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# The Radioactivity of the Earth's Crust and Its Influence on Cosmic-Ray Electroscope Observations Made Near Ground Level

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Radium analyses of 23 rock specimens from the scene of cosmic-ray observations made by R. A. Millikan and his co-workers discloses a linear relationship between the average radium content of the terrain and the local gamma-ray ionization in the cosmic-ray electroscope. Ionization due to soft gamma-radiation stimulated in the earth's crust by the impact of cosmic rays, or due to the reflection of cosmic rays by the earth's crust is therefore negligible, and the ratio of thorium to uranium is approximately constant in the materials examined. A local gamma-ray ionization of 5.5 ion pairs per sec. per cm³, in air at one atmosphere pressure, is normally associated with a content of 1.0 $\times 10^{-12}$ g Ra per g rock in the neighboring terrain. It is shown that the gamma-ray ionization $n$ ion pairs per cm³ per sec., at $h$ cm above a broad radioactive surface is $n=(2\pi\rho mK/\mu_1)G_{\mu h}$, where $G_{\mu h}\equiv\int_1^\infty z^{-2}e^{-\mu h z}dz$, $\mu_1/\rho$ is the mass absorption coefficient and $m$ the Ra concentration of the emitter, $\mu$ is the gamma-ray absorption coefficient of air, and $K$ is Eve's number. Application to the experimental data suggests that the Th/U ratio is the order of 5 to 10 for many common geological materials.

## INTRODUCTION

THE extra-terrestrial origin of the penetrating radiation now called cosmic rays was first demonstrated by Gockel,[1] who discovered that the residual ionization in a sealed electroscope at first decreased but then increased markedly as the instrument was raised away from the earth's surface. The initial decrease shows that a part of the residual ionization is of terrestrial origin. Whereas the bulk of this terrestrial radiation was believed to be due to gamma-radiation from radioactive elements in the earth's crust, Millikan[2] suggested that experiments were needed to demonstrate whether an appreciable fraction of this radiation was due to soft secondary rays stimulated by the cosmic rays or to cosmic rays reflected or scattered back by the earth's crust. That practically all of the terrestrial radiation is due to gamma-rays from rocks in the earth's crust has now been demonstrated by ground level and airplane comparisons of the ionization in unshielded electroscopes with the ionization in similar electroscopes shielded only from below by lead.[3] Further evidence is furnished by the cloud-chamber photographs of Anderson[4] and Blackett and Occhialini,[5] which show a pronounced distribution maximum for cosmic-ray tracks entering from vertically overhead, and also show very few tracks entering the apparatus from below. The present observations also support the conclusion that all of the terrestrial radiation is due to gamma-rays from the earth's crust, by corre-

* Experimental work done as National Research Fellow, University of California, Berkeley, 1932–1934.

[1] Gockel, Physik. Zeits. **11**, 280 (1910); Hess, Physik. Zeits. **12**, 998 (1911); **13**, 1084 (1912); Kolhörster, Physik. Zeits. **14**, 1153 (1913); Millikan and Bowen, Phys. Rev. **27**, 353 (1926).

[2] Millikan and Cameron, Phys. Rev. **28**, 851 (1926).

[3] Anderson, Millikan, Neddermeyer and Pickering, Phys. Rev. **45**, 352 (1934).

[4] Anderson, Phys. Rev. **44**, 406 (1933).

[5] Blackett and Occhialini, Proc. Roy. Soc. **A139**, 699 (1933).

TABLE I. *Description and geographical origin of the samples, local gamma-radiation in ion pairs per cm³ per sec. at 30 atmospheres of air, and radium content of the geological samples in g Ra per g of specimen.*

