Alfred W. Klement, Jr. (Editor)
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If you have any questions regarding this material please don't hesitate to contact us.

Sincerely,

Harold L. Beck

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Radiation Physics Division

5 Enclosures:
3 Reprints
2 HASL Reports

NAT. [illegible] SPECIAL [illegible]

[illegible] DOSES [illegible] PHOTONS - YES

Spectrometric Determination of Dose Rates from Natural and Fallout Gamma-Radiation in the US 1962-63

Lowder, Beck Condon

Nature, Vol, 202, No 4934 pp 745-49 May 23 1964

Measurement of the Exposure of Human Populations to Environmental Radiation. Lowder + Condon

Nature, Vol 204, No 4985, pp 658-662 May 15, 1965

HASL Surface Air Sampling Program

Environmental Gamma Radiation from Deposited Fission Products, 1960-64

Beck

H P. 1966 Vol 12 pp 313-322

HASL-145

Environmental Radiation Measurements in the Southeastern, Central and Western United States

Beck, Condon, Lowder Apr 1964

sample radiometry or inferences from information on similar formations elsewhere[5]. It seemed possible that these differences in mean bedrock radioactivity might be reflected in significant differences in mean radiation exposure between the populations of these areas.

Entirely unrelated approaches were utilized by the Health and Safety Laboratory and Harvard groups in

Harvard group undertook a more direct approach and distributed integrating dosimeters to representative members of the population to be worn on the person.

For the purpose of comparing the two sets of results, we have defined 'population exposure' as the free air dose 'experienced' by typical individuals in their usual rounds of activity. We consider only the air dose from the more penetrating components of the environmental radiation field, that is, the terrestrial γ- and ionizing cosmic radiation. This definition can be justified by the fact that the mean γ-doses to the skin and at various depths in the body are fairly well-known fractions of the free air γ-dose[9,10], and only the more penetrating components of the external radiation field significantly contribute to the dose at the depths of greatest interest, that is, those of the gonads and bone. It should be noted that the possibly important tissue dose contribution from cosmic ray neutrons is not determined by the techniques used in these investigations.

The Health and Safety Laboratory radiation survey was carried out in July and August 1962, and subsequent check measurements were made in May and September 1963 and May 1964. The instrumentation included high-pressure argon ionization chambers for total dose-rate measurements, a γ-spectrometer system for determination of component dose rates (particularly necessary for discrimination between the natural and fall-out γ-radiation), and portable scintillation detectors for surveys of the areas surrounding each outdoor measurement location as well as for the indoor measurements. The survey techniques, described in detail elsewhere[11-13], provide an overall accuracy of ± 5 per cent (*S.D.*) for the measured total dose-rate values and approximately ± 10 per cent for each of the various components of the total radiation field. The outdoor readings were taken in large, flat open spaces situated in populated areas (for example, parks, fields, lawns, vacant lots) with the instruments placed 3 ft. above the ground. The number of measurements in each area was determined by its population and size, the availability of proper sites, and the observed range and pattern of the previous readings. A sufficient number was taken to ensure that a reasonable radiation profile could be constructed for each area. The quite limited

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range in the measured dose rates in each case (±10 per cent about the mean for most field readings) meant that relatively few locations were required for this purpose. Most of the measurements were made in the larger towns, as these contained the bulk of the population.

The mean γ-dose rates over open ground derived from the spectrometer readings are given in Table 1 for each of the eight regions chosen for examination. The main towns and the corresponding bedrock formations are indicated. The spectrometric procedures allow determinations of the component dose rates from potassium-40, the uranium and thorium series, and the main fall-out γ-emitters (^{95}Zr–^{95}Nb, ^{103}Ru, ^{106}Rh, ^{137}Cs). Estimates of mean soil concentrations of the natural radioisotopes can be calculated from the component dose rates, assuming uniform depth distribution in the ground[10,11]. Such concentration values are representative of the true soil contents of radium, uranium and thorium only when these isotopes are in radioactive equilibrium with their γ-emitting daughters. These results are included in Table 1, and the mean values for the natural emitters are combined to give a mean 'equivalent uranium' (eU) concentration for the upper layers of the ground, that is, the amount of uranium in equilibrium with its daughters that would yield the same γ-dose rate as the potassium and the uranium and thorium series in the actual situation. Also listed for comparison purposes are the mean equivalent uranium contents of the various bedrocks as estimated by Billings[5]. It is noteworthy that the range of soil activities is much narrower than that inferred for the bedrock formations.

