measured by three methods in relation to depth.
- Fig. 4. Soil thermoluminescence as measured by three methods in relation to Cs137 radiation exposure level (Experiment I)
-- Hanford sandy loam.
- Fig. 5. Soil thermoluminescence as measured by three methods in relation to Cs137 radiation exposure level (Experiment I)
-- Soil No. 4FF.
- Fig. 6. Lithium fluoride (TLD-100) dosimeter readings in relation to distance from the base of the Cs137 source tower.
- Fig. 7. Soil thermoluminescence as measured by three methods in relation to distance from the base of the Cs137 source tower.
- Fig. 8. Soil thermoluminescence as measured by three methods in relation to Cs137 radiation exposure level (Experiment II)
-- Hanford sandy loam.
- Fig. 9. Soil thermoluminescence as measured by three methods in relation to Cs137 radiation exposure level (Experiment II)
-- Soil No. 4FF.

REFERENCES

1. Darnell, T. C. 1967. In vivo dosimetry in the lung for internally administered radioactive isotopes. Ph.D. Thesis, University of California, Los Angeles, California.
2. French, N. R. 1964. Description of a study of ecological effects on a desert area from chronic exposure to low level ionizing radiation. U.S. Atomic Energy Commission Document, UCLA 12-532, Biology and Medicine TID-4500, 34th ed.
3. Lucas, A. C., Burson, Z. G., and Lagerquist, R. E. 1966. Design of a shielded source for the irradiation of natural animal populations. U.S. Atomic Energy Commission Document, CEX-63.10.
4. Lucas, A. C., and French, N. R. 1967. A miniature thermoluminescent dosimeter and its application to radioecology. In F. H. Attix, Editor, "Luminescence Dosimetry," CONF-650637, pp. 402-411.
5. Nishita, H., and Hamilton, M. 1968. Some thermoluminescent characteristics of gamma irradiated soils. Soil Sci. 106: 76-84.
6. Nishita, H., and Hamilton, M. 1969. Heating effects on the thermoluminescence of gamma irradiated soils. Soil Sci. 108: 1-10.
7. Nishita, H., and Hamilton, M. 1970. Spurious thermoluminescence of soils. Soil Sci. 110: 371-378.
8. Nishita, H., and Hamilton, M. 1971. Influence of temperature during the gamma irradiation period on the subsequent thermoluminescence of soils and LiF and CaF_2 dosimeters. Soil Sci. 112: 393-398.

SOIL THERMOLUMINESCENCE IN RELATION TO RADIATION

EXPOSURE UNDER FIELD CONDITIONS

H. Nishita and M. Hamilton

The detection and measurement of ionizing radiation have long been subjects of investigation in the physical and the life sciences. Over the years, various types of radiation dosimeters have been developed. Among these are the thermoluminescent dosimeters, some of which are in use in measuring radiation exposures in natural environments such as radiation fields to study the effect of ionizing radiation on an ecosystem. In such studies, the dosimeters can be placed at selected sites before the area is exposed to the radiation source. The pre-irradiation placement, however, may not have been done always at the appropriate sites or in sufficient number to fully assess the radiation field. Under these circumstances, it may be advantageous if soil or other materials in the environment could be used to estimate the radiation exposure. We have been studying soil thermoluminescence with this in mind (5 - 8). The radiation exposures for the previous studies have been done in the laboratory in which the environmental conditions were quite constant.

The objective of this study was to determine the relationship of soil thermoluminescence to ionizing radiation exposure under the conditions of natural environment where the temperature fluctuates considerably diurnally and seasonally. It has been shown previously that temperature of the soil during exposure to ionizing radiation can affect the subsequent thermoluminescence of the soil (8).

MATERIALS AND METHODS

Two soils (Hanford sandy loam and soil No. 4FF) were used for this study. The chemical characteristics of these soils have been reported previously (5). Hanford s.l. is an agricultural soil collected in California. Soil No. 4FF is a non-saline highly calcareous soil collected at Frenchman Flat (Nevada Test Site).