| DESCRIPTION | SPECIMEN NUMBER | LOCAL GAMMA-RADIATION | RADIUM CONTENT |
|---|---|---|---|
| Gravel. Top of Pike's Peak, Colorado. | C1 | 97 | $0.41\times10^{-12}$ |
| Gravel. Roadway on Mt. Manitou, Colorado. | C2 | 97 | 0.25 |
| Deep glacial rubble. Churchill, Manitoba. | C3 | 32 | 0.33 |
| Glacial sand. Churchill, Manitoba. | C4 | 32 | 0.44 |
| Gravel. Windy Point, Colorado. | C5 | 98 | 0.52 |
| Granite. Churchill, Manitoba. | C6 | 12 | 0.23 |
| Red Granite. Pike's Peak, Colorado. | C7 | 97 | 1.52 |
| Hard granite outcrop. 30 ft. stone in Red River, Winnipeg, Manitoba. | C8 | 45 | 0.19 |
| Granite. Mt. Manitou, Colorado. | C9 | 97 | 1.34 |
| Gravel. Superior pits, Minneapolis, Minn. | C10 | 32 | 0.48 |
| Limestone. Quarry in Minneapolis, Minn. | C11 | 34 | 0.60 |
| Quartzose sandstone. Zion Canyon, Utah. | C12 | 25 | 0.32 |
| Granite boulder. Glen Cove, Colorado. | C13 | 135 | 1.83 |
| Soil. Ellendale, North Dakota. | C14 | 42 | 0.78 |
| Soil. Felts Field, Spokane, Washington. | C15 | 47 | 0.68 |
| Limestone. Cormorant Lake, Manitoba. | C16 | 6 | 0.11 |
| Limestone. Riverside, California. | C17 | 14.5 | 0.32 |
| Kaibab limestone. Rocky point with railing, North Rim, Grand Canyon, Colorado. | C18 | 24 | 0.95 |
| Acidic lava. Arequipa, Peru. | C19 | 148 | 1.75 |
| Rhyolitic lava. Crucero Alto, Peru. | C20 | 60 | 2.38 |
| Quartzite. Crucero Alto, Peru. | C21 | | 0.20 |
| Limestone. Cormorant Lake, Manitoba. | C22 | 6 | 0.12 |
| Quartz-mica schist. Inside railroad tunnel, San Gabriel Canyon, Cal. | C23 | 73 | 0.20 |

lating the measured radium content of certain rocks with the soft gamma-radiation observed in a cosmic-ray electroscope operated on or near these rocks.

## DESCRIPTION OF SPECIMENS

All of the rock specimens were furnished by R. A. Millikan and his co-workers, and represent samples of the rocks or soil underlying the electroscope at the scene of the cosmic-ray observations which they have made during the past four years. A brief description of the specimens and the location of the stations is given in Table I.

## LOCAL GAMMA-RADIATION *vs.* RADIUM CONTENT

The intensity of the local gamma-radiation at each of these sites was determined by comparing the discharge rates for the unshielded electroscope with the rate when the electroscope was completely surrounded by a lead shield 7.64 to 10 cm thick. Corrections were of course made[6] for the fact that the lead shield absorbs some (24 to 39 percent, depending on altitude) of the cosmic radiation while absorbing practically all (97.6 percent) of the local gamma-radiation.

The field specimens of local geological material were ground to about 60 mesh and 3 to 5 g samples analyzed for radium by the emanation method, employing the direct-fusion furnace and double ionization-chamber technique.[7] Two or more duplicate analyses were made on nearly every specimen, and individual values seldom varied from the mean value by as much as $0.05\times10^{-12}$ g Ra per g. Sample variations of this magnitude are to be expected on statistical grounds because of the local concentration of radium in the grains of certain heavy mineral constituents of the samples. As is to be expected, these sample variations are considerably smaller in the fine grained materials than in coarse grained igneous or conglomerate samples. Those specimens which are free from limestone and opal were analyzed by Raitt with Evans' earlier apparatus[8] at the California Institute of Technology. These two independent sets of data are in splendid agreement as is shown by Fig. 1.

Table I presents the local gamma-ray ionization found by Millikan and his co-workers as observed in, or else reduced to, a 15 cm diameter steel electroscope filled with air at 30 atmospheres pressure. The average radium content of the field specimen is given in g Ra per g of material.

[6] Millikan and Cameron, Phys. Rev. **28**, 851 (1926); **37**, 235 (1931).

[7] Evans, Rev. Sci. Inst. **6**, 99 (1935).

[8] Evans, Rev. Sci. Inst. **4**, 223 (1933).

