## Radiogeology and Population Exposure to Background Radiation in Northern New England

The possibility that chronic exposure to low doses of ionizing radiation is of etiologic significance in the genesis of certain human diseases has, in recent years, stimulated interest in the effects of exposure to natural background radiation. In connection with epidemiologic studies of cancer and of congenital malformations in northern New England, we have during the last 3 years undertaken a variety of surveys of natural radioactivity in this area. These include: (i) a radiogeological survey of bedrock radioactivity in Maine, New Hampshire, and Vermont (*1*); (ii) a personnel-monitoring survey of population exposure to external gamma radiation in selected areas of high and low bedrock radioactivity (*2*); (iii) a survey of the concentration of radium-226, radium-224, and polonium-210 in teeth extracted from life- or long-time residents of these same areas (*3*).

In this report the findings of these surveys will be correlated in an attempt to characterize the dose from external and internal natural emitters received by populations resident in areas of differing bedrock radioactivity.

The bedrock of northern New England is of Paleozoic age, except for some Precambrian in the southern part of the Green Mountains of Vermont. In northwestern Vermont, in and immediately east of Lake Champlain, limestone and dolomite, along with some shale and sandstone, are common. In the Taconic Mountains, the Green Mountains, all of eastern Vermont, and in all of New Hampshire and Maine the original sedimentary rocks were largely clastic—originally shale and limestone. All the rocks, except in northwestern Vermont, are regionally metamorphosed primarily to schists and gneiss. While Paleozoic granitic rocks are not abundant in Vermont or in the northern third of Maine, they are extensively developed throughout New Hampshire and the southern two-thirds of Maine.

For comparative purposes, the radioactivity of bedrock may be expressed in terms of equivalent uranium in parts per million. This is the amount of uranium which, by itself, would yield the same quantity of gamma radiation in roentgens as the uranium, thorium, and potassium-40 in the particular rock.

The equivalent uranium content was calculated for 58 geological formations in New Hampshire, 5 in Maine, and 24 in Vermont. Five principal sources of data were used: chemical analysis of rock specimens, alpha counts of rock specimens, direct measurements of outcrops in traverses, calculations based on counts obtained in airborne surveys, and lithological similarity to rocks for which data are available in the literature (*1*).

An isorad map of northern New England indicating the distribution of equivalent uranium is shown in Fig. 1.

Table 1. Radiogeology of areas surveyed in northern New England.

| Geological category | Rock type | Average equivalent uranium (ppm) |
|---|---|---|
| Dunham dolomite (Vt.) | Mainly a siliceous, buff-weathered dolomite containing well-rounded sand grains irregularly distributed. | 5 |
| Beldens formation (Vt.) | Interbedded buff to brown dolomite and white to blue-gray marble and limestone. | 5 |
| Glacial drift (Vt.) | Till, sand, and gravel overlying Monkton quartzite, Dunham dolomite, or Clarendon Springs dolomite. | 9 |
| Monkton formation (Vt.) | Red quartzite interbedded with lesser amounts of buff and white quartzite and relatively thick gray dolomite. | 11 |
| Fitchburg granite (N.H.) | A heterogeneous, primarily medium-grained to coarse-grained pink granite gneiss; locally, as at Milford, intrusive binary granites are present. | 23 |
| Littleton formation (N.H.) | A very heterogeneous formation composed primarily of mica schist, quartz-mica schist, and gneiss. | 23 |
| Binary granite (N.H.) | Fine-grained to medium-grained, locally coarse-grained, light-gray to white granite composed of potash-feldspar, quartz, oligoclase, and lesser amounts of biotite and muscovite. | 26 |
| Conway granite (N.H.) | Fine-grained to coarse-grained pink biotite granite. | 45 |