The average fall-out levels indicated in Table 1 apply specifically to July and August 1962, when almost all readings fell between 2 and 3 μr./h. When some of these sites were re-checked in 1963, the fall-out dose rates were nearly twice as great, ranging from 3 to 5 μr./h. In all cases, ^{95}Zr–^{95}Nb was the dominant contributor. By May 1964, levels of 1 μr./h were typical, the ^{95}Zr–^{95}Nb and other short-lived emitters having nearly disappeared, leaving ^{137}Cs as the most important fall-out γ-emitter. These results are quite consistent with the more detailed information available for the New York City area during this period[14].

In addition to the combined ionization chamber and spectrometer readings summarized in Table 1 (5–10 in each region, except for 16 at Conway), separate ionization-chamber readings were made at many other sites along with a number of independent portable scintillometer surveys. The uniformity of the outdoor radiation levels in each area was remarkable. In general, streets and side-walks did not significantly alter the observed profile.

Any attempt to estimate population exposure to environmental radiation must take into account the

Table 1. MEAN OUTDOOR γ-RADIATION LEVELS

Region	Bedrock	eU* (p.p.m.)	K		U		Th		Natural γ		Fall-out γ
			μr./h	%	μr./h	p.p.m.	μr./h	p.p.m.	μr./h	eU (p.p.m.)	μr./h
New Hampshire											
1. Manchester	Fitchburg Granite	23	2·5	1·5	1·1	1·5	3·4	9·4	7·0	9·2	2·2
2. Concord	Binary Granite	26	2·9	1·7	1·2	1·6	4·3	11·9	8·4	11·1	2·2
3. Franklin	Littleton Formation	23	2·4	1·4	1·2	1·6	3·5	9·7	7·1	9·4	1·8
4. Conway	Conway Granite	45	3·6	2·1	1·7	2·2	5·6	15·6	10·9	14·3	2·7
Vermont											
1. Rutland	Dunham Dolomite	5	2·6	1·5	1·0	1·3	2·0	5·6	5·6	7·4	2·3
2. Middlebury	Beldens Formation	5	3·2	1·9	0·8	1·1	2·6	7·2	6·6	8·7	2·0
3. Bennington	Glacial Drift	9	3·3	1·9	1·0	1·3	2·2	6·1	6·5	8·6	2·0
4. Burlington	Monkton Formation	11	2·4	1·4	1·0	1·3	1·8	5·0	5·2	6·8	2·4

* Mean bedrock radioactivity as estimated by Billings[5].

Table 2. MEAN WEEKLY DOSES IN MR./WEEK

Geological category	eU (p.p.m.)	Outdoor Natural γ	Outdoor Total γ*	Cosmic	Mean population exposure		
					1962†	Natural	Dosimeters
Dunham Dolomite	5	0·94	1·34	0·62	1·69	1·37	2·97
Beldens Formation	5	1·11	1·43	0·60	1·74	1·49	2·86
Glacial Drift	9	1·09	1·43	0·62	1·76	1·49	3·01
Monkton Formation	11	0·87	1·27	0·59	1·61	1·29	2·70
Fitchburg Granite	23	1·18	1·52	0·60	1·82	1·54	3·13
Littleton Formation	23	1·19	1·63	0·61	1·91	1·56	2·97
Binary Granite	26	1·41	1·78	0·60	2·05	1·73	3·21
Conway Granite	45	1·83	2·27	0·61	2·43	2·07	3·67

* Includes fall-out, averaged over all locations within an area.
† Obtained as indicated in the text; in units of air dose rate.

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Monkton Formation	11	0·87	1·27	0·59	1·61	1·29	2·70
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Littleton Formation	23	1·19	1·63	0·61	1·91	1·56	2·97
Binary Granite	26	1·41	1·78	0·60	2·05	1·73	3·21
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effect of man-made structures on ambient radiation fields, since most individuals spend a large fraction of their time indoors. Portable scintillation detector readings were made in 160 private homes and apartments in the main towns to ascertain whether any consistent relationship existed between indoor and outdoor radiation-levels. Several rooms in each dwelling were surveyed, including the living-room and at least one bedroom. Again, a strong uniformity exhibited itself in that the mean indoor levels were close to 70 per cent of the corresponding outdoor levels in each area[4]. This may be related to the fact that the vast majority of the dwellings were of wood-frame construction, with the building materials appearing to act generally as γ-ray shields with relatively little activity of their own.