In experiment I, 5 columns of each soil were buried in the native soil and irradiated at a constant distance (21.2 m) from the radiation source. Two columns, one of each soil type, were removed every 14 days until all were removed. The soil columns were constituted in the following manner. The soil was placed in lucite tubes (3.81 cm. I.D., 0.32 cm. wall) that were formed by stacking twelve 2.54-cm. high rings. The rings were held together in a column by black plastic tape. Each ring containing the soil was separated from each other by a Mylar film (0.025 mm. thickness) to allow easy separation of each increment. An extruded LiF (TLD-100) dosimeter (1 mm. dia. x 6 mm.) contained in a No. 5 gelatin capsule was placed in the center of the soil mass in each ring. An additional LiF dosimeter in a capsule covered with black plastic tape was placed at the surface of the top increment. Since the columns were buried with their top surfaces flush with the surface of the surrounding native soil, the surface dosimeter was not interposed from the radiation source by any soil. The columns were exposed to the radiation source during the period of 28 September to 7 December, 1970. During this period, the mean daily air temperature varied between 2° and 24°C.

In experiment II, the soils were irradiated at 12 different distances from the radiation source for the same period of time. The soil sites were located on a straight line extending away from the base of the source tower in 15-m. increments of distance. During the experimental period (12 February to 4 May, 1971), the mean daily air temperature varied between 3 and 18° C. The soils were placed on the surface of the bare native soil in 15 x 20 cm. polyethylene bags (0.025 mm. wall thickness) in 20-g. aliquots. The soils formed a thin layer in the bags, since their bulk volumes were small in relation to the bag volume. One of the bags at each site contained a calibrated LiF dosimeter in No. 5 gelatin capsule covered with black plastic tape.

These experiments were conducted at the 9 hectare radiation ecology sites at Rock Valley, Nevada Test Site. The radiation source is a 33,600 Ci Cs137, which is differentially shielded with lead. This radiation facility has been described in detail previously (2 - 4). Except when it was lowered into a lead cask below ground to permit the placement and the collection of samples, the source was raised to 15-m. height above the ground surface. During the experimental periods indicated above, the source was up for a total time of 1679 and 1461 hours for experiments I and II, respectively.

The measurements of soil thermoluminescence were done on 30 mg. aliquots of loose sample using the apparatus and the methods described before (7). In order to minimize any variation among samples due to thermoluminescent decay, all readings were done 28 days after irradiation, except the experiment II samples of soil No. 4FF, which were read out 29 days after irradiation. At these times, the thermoluminescent decay

was at a very slow rate. It was shown previously (5) that the most rapid decay of soil thermoluminescence occurs within the first 24 hr. after irradiation.

The LiF dosimeters were read out with a reader purchased from Madison Research and Development Laboratories, Inc. (Model S-2LC). The calibration curves for the LiF rods were obtained by exposing them to various amounts of Cs137 gamma rays in air. As a standard operating procedure, the rods were annealed for one hour at 400°C and then for 24 hours at 70°C before irradiation. A post-irradiation annealing regimen (100°C for 10 min.) was used for both the calibration and the unknown determinations in order to minimize the influence of fading during exposure and storage. Since the readings of the LiF rods exposed inside the soil mass were referred to the calibration curves obtained in the manner described above, the word "exposure" as used in this paper indicates "exposure in roentgens referred to air."