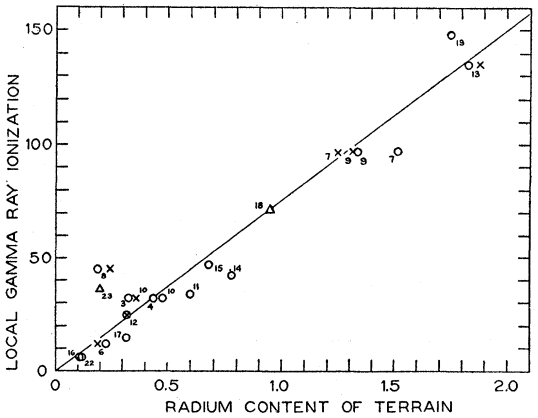


FIG. 1. Ionization due to soft local radiation *vs.* radium content of specimens of the surrounding rocks or soil. Ionization is in ion pairs collected per sec. per $cm^3$ in air at 30 atmospheres. Radium content is in $10^{-12}$ g Ra per g material. Analyses marked ○ or △ are by Evans, × by Raitt.

The local gamma-ray ionization is due to the combined effect of all the gamma-emitters in the uranium, actinium and thorium series. The thorium content of ordinary rocks is usually several times greater than the uranium content, and hence the thorium series gamma-rays are at least as strong as those from the uranium series. McCoy and Henderson[9] found 1 g Ra to be the gamma-ray equivalent of $6.85\times10^6$ g Th, hence 1 g U and its decay products is slightly more active than 2.3 g Th and its decay products in the production of gamma-ray ionization. The radium content of a rock body is proportional to the total uranium series gamma-rays emitted by that body. But in a series of rock bodies the radium content can only be proportional to the *total* gamma-radiation if the ratio of the thorium to uranium content is constant. The close correlation here found between the total gamma-radiation and the radium content, shown in Fig. 1, indicates that in at least 15 of the samples in this series the thorium-uranium ratio does not vary widely.

Fig. 1 omits four of the specimens found in Table I for the following reasons. Samples C1, C2 and C5 are gravels from the Pike's Peak area, whose radium content shows no agreement with the local gamma-radiation at these stations. This fact prompted inquiries which confirmed the radioactive evidence that these materials are not really indigenous to their areas, but are thin surface layers which were hauled into their present site during road building operations. At Crucero Alto, the rocks in the neighborhood of the cosmic-ray station are a mixture of C20 and C21, mostly the latter. The local gamma-radiation corresponds to a mixture of 73 percent C21 and 27 percent C20, which is in agreement with the field notes on the terrain, but cannot be readily plotted in Fig. 1.

In all but two cases, the electroscope stood about a foot or two above the surface of a rather flat area, hence subtended on earthward solid angle of nearly $2\pi$. For C18, however, the electroscope was on a narrow rocky point jutting out from the rim of the Grand Canyon, and subtended a solid angle of only about $2\pi/3$ steradians, and for C23 the electroscope was inside a mountain tunnel and subtended nearly $4\pi$ steradians. In plotting Fig. 1, triangular points are shown for these two stations to indicate that the observed local radiation has been corrected for solid angle.

The local radiation for C23 and C8 is about 2.5 to 3 times the value expected from the radium content if the thorium-uranium ratio is the same as for the other samples in the series. This suggests that these two specimens are either not truly representative of the average rock in their vicinity or else that they have a higher thorium-uranium ratio than the other members of the series.

Employing the newly developed technique[10] for counting alpha-rays emitted from the surface of rocks, Dr. Gladys Finney made a preliminary estimate of the Th/U ratio for C8, 11, 18 and 23. The numerical values depend upon the alpha-ray stopping power of the rock, which was inferred from the known radii of plechroic halos in various minerals. Under such assumptions the Th/U ratios obtained from measurements on flat polished surfaces ground onto the solid specimens indicate that C11 and C18 have the same Th/U ratio, while C8 has 2.8 times and C23 has 3.3 times as high a Th/U ratio as C11 and C18. The high Th/U ratios thus indicated for C8 and C23 bring their local gamma-radiation into agreement

[9] McCoy and Henderson, J. Am. Chem. Soc. **40**, 1316 (1918).

[10] Evans, Finney, Kip and Mugele, Phys. Rev. **47**, 791 (1935).