With such data at hand, an estimate of mean population exposure to environmental radiation can be obtained by calculating a suitably weighted average of the indoor and outdoor readings of the survey instruments. Taking into consideration the greater occupancy time indoors of the average individual, the mean exposure levels have been estimated to be 80 per cent of the mean outdoor terrestrial γ-dose rates given in Table 1, plus the contribution from the ionizing components of the cosmic radiation at the ground altitudes of the various areas[15]. No correction of the cosmic-ray figures for typical structural shielding has been made, since this would be a reduction of the order of 10 per cent or less, which is comparable to the present uncertainty in the absolute cosmic-ray ionization intensity.

Table 2 shows the population exposure data arranged by geological region. The mean weekly outdoor doses in air are given for both natural and total (natural plus fall-out) γ-radiation and for cosmic rays, and the Health and Safety Laboratory population exposure estimates for the time of the survey (August 1962, including fall-out) and for the natural emitters only (that is, the mean life-time levels neglecting fall-out) are also given. The importance of the spectrometric technique is emphasized by the fact that estimates of the integrated natural γ-dose were obtainable even under conditions of near-maximum fall-out contamination. In many population investigations, it is just this quantity that is desired.

The Harvard investigation involved the use of a set of 200 Victoreen model 362 condenser ionization-chamber pencils, along with a stable pulse height readout system[16,17] which is designed to allow readings of $1{\cdot}0 \pm 0{\cdot}2$ mr. at the 95 per cent confidence level with a single pencil. Mechanical and thermal stability was tested, and corrections made for average leakage rates observed in the laboratory.

These dosimeters were distributed in pairs to five individuals in standard occupational categories in each of 16 areal units, half urban and half rural. The dosimeters

were worn for one week, collected, read, and then redistributed. The experiment was conducted for five weeks, resulting in a total sampling of 400 individuals, 25 in each areal unit (that is, 50 in each of the eight geological regions). The details of this study are discussed by Segall[1,3].

Estimates of population exposure from the mean values of the Harvard dosimeter data in the various areas are given in the last column of Table 2. These air dose values are derived from Segall's data[1,3] by assuming that each of the dosimeters, worn on the body surface, read 100 per cent of the cosmic-ray ionization and 85 per cent of the γ-ray ionization in free air. The latter figure is based primarily on the recent measurements of body attenuation factors by Spiers and Overton[9].

The Health and Safety Laboratory and Harvard population exposure results are plotted as a function of estimated mean bedrock radioactivity in Fig. 1, with the respective regression lines indicated. Plotted also in Fig. 1

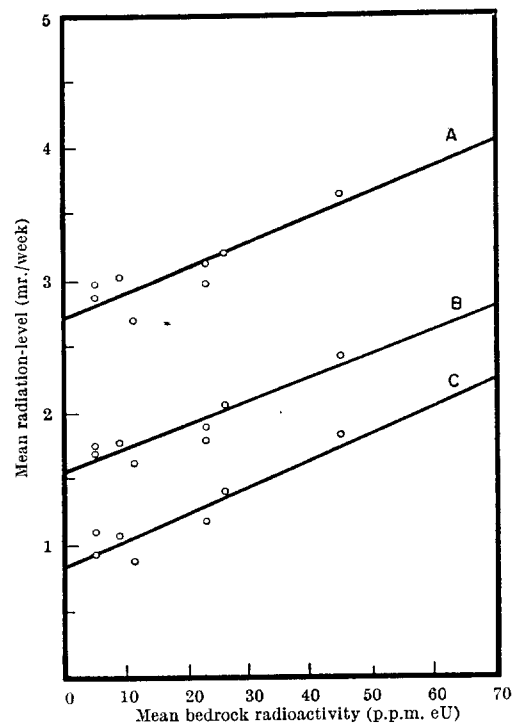

Fig. 1. Environmental radiation in the eight selected areas of northern New England as a function of estimated mean bedrock radioactivity. *A*, Population exposure estimates from Harvard dosimeters; *B*, population exposure estimates based on Health and Safety Laboratory *in situ* measurements; *C*, mean weekly outdoor γ doses