RESULTS AND DISCUSSION

Figure 1 shows the glow curves for Hanford s.l. and soil No. 4FF before and after exposure to Cs137 photons (experiment I). The shapes of natural thermoluminescence glow curves (curves labelled III) of the two soils were different. The natural thermoluminescence of Hanford appeared as one broad peak, while that of soil No. 4FF appeared as two peaks, in the high temperature range ($>200^{\circ}\text{C}$). The threshold temperature for the natural thermoluminescence for Hanford s.l. and soil No. 4FF was about 230° and 175°C , respectively. Our experience in working with many different soils has shown that very little, if any, natural thermoluminescence occurs in the temperature range below 200°C . Note that after

the soils have been exposed to radiation, their glow peaks show growth in the temperature range below 200°C , in which there is only little or no natural thermoluminescence. This characteristic of soil glow curves is utilized to circumvent the complication of the presence of natural thermoluminescence in relating radiation-induced (other than by natural radiation) soil thermoluminescence to exposure level. Figure 1 also shows that most of the Cs137 radiation-induced thermoluminescence in Hanford s.l. occurred in the temperature range below 250°C , while that in soil No. 4FF was vice versa. The effect shown by Hanford s.l. was followed by all other noncalcareous soils that we have examined (7). With highly calcareous soils as soil No. 4FF, the radiation-induced thermoluminescence is lower in the temperature range below 250°C for low radiation exposure levels, but it increases at a greater rate in this temperature range with increasing exposure level. When a sufficiently high exposure level (approximately 2×10^4 R) is attained, the radiation-induced thermoluminescence below 250°C becomes greater than that above 250°C (5). Thus, working in the lower temperature range ($< 250^{\circ}\text{C}$) has the advantage of obtaining greater sensitivity to increase of radiation dosage as well as affording the possibility of avoiding the presence of natural thermoluminescence.

The exposure readings of LiF dosimeters that were imbedded in the soil columns are shown in figure 2. The exposure readings at the surface of the columns in the order of the irradiation termination time were 208, 276, 324, 500 and 548 R. Due to energy attenuation of the gamma rays, the exposure values decreased progressively from the surface to the bottom increments

of the columns. The lowermost dosimeters, which were at 29.2 cm. depth, read out < 5, 7, 11, 14 and 15 R for columns I through V, respectively. The small variations of the exposure readings that did occur are believed to be due primarily to the shadowing and scattering of the gamma rays. The native soil in which the columns were buried was quite gravelly and contained few stones up to 6 inches across. None of these large stones, however, occurred within 12 inches of the buried columns. In any case, each LiF dosimeter reading represented the actual exposure level at the center of each soil column increment.

The thermoluminescence of the irradiated soils in relation to depth is shown in figure 3 in three ways, i.e., the total area under the glow curve with that of the natural thermoluminescence (TL) subtracted, the glow curve area in the sample heating temperature range below 200°C. and the glow curve height at 200°C. The limiting temperature of 200°C. was selected arbitrarily in order to work in the glow curve region showing no natural thermoluminescence (Hanford s.l.) or only a minimal amount of it (soil No. 4FF). The small amount that did occur at 200°C. and below in the case of soil No. 4FF was subtracted from the gross to obtain the radiation-induced thermoluminescence in the last 2 methods listed above. The figure shows that the radiation-induced soil thermoluminescence decreased with increasing depth. If these results are compared with those of figure 2, it is apparent that in general soil thermoluminescence increased as the radiation exposure level increased. Also, the response of the soils to radiation exposure was less sensitive and more variable than the LiF dosimeters.

The correlation coefficient of the variables fitted by least square

line indicates that soil thermoluminescence was indeed related to radiation exposure (figs. 4 and 5). (The column numbers indicated in the figures correspond to the removal numbers and dates shown in figures 2 and 3.) The results for Hanford s.l. (fig. 4) showed high correlation coefficients ($r \geq 0.967$). The "glow curve height" method showed higher correlation coefficient ($r = 0.978$) and lower standard deviation of the regression line slope ($s_b = 0.0001$) than either of the "area" methods. Of the latter methods, the "total area" method showed slightly higher correlation coefficient ($r = 0.975$) and lower standard deviation of the regression line slope ($s_b = 0.0010$) than the "area below 200°C " method ($r = 0.967$, $s_b = 0.0025$). With soil No. 4FF (fig. 5), the "glow curve height at 200°C " ($r = 0.946$, $s_b = 0.0002$) and the "area below 200°C " ($r = 0.953$, $s_b = 0.0004$) methods showed good correlation. The "total area" method ($r = 0.769$, $s_b = 0.0147$), however, showed relatively poor correlation, indicating that this method was not suitable for this soil. Although the data are not given in this paper, the glow curve height at 312° and 376° were also examined for soil No. 4FF. The correlation coefficient for the glow curve height at 312°C was $r = 0.80$ and that at 376°C was $r = 0.32$.