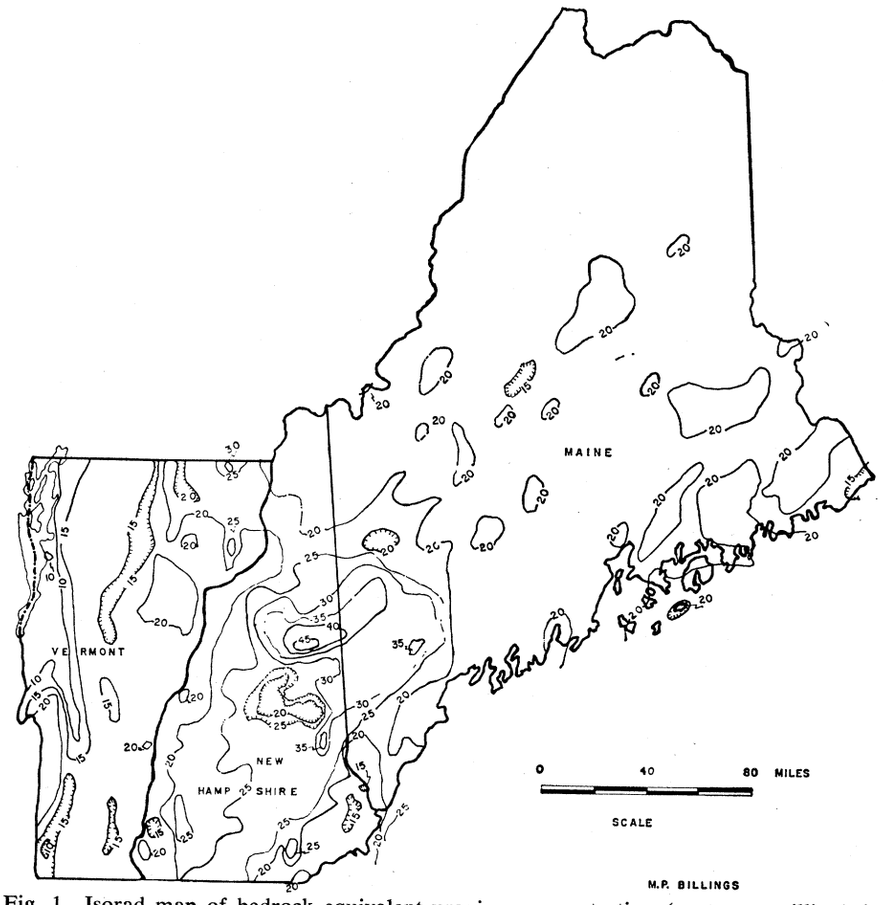


Fig. 1. Isorad map of bedrock equivalent uranium concentration (parts per million) in northern New England.

Table 2. Population exposure to external radiation by geological category; mean weekly dose rate (mr).

| Geological category | Personnel-monitoring survey | | Pressurized ion chamber survey | | |
|---|---|---|---|---|---|
| | Number of measurements | Total exposure (mr/wk) | Number of measurements | Terrestrial component (mr/wk) | Cosmic component, estimated from altitude (mr/wk) |
| | | *Urbanized areas* | | | |
| Dunham dolomite | 50 | 2.79 | 8 | 1.68 | 0.71 |
| Beldens formation | 50 | 2.41 | 6 | 1.64 | .68 |
| Glacial drift | 50 | 2.68 | 3 | 1.42 | .70 |
| Monkton formation | 50 | 2.29 | 10 | 1.30 | .63 |
| Fitchburg granite | 50 | 2.68 | 12 | 2.03 | .65 |
| Littleton formation | 50 | 2.53 | 3 | 1.79 | .66 |
| Binary granite | 50 | 2.81 | 4 | 1.96 | .64 |
| Conway granite | 50 | 3.15 | 7 | 3.13 | .68 |
| | | *Rural areas* | | | |
| Dunham dolomite | 50 | 2.48 | 6 | 1.88 | 0.68 |
| Beldens formation | 50 | 2.63 | | | |
| Glacial drift | 50 | 2.61 | 4 | 1.50 | .70 |
| Monkton formation | 50 | 2.47 | 4 | 1.33 | .64 |
| Fitchburg granite | 50 | 2.83 | 2 | 1.99 | .66 |
| Littleton formation | 50 | 2.71 | 2 | 1.74 | .70 |
| Binary granite | 50 | 2.83 | 6 | 2.32 | .69 |
| Conway granite | 50 | 3.28 | 4 | 2.63 | .69 |

Values range from 5 parts per million in the limestone areas of Vermont to 45 parts per million in the granitic regions of New Hampshire.

To determine the relationship between bedrock radioactivity and the population dose from external background radiation, a personnel-monitoring survey was conducted in selected areas of Vermont and New Hampshire. The dosimetric system used consisted of Victoreen model 362 condenser chamber pencils and a pulse-height readout and recharging unit. The specifications and characteristics of this system relevant to its application in population studies have been discussed elsewhere (*4*).