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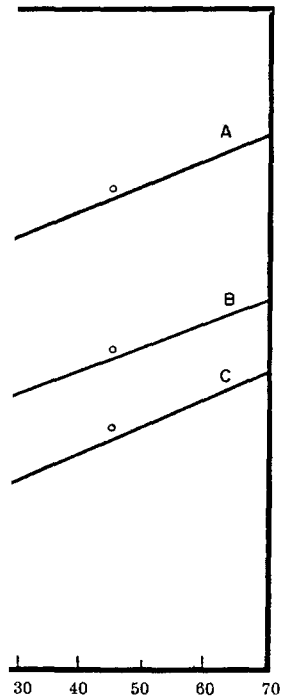

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(lowest curve) are the mean outdoor natural γ-levels obtained from the Health and Safety Laboratory spectrometer readings. These values are directly proportional to mean soil content of natural radioactivity (1 mr./week $\rightarrow$ 7·83 p.p.m. eU).

It is readily apparent from the figure that the two sets of population exposure data show a considerable systematic difference. The large intercept at zero bedrock radioactivity for the linear fit to the Harvard dosimeter data cannot be explained simply on the basis of the relatively constant cosmic-ray and fall-out dose contributions, which are less than 1·0 mr./week. There is certainly no evidence to suggest that building materials produce elevated radiation levels indoors in any consistent manner. The intercept for the Health and Safety Laboratory results is more reasonable, although also somewhat high. It is interesting to note that, while an apparent linear trend appears to exist for all sets of data, the interpretation of this trend is not obvious. For example, the slope of the natural γ-dose regression line is only one-sixth of that expected if the bedrock were the source of the radiation[11]. In a sense, the results shown in Fig. 1 provide a rough indication of the influence of bedrock geology on soil radioactivity and natural radiation exposure in these areas. The effect is small and may be of practical significance only in the Conway area, where the reddish sand derived from the thorium-rich Conway granite is present in the soil throughout the populated areas. The trend of the results in Fig. 1 may be indicative of some fairly consistent relation in these areas between the bedrock formations and their respective overburdens of soil in terms of natural radioactivity. The apparent near-linearity of the population exposure estimates as a function of bedrock radioactivity derives from the similar relationship between estimated mean soil and bedrock radioactivities, since the outdoor (and to some extent the indoor) radiation-levels to which the general population is exposed are closely related to the content of natural γ-emitting radioisotopes in the upper layers of the soil.

Fig. 2 shows the Harvard dosimeter data plotted directly as a function of the Health and Safety Laboratory 1962 total exposure results. The high degree of correlation ($r > 0{\cdot}9$) between the Harvard and Health and Safety Laboratory estimates of population exposure is evident; a line of unit slope fits the data quite well. The 1·2 mr./week value for the Y-axis intercept of this line is a measure of the apparently systematic deviation between these two sets of data. While not enough information is at present available to explore this problem fully, one obvious possibility is that the pocket dosimeters consistently exhibited enhanced leakage under field conditions as compared with that measured in the laboratory and corrected for in the data interpretation.

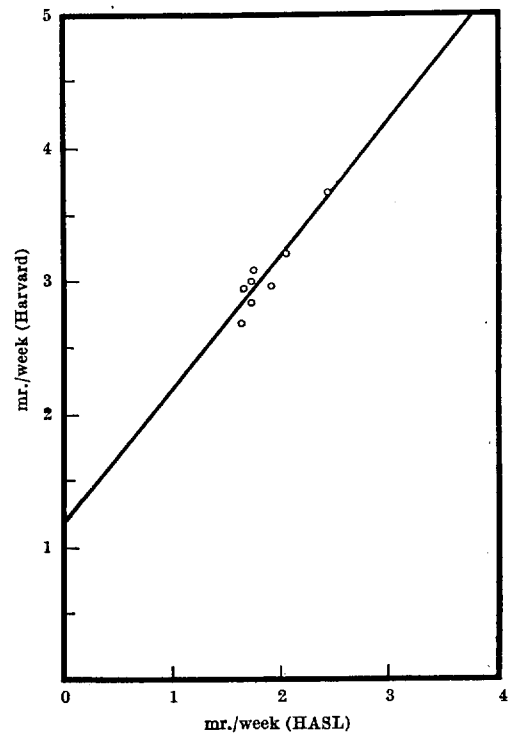
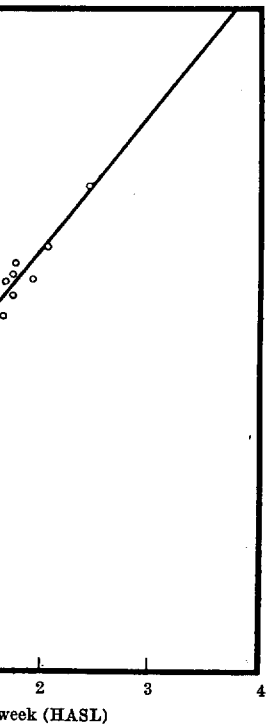