Figure 6 shows the ground surface Cs137 radiation exposure readings in relation to distance from the base of the source tower (experiment II). The points on the graph represent the radiation exposures at the sites where the experimental soils were located. At the site nearest to the tower, the exposure was 466R and that at the outermost site was 75 R. Note that the exposure readings varied somewhat and did not decrease in accordance to the simple inverse square law, although the soil stations

were located on a straight line from the source tower. Part of the variation may be caused by the variation of the terrain, but the primary cause is the design of the Cs137 source mount. As described in detail by French (2), the radiation source is differentially shielded with lead. The source, as seen from the ground near the base of the tower, is completely hidden behind lead shielding of differential thickness. At the periphery of the radiation field, 168 m. from the tower, no shielding interposes the source and the ground. Thus, the ratio of the radiation from the shielded portion to that from the unshielded portion changes with increasing distance from the tower. As indicated by the hump in the graph (fig. 6), the radiation field changes somewhat as the bare source, as seen from the ground, begins to emerge above the lead shielding. In the present experiment the radiation field served well in providing photon radiation of various energies and different exposure levels.

The soil thermoluminescence in relation to distance from the source is shown in figure 7. With Hanford s.l., the trends of the curves for all three methods of measuring thermoluminescence were quite similar to that of the radiation exposure readings with respect to distance (fig. 6). The curves for both the soils and the radiation exposure readings generally showed a minimum at 60-m. distance and then an increase to a peak at about 75 m. and a gradual decrease at farther distances from the source.

The correlation coefficient of the variables fitted by least square line showed close relationship between radiation exposure readings and

soil thermoluminescence (figs. 8 and 9). Hanford s.l. showed correlation coefficients of 0.943 or greater for the three methods of measurement (fig. 8). As in experiment I, the "glow curve height at 200°C" showed a lower standard deviation of the regression line slope, s_b , than either of the "area" methods. With soil No. 4FF (fig. 9), the "glow curve height" ($r = 0.938$, $s_b = 0.0003$) and the "area below 200°C" ($r = 0.915$, $s_b = 0.0008$) methods showed high correlation coefficients and low standard deviation of the regression line slope compared to the "total area" method ($r = 0.577$, $s_b = 0.0451$). This again indicates that the latter procedure was unsuitable for this soil. In general, the results of this experiment showed lower correlation coefficients for all methods than those of experiment I. Perhaps, this reflected the wider variation of photon energies to which the soils of experiment II were exposed. The variation of photon energies were, however, only a small factor, since high correlation coefficients were obtained with both experiments. Another factor that may have had an influence is the temperature during the irradiation period. The soils of experiment II were exposed to much wider and more intense temperature fluctuations, because their entire mass was exposed in thin layers on the ground surface. The effect of the environmental temperature fluctuations, however, were minimized, since the readouts were done 28 days after the removal of the soils from the radiation field. It has been shown previously (8) that the effect of temperature during irradiation can be eliminated or minimized by using an appropriate post-irradiation, pre-readout sample heating regime, or by allowing thermoluminescent decay to progress for a sufficient length

of time before readout.

In relating soil thermoluminescence to radiation exposure level, an important quantity is the minimum exposure level that can be measured with useful precision. The soil column data (experiment I) was examined to determine this quantity for the soils used and for the three techniques of measuring thermoluminescence. Since the quantity could not be determined precisely because of the limited number of samples and varying exposure levels and photon energies due to varying soil depth, only the range within which it might occur is given (table 1). In any case, the minimum measurable exposure level appeared to fall within the 20 to 85 R range. This is far above the sensitivity of LiF dosimeters which is in the low milliroentgen range (1).