The survey encompassed 16 areal units located in a sample of rural and urbanized towns overlying eight geological formations which include the full range of equivalent uranium concentration in northern New England (Table 1). In Vermont, the towns surveyed are built on various calcareous and sandstone formations which are representative of the lower values in the region (5 to 11 parts per million). The towns in New Hampshire overlie several types of granitic and gneiss bedrock which is characteristic of the regional upper limit (23 to 45 parts per million).

Factors such as age, sex, and occupation were controlled by limiting distribution of the dosimeters to five standard occupational categories. The cumulative dose was measured over a period of 1 week for five successive weeks. In each of the 16 areal units a weekly sample of five persons consisting of one representative from each of the five occupations was monitored.

The mean weekly dose rate by geological category for urbanized and rural areas is shown in Table 2. Values ranged from 2.29 to 3.28 mr/week. Analysis of variance revealed intercategory differences to be statistically significant ($p < .01$). When the mean weekly dose rate is plotted against local equivalent uranium concentration (Fig. 2) a linear trend is observed. This trend is significant ($p < .01$). The data are therefore consistent with the hypothesis that the level of population exposure to sources of external background radiation is determined both by a linear effect of radioactivity in the underlying bedrock and by local factors unrelated to this linear component; the latter might include the nature and depth of overlying soil and the types of construction materials used in buildings.

The terrestrial component of external background radiation in the same 16 areal units was measured concommitantly by a team from the Health and Safety Laboratories, U.S. Atomic Energy Commission, with a pressurized ion chamber (Table 2). Values ranged from 1.30 to 3.13 mr/week. The total dose from external sources as recorded by personnel monitoring shows significant correlation with the terrestrial component as measured with the pressurized ion chamber ($r = .97$; $p < .001$). Geographic variation in the ionizing component of the cosmic flux was estimated as a function of altitude (*5*).

The relationship between bedrock radioactivity and the skeletal dose from natural internal emitters was examined by measuring the concentration of radium-226 and polonium-210 in 160 teeth extracted from life- or long-time residents of eight municipalities in the same geological regions. In this connection, a method of measuring the concentration of radium-226, radium-224, and polonium-210 in a single

Table 3. Population exposure to internal emitters by geological category. Skeletal data are estimated annual skeletal dose rates.

| Number of teeth examined | Radium-226 | | Polonium-210 | | Radium-226 in tap water (pc/lit.) |
|---|---|---|---|---|---|
| | Mean concn. per gram ash (pc/g) | Skeleton (mrad/year) | Mean concn. per gram ash (pc/g) | Skeleton (mrad/year) | |
| | | *Dunham dolomite* | | | |
| 25 | 0.014 | 1.014 | 0.050 | 3.406 | 0.04 |
| | | *Beldens formation* | | | |
| 15 | .020 | 1.486 | .057 | 3.865 | .03 |
| | | *Glacial drift* | | | |
| 20 | .009 | 0.682 | .052 | 3.546 | .08 |
| | | *Monkton formation* | | | |
| 20 | .010 | .744 | .047 | 3.153 | .02 |
| | | *Fitchburg granite* | | | |
| 20 | .018 | 1.352 | .056 | 3.783 | .04 |
| | | *Littleton formation* | | | |
| 20 | .016 | 1.184 | .061 | 4.136 | .04 |
| | | *Binary granite* | | | |
| 20 | .014 | 1.008 | .050 | 3.397 | .02 |
| | | *Conway granite* | | | |
| 20 | .025 | 1.814 | .059 | 4.025 | .01 |

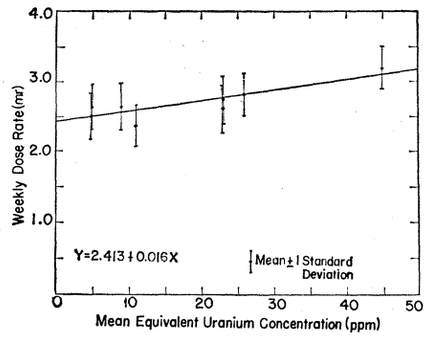


Fig. 2. Weekly dose rate from external radiation and local equivalent uranium concentration.

tooth specimen was developed (*6*). Prior bovine and human studies had suggested that teeth may be used to indicate the body burden of radium-226 if it is acquired by chronic exposure (*7*). Each sample included teeth from persons ranging in age from under 15 to over 65. No association between the concentration of either radium-226 or polonium-210 with age was noted.