TABLE II. *Average radium content of the classified samples.*

| NUMBER OF SPECIMENS | CLASSIFICATION | G RADIUM PER G SPECIMEN LIMITS | AVERAGE |
|---|---|---|---|
| 1 | Quartz-mica schist | $0.20\times10^{-12}$ | $0.20\times10^{-12}$ |
| 1 | Quartzite | 0.20 | 0.20 |
| 4 | Limestones | 0.11 to 0.60 | 0.29 |
| 1 | Sandstone | 0.32 | 0.32 |
| 2 | Glacial sand and rubble | 0.33 to 0.44 | 0.38 |
| 4 | Gravels | 0.25 to 0.52 | 0.41 |
| 2 | Soils | 0.68 to 0.78 | 0.73 |
| 1 | Kaibab limestone | 0.97 | 0.97 |
| 5 | Granites | 0.19 to 1.83 | 1.02 |
| 2 | Peruvian Lavas | 1.75 to 2.38 | 2.06 |
| Total 23 | All specimens | 0.11 to 2.38 | $0.70\times10^{-12}$ |

with that of the other specimens. It must be emphasized, however, that this alpha-ray method for Th is still in a state of development and that absolute Th values are uncertain, although there can be no doubt that the relative values for Th/U given above are essentially correct. No field specimen in this series was large enough to permit confirming these Th/U values by the $\gamma$-ray methods recently developed by Evans and Mugele.[11]

The radium content of the sample of Kaibab limestone C18 is much higher than for most limestones, but is certainly correct as it was checked by a variety of alternative analytical techniques, employing both solution and direct-fusion methods. This result is in accord with the rather large contamination, by sand and other silicates, noticeable in the Kaibab formation.

There are six simultaneous criteria for a linear relationship between the local gamma-radiation from the earth and the radium content of a small sample of rock or earth, taken near the cosmic-ray electroscope.

(1) The sample is truly a representative specimen of all the neighboring and underlying terrain.
(2) The ratio of the thorium to the uranium content is the same in each specimen of the series.
(3) Radioactive equilibrium exists between thorium, uranium, actinouranium and their individual decay products.
(4) No new strong gamma-radioactive element is present in the rocks which is not a member of one of the three known radioactive families.
(5) No appreciable gamma-radiation is produced in the rocks by cosmic-ray bombardment.

[11] Evans and Mugele, Phys. Rev. **47**, 427 (1935).

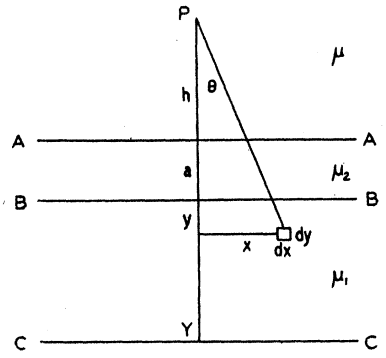


FIG. 2. Geometry for Eqs. (1) to (4).

(6) No appreciable fraction of the cosmic rays incident on the earth's crust are reflected or scattered back toward celestial space.

Criteria 4, 5 and 6 may be regarded as established by the data of Fig. 1 and Table I. Criterion 2 is obeyed by most of the specimens, and the constancy of the Th/U ratio thus disclosed is of considerable geochemical interest. The ratio is not sufficiently constant, however, to lend strong support to belief in a generic relationship between the uranium and thorium families, moreover such a belief is embarrassed by the existence of radioactive ores bearing either thorium or uranium exclusively.

The cosmic-ray electroscope is here shown to be a geophysical instrument which is useful for rapid radioactive surveying, even where no radioactive ore bodies are involved.

Bearing in mind that the number of specimens in each petrographic classification is small, and that therefore the average values cannot be regarded as universally applicable, it is of interest to compute the average radium content of each class of material which entered the present investigation. These averages are shown in Table II.