Fig. 2. Harvard population exposure estimates as a function of comparable Health and Safety Laboratory (HASL) estimates for the eight New England areas examined

In any event, there is little doubt that the dosimeter results are too high. This can be shown by carrying out a simple mathematical analysis of the various contributions to the population exposure-level, P, utilizing the accurate Health and Safety Laboratory measurements of outdoor environmental radiation dose rates. If I_c, I_n and I_f are the measured mean outdoor dose-rate contributions from cosmic, natural γ-, and fall-out γ-radiation, respectively, and I_h is the mean indoor γ-dose rate pro uced by sources in the building materials, we can write the following expression for P:

$$P = f_i\,(I_c + s_f I_f + s_n I_n + I_h) + f_o\,(I_c + I_f + I_n)$$

where f_i and f_o are occupancy time factors for indoor and outdoor locations, respectively, and s_f and s_n are mean transmission factors of the buildings and residences for outdoor fall-out and natural γ-radiation. Substituting



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reasonable values for these factors and the various weekly doses into the formula, we get:

$$P = 0{\cdot}8\,[0{\cdot}60 + (0{\cdot}2)\,(0{\cdot}37) + (0{\cdot}3)\,(1{\cdot}18) + I_h] + 0{\cdot}2\,[0{\cdot}60 + 0{\cdot}37 + 1{\cdot}18] = 1{\cdot}25 + 0{\cdot}8\,I_h \text{ mr./week}$$

Since we have determined that the indoor total γ-levels average 0·7 of the outdoor levels in these areas, we find that:

$$I^h \simeq 0{\cdot}5\,I_f + 0{\cdot}4\,I_n = 0{\cdot}66 \text{ mr./week}$$

Substituting this in the above expression for P, we get:

$$P \cong 1{\cdot}8 \text{ mr./week}$$

This result is not strongly dependent on the particular values assumed for the various factors in the above equation. It is quite consistent with the similarly calculated Health and Safety Laboratory population exposure estimates, and much lower than the dosimeter results. The mean contribution from building materials to population exposure would have to be close to 2 mr./week to validate the dosimeter data, which is considerably higher than the measured values for the total indoor γ-dose rate in most of the 160 residences where scintillation detector readings were made. Even without such evidence, it seems to be an unreasonably high value to assign to mean regional indoor radiation-levels produced by radioactivity in building materials. For it implies total indoor γ-doses averaging approximately 3 mr./week, whereas the scattered data given in the 1962 United Nations report[18] indicate that readings of 1 mr./week are typical of normal situations in wood or brick houses.

The results of both surveys indicate that the range of population exposure to environmental radiation is quite narrow throughout the regions studied. It follows that northern New England does not provide a good 'laboratory' for the study of the effect on large human populations of differences in long-term environmental radiation exposure. Of much greater significance is the correlation between the two entirely independent and undoubtedly somewhat imprecise techniques for estimating these exposure-levels. This correlation can be at least partially understood as a consequence of the relatively high degree of uniformity in radiation-levels observed within each area. Under such fortunate conditions, the method of using a few hundred field measurements to infer the total radiation profile has yielded apparently realistic values for population exposure, for which the Harvard dosimeter data provide strong qualitative support. Spiers *et al.*[7], in their discussion of the extensive population investigation in Scotland, have already indicated some of the difficulties involved in obtaining and interpreting data of this type. But it can be concluded from the work recorded here that

the *in situ* approach is capable of yielding useful quantitative results in a reasonably uniform radiation environment, and at the very least can be used as a basis for evaluating more direct—but not necessarily more accurate—methods of estimating mean population exposure-levels.