The mean concentrations of radium-226 in teeth ranged from 0.009 to 0.025 pc/g of ash (Table 3). For analysis of variance, a log transformation was used in view of the marked differences in intracategory variability and the tendency of this variability to increase as the mean value for the category increases. Differences between geological categories were significant ($p < .01$). However, in contrast to the findings with respect to external radiation, the linear component of intercategory variability in the concentration of radium-226 in teeth which is related to equivalent uranium concentration is not significantly greater than the residual component which is associated with other factors of a local nature ($p > .05$). The present data would therefore suggest that if an effect of local bedrock radioactivity on the dose from radium-226 is present, it is too small to be detected in the number of teeth analyzed. Consistent with these findings is the observation that the concentration of radium in samples of tap water from the eight municipalities varies from 0.01 to 0.08 pc/lit. but shows no significant correlation with bedrock radioactivity ($r = -.51$; $p > .05$) (*8*).

Table 4. Estimated mean annual bone tissue dose (mrem/year) from natural internal emitters in northern New England.

| Emitter | Relative biological effect | |
|---|---|---|
| | 4 | 10 |
| Radium-226 | 4.6 | 11.6 |
| Thorium-228 | 3.3 | 8.3 |
| Lead-210 | 14.7 | 36.6 |
| Potassium-40* | 15.0 | 15.0 |
| Carbon-14* | 1.6 | 1.6 |
| Total | 39.2 | 73.1 |

* Standard value.

Values for the concentration of polonium-210 in teeth range from 0.047 to 0.061 pc/g of ash. Intercategory variation is not statistically significant ($p > .05$). Differences between individuals with respect to this radioisotope are more marked than is the case for radium-226.

The concentration of radium-224 was measured in only 46 teeth with an unequal distribution between the eight geological categories. It was considered inadvisable, on the basis of this small sample, to evaluate the relationship between the tissue dose from this element and bedrock radioactivity. The mean value for teeth from all regions is 0.007 pc/g of ash.

The annual dose to bone tissue from radium-226, lead-210, and thorium-228 may be estimated from their concentration in teeth. Although polonium-210 and radium-224 were measured, the amounts of these daughter products may be assumed to be in equilibrium with their longer-lived parents lead-210 and thorium-228, since the teeth were extracted a year or more before analysis. Table 4 shows the estimated annual dose to bone tissue for a relative biological effect of 4 and 10. These values were calculated from the overall mean concentration of the relevant radioisotopes from all municipalities, since there was no evidence in the present data to suggest that the tissue dose from these natural internal emitters varies significantly in the study area as a function of bedrock radioactivity.

These results indicate some of the difficulties inherent in the use of bedrock radioactivity as an index of geographic variation in population exposure to background radiation. As far as external emitters are concerned, it was possible to demonstrate a significant linear relationship between population dose rate and equivalent uranium concentration. However, a ninefold difference in equivalent uranium concentration between the categories of highest and lowest bedrock radioactivity was found to correspond to a difference in dose rate in the order of only 43 mr/yr. No significant relationship between equivalent uranium concentration and the level of radium-226 and polonium-210 in teeth was observed. Thus, while the geological data suggested considerable intercategory variation in terrestrial sources of radioactivity, dosimetric evaluation of population exposure revealed relatively small differences only with respect to external gamma radiation (*9*).

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## Microdetermination of Calcium by Aequorin Luminescence

Abstract. *A bioluminescent protein, aequorin, isolated from the jellyfish* Aequorea *in dilute disodium ethylenediaminetetraacetate solution, emits light on addition specifically of $Ca^{++}$ or $Sr^{++}$, thus providing the basis for a simple, quantitative micromethod for the determination of these cations, especially in biological fluids.*

Homogenates of photogenic tissues of the jellyfish *Aequorea* contain a bioluminescent protein, aequorin, which has recently been extracted in disodium ethylenediaminetetraacetate (EDTA-2Na) solution and purified chiefly by chromatography on diethylaminoethyl cellulose (DEAE) (*1*). Light emission at a rate that is first order with respect to the concentration of aequorin takes place on addition specifically of calcium (or to less extent strontium) salts in slight excess over the molar equivalent of EDTA-2Na in the solvent. No other factor, among many tried, replaced the $Ca^{++}$ or $Sr^{++}$ in this reaction. Unlike most other bioluminescent systems (*2*), neither the