## THEORY OF GAMMA-RAY IONIZATION FROM LARGE RADIOACTIVE BODIES

The gamma-ray ionization above a flat radioactive solid such as a region of the earth's surface may be computed as follows. For greater generality, consider the ionization $n$ produced at the point $P$, Fig. 2, located $h$ cm above the plane surface $AA$ of a nonradioactive absorbing layer $a$

cm thick, which overlies the radioactive solid $BB-CC$. Let the thickness of this radioactive material be $Y$ cm, its density $\rho_1$, g cm$^{-3}$, its Ra content $m$ g Ra per g, and its effective absorption coefficient $\mu_1$ cm$^{-1}$. Let $\rho_2$ and $\mu_2$ be the density and absorption coefficient of the absorber, and $\mu$ the absorption coefficient of the medium above $AA$ (air in the present application).

The ionization $dn_1$ ion pairs per cm$^3$ per sec. at $P$ due to the Ra in the volume element $2\pi x dx dy$ formed by rotating $dxdy$ about the vertical $y$ axis is:

$$dn_1=\frac{2\pi x dx dy\rho mK}{(h+a+y)^2+x^2}e^{-\mu_1 y/\cos\vartheta}e^{-\mu_2 a/\cos\vartheta}e^{-\mu h/\cos\vartheta}, \tag{1}$$

where $K$ is a proportionality constant.

We now change variables by substituting $z=1/\cos\vartheta$, and then integrate over the region $1\leqslant z\leqslant\infty$ which is equivalent to $0\leqslant\vartheta\leqslant\pi/2$. From the geometry of Fig. 2, $x=(h+a+y)\tan\vartheta$, and by substitution of the new variable $z$ we find:

$$n_1=2\pi\rho mK\int_1^\infty z^{-1}e^{-(\mu_2 a+\mu h)z}dz\int_0^Y e^{-\mu_1 yz}dy \tag{2}$$

$$=(2\pi\rho mK/\mu_1)\int_1^\infty\{z^{-2}e^{-(\mu_2 a+\mu h)z}-z^{-2}e^{-(\mu_1 Y+\mu_2 a+\mu h)z}\}dz. \tag{3}$$

A table of the integral $G_b=\int_1^\infty z^{-2}e^{-bz}dz$ was given by Gold,[11a] whose values are shown in Fig. 3. From Eq. (3) and Fig. 3 one may compute the gamma-ray ionization for the general case, such as for specimens C1, C2 and C5 where the absorbing layer is also radioactive.

In the special case which applies to most of the specimens, $a=0$ and $Y=\infty$. We therefore have:

$$n=(2\pi\rho mK/\mu_1)G_{\mu h}. \tag{4}$$

Eve's number[12] $K$ is evaluated by noticing that for any radiation which is exponentially absorbed the ionization $n$ ion pairs per cm$^3$ per sec. at a distance $r$ cm from a source of unit strength will be $n=Ke^{-\mu r}/r^2$ in a medium whose absorption coefficient is $\mu$ cm$^{-1}$. The total ionization produced by such a unit source will be

$$N=\int_0^\infty 4\pi r^2 n dr=4\pi K\int_0^\infty e^{-\mu r}dr$$

$$=4\pi K/\mu \text{ ion pairs per sec.}$$

Eve's number $K$ depends upon temperature pressure and ionization chamber wall materials, and is not accurately known, but is approximately $60/4\pi\times10^9$ ion pairs per sec. per cm for the gamma-rays from Ra $(B+C)$ in equilibrium with 1 g of Ra. (For $\mu N$ in air Eve gives $50\times10^9$; Hess 61 and $72\times10^9$; Moseley and Robinson $73\times10^9$. Reitz gives $K=4.3\times10^9$ for RaC in free air at 0° and 760 mm, or up to $6.8\times10^9$ in an iron ionization chamber with 3 mm thick walls.)[13]

The mass absorption coefficient $\mu_1/\rho$ is not accurately known for geological specimens but has a value somewhere near or between 0.034 (McClelland[14]) and 0.065 cm$^2$ g$^{-1}$ (Behounek[15]). Since $\mu$ is the order of $6\times10^{-5}$ cm$^{-1}$ for air, any observations made within a few meters of the earth's surface will correspond to $\mu h\to0$, hence $G_{\mu h}\to1$. If we substitute $G=1$, $\mu_1/\rho=0.06$, $K=6.8$