Another conclusion which is suggested by the New England results is that the basic limitation of the pocket ionization-chamber technique in terms of measuring normal human exposure to environmental radiation is now the difficulty in determining mean leakage rates under actual field conditions while being worn and handled. There appears to be no fundamental reason why this difficulty cannot be at least partially overcome by suitably controlled experimentation, and thus the pocket chamber technique can be considered as a potentially practical one for this kind of measurement. It should be remarked that the dosimeters admirably fulfilled their basic purpose in the Harvard investigation, namely, the determination of differences in population exposure-levels between areas.

There are, of course, a number of other possible methods for determining mean population exposure to environmental radiation. For example, photographic film dosimetry techniques have been applied to this general problem area with some success. O'Brien *et al.*[19] described a film-scintillator (sodium iodide) system which Roser and Cullen[20] have utilized in the measurement of population exposure in Brazil on a limited scale. The approximately thousand-fold enhancement of the film response produced by the scintillator is almost too great for the high-background areas of Brazil; such a method would almost certainly be feasible in areas of more normal background levels for certain kinds of studies. The basic limitation here is the cost of the dosimeters, which precludes their widespread use. The problem of reciprocity law failure must also be taken into account in the calibration of the dosimeters.

A similar kind of dosimeter has been described by Henson[21], using photographic film and a plastic scintillator (*N.E.* 102). While less sensitive than the sodium iodide system, it exhibits little energy dependence and good precision (± 10 per cent *S.D.* for two weeks' exposure at normal background). Reciprocity failure was observed but has not proved excessive. The main problem seems to be a strong dependence on temperature in its response, which varies with the dose rate. The error present in any particular reading is not known, so that the use of this dosimeter has not been recommended.

There has also been recent progress in increasing the sensitivity of normal radiographic film by means of post-exposure to visible light and improved development techniques that may render such film useful for environmental radiation studies without the necessity for external enhancement of its response. McLaughlin[22] has reported

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respectively, with approximately ± 20 per cent accuracy (*S.D.*). These limits may eventually be somewhat lowered and the precision improved with refinements in read-out techniques. Cullen[25] has recently utilized 156 lithium fluoride dosimeters for a population exposure investigation in a high background area in Brazil, with 50 mg of the material placed in religious medals to be worn for a three-month period. This exposure time provided a total γ-dose of several hundred milliroentgens, well above the minimum now routinely detectable. In general, thermoluminescent dosimeters have the significant advantages of small size and relatively low unit cost, and may prove to be a useful tool for future population investigations. The Health and Safety Laboratory is at present engaged in evaluating the available thermoluminescent dosimeter systems for their applicability to the routine measurement of human exposure to environmental radiation, and field tests along the lines of the New England survey are planned when sufficiently promising dosimeter systems are developed.

These recent advances in direct personnel dosimetry, particularly in the extension of the sensitivity limits to ever lower γ-dose levels, render the detailed examination of human exposure to environmental radiation on a routine basis increasingly feasible, even in the extremely low-level radiation fields that are characteristic of the normal environment. But the reliability and reproducibility of the readings of the various types of dosimeter in terms of absolute dose under the stresses of actual field use remain to be thoroughly explored. The New England survey results seem to indicate the adequacy of *in situ* measurements in establishing a radiation profile over extensive areas, a profile that when sufficiently uncomplicated may be properly interpreted in terms of population exposure to environmental radiation. These results also emphasize some of the problems associated with adequately calibrating the response of personnel dosimeters under field conditions. It appears that the use of highly accurate ionization-chamber and spectrometric techniques for *in situ* measurements will be required in the near future for all population studies of the type described here, if only to provide a standard by which the adequacy of the new techniques for direct human exposure measure-

ments can be determined (for example, ref. 20). Eventually, it is anticipated that the logistically difficult *in situ* approach will be completely superseded by direct measurements of radiation incident on representative individuals in their daily rounds.

We thank our colleague, H. L. Beck, as well as several Health and Safety Laboratory summer students, including J. Grebowsky, H. Grotch and I. Hammerman, for their assistance with the outdoor survey. A. Spiegal carried out the indoor measurements. Many helpful discussions have been held with Dr. A. Segall of the Harvard School of Public Health, who co-ordinated the various parts of the New England survey and who kindly supplied the dosimeter results. We also thank J. E. McLaughlin, director, Radiation Physics Division, Health and Safety Laboratory, for his advice and support.

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