In both experiment I and II, the effect of soil type was quite evident. Soil No. 4FF consistently showed the lower r values and the higher s_b values than Hanford s.l. This effect is due primarily to the high carbonate content of soil No. 4FF. Since carbonates show very high thermoluminescence compared to other components of the soil (5), a small variation of the carbonate content in the soil samples taken for readout can cause an appreciable variation in the amount of thermoluminescence.

The results of experiments I and II also showed that soil thermoluminescence is highly correlated with radiation exposure level. It appears that soil could be used as radiation dosimeters in terrestrial ecosystems exposed to relatively high levels of ionizing radiation. Any complicating effect that natural thermoluminescence might have

in determining the net exposure level could be avoided by using the "area below 200°C" or the "glow curve height at 200°C" methods. In these methods, a limiting temperature lower than 200°C could be used also, particularly at higher exposure levels than that which prevailed in the present experiments. In making the choice between the two methods, perhaps the "glow curve height" method should be selected, since its standard deviation of the regression line slope, s_b , was always less than that of the "area" methods. On the other hand, its regression line slope was in every case (figs. 4, 5, 8 and 9) lower than that for the "area" method. This fact implies that the "glow curve height" method was less sensitive in measuring the change of thermoluminescence in relation to radiation exposure. It will be noted that the maximum radiation dose to which the soils were exposed in experiments I and II were 548 and 466 R, respectively. At higher dose levels, which will cause the area below 200°C to increase, the precision of the "area" method may improve and become the better one to use. This, however, remains to be tested.

SUMMARY

Experiments were conducted to determine the relationship of soil thermoluminescence to ionizing radiation exposure under the conditions of natural environment using the Cs137 radiation field at the Nevada Test Site. In experiment I, soil columns were buried in the ground and irradiated at a fixed distance from the Cs137 source. In experiment II, they were exposed on the ground surface at 12 different distances from the source. Soil thermoluminescence was determined in three ways, i.e., glow curve area below 200°C, glow curve height at 200°C, and total area.

Both experiments showed that soil thermoluminescence is highly correlated with radiation exposure level. The first two methods of measuring thermoluminescence indicated above gave correlation coefficients $r > 0.915$. The "total area" method gave high correlation coefficient $r > 0.951$ for the noncalcareous soil, but low coefficient $r < 0.769$ with the highly calcareous soil studied, indicating that this method cannot be applied to the latter soil type. Under the experimental conditions that prevailed, the minimum radiation exposure level required to produce measureable soil thermoluminescence appeared to be in the 20-85 R range.

Fig. 1. Glow curves for Hanford sandy loam and soil No. 4FF before and after exposure to Cs137 photons (Experiment I)

Fig. 2. Lithium fluoride (TLD-100) dosimeter readings in relation to soil depth for different lengths of irradiation time

Fig. 3. Soil thermoluminescence as measured by three methods in relation to depth.

Fig. 4. Soil thermoluminescence as measured by three methods in relation to Cs137 radiation exposure level (Experiment I)
Hanford sandy loam

Fig. 5. Soil thermoluminescence as measured by three methods in relation to Cs137 radiation exposure level (Experiment I)
Soil No. 4FF

Fig. 6. Lithium fluoride (TLD-100) dosimeter readings in relation to distance from the base of the Cs137 source tower

Fig. 7. Soil thermoluminescence as measured by three methods in relation to distance from the base of the Cs137 source tower

Fig. 8. Soil thermoluminescence as measured by three methods in relation to Cs137 radiation exposure level (Experiment II) Hanford sandy loam

Fig. 9. Soil thermoluminescence as measured by three methods in relation to Cs137 radiation exposure level (Experiment II)
Soil No. 4FF