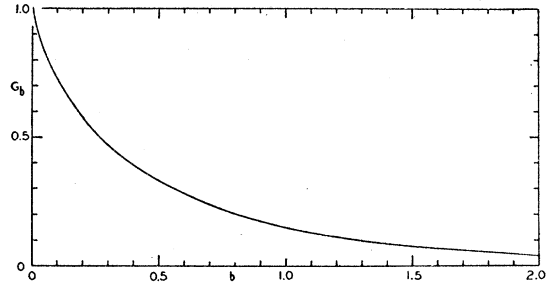


FIG. 3. Values of the integral $G_b=\int_1^\infty z^{-2}e^{-bz}dz$ for $0\leq b\leq2$. For larger $b$, some values are:

| $b$ | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|
| $G_b$ | 0.03754 | 0.01064 | 0.00320 | 0.00099 | 0.00032 |

The series $G_b=e^{-b}(1/b-2/b^2+3!/b^3-4!/b^4+\cdots)$ represents the integral asymptotically.

[11a] E. Gold, Proc. Roy. Soc. **A82**, 43 (1909).

[12] Eve, Phil. Mag. **22**, 551 (1911); **27**, 394 (1914); Rutherford, Chadwick and Ellis, *Radiations from Radioactive Substances* (1930), p. 496.

[13] A. S. Eve, Phil. Mag. **12**, 189 (1906); **22**, 551 (1911); **27**, 394 (1914); V. F. Hess, Akad. Wiss. Wien, Ber [IIa] **122**, 1053 (1913); H. G. J. Moseley and H. Robinson, Phil. Mag. **28**, 327 (1914); A. W. Reitz, Zeits. f. Physik **69**, 259 (1931).

[14] Eve, Phil. Mag. **21**, 26 (1911).

[15] Behounek, Zeits. f. Physik **79**, 590 (1932).

$\times 10^9$, and $m = 1 \times 10^{-12}$ g Ra per g in Eq. (4) we obtain $n = 0.7$ ion pairs per cm$^3$ per sec., for the ionization near the ground[16] due to the penetrating gamma-rays of RaC from material containing $10^{-12}$ g Ra per g. The softer gamma-rays from higher members of the uranium series will increase this value to about unity, but accurate data are not available.

This theoretical ionization value is to be compared with the observations plotted in Fig. 1. Incomplete saturation[17] of the ion current permits only 13.8 times[18] as many ions to be collected in air at 30 atmospheres as at 1 atmosphere pressure. Consequently, the data of Fig. 1 show that the radioactive content of average terrestrial materials, be they granites, limestones, sandstones, rubble or soil, will cause a gamma-ray ionization[19] of about 5.5 ion pairs per sec. per cm$^3$ in air at a pressure of one atmosphere when the terrain contains $1.0 \times 10^{-12}$ g Ra per g.

Recalling[9] that the gamma-rays from 2.3 g Th and its decay products are nearly as effective as those from 1 g U and its decay products, it is readily seen that the Th/U ratio must be between 5 and 10 in order to account for the 5.5 ion pairs per cm$^3$ per sec. observed in the cosmic-ray electroscope. More accurate knowledge of $K$ and $\mu_1/\rho$ may alter this figure somewhat, but there is here clear support for the view that the Th/U ratio is considerably greater than the value 2.5 which has been used by Holmes and others.[20]

Entirely independent measurements on another suite of rock samples[10] has confirmed this high Th/U ratio. This conclusion has considerable geophysical importance because it shows that the thorium series is more important than the uranium series in such questions as geological age measurements based on helium ratios in common materials, and in geothermal problems concerning earth movements and the earth's internal heat.

## Acknowledgments

This research was originally initiated by R. A. Millikan at the California Institute of Technology, where it followed the carbonate-fusion investigations by J. Lloyd Bohn[21] and E. M. McMillan,[22] and was a part of Professor Millikan's cosmic-ray research program, supported by a grant from the Carnegie Corporation of New York. Completion of the experimental phases awaited the recent final development of the direct-fusion furnace technique[23] in the laboratories of the University of California, where the able assistance of Mr. Fred Ludecke and Mr. Arthur Kip was of great value.

[16] The variation of ionization with altitude is given by Eq. (4) and Fig. 3. Thus at 200 meters above ground level $\mu h = ca$. 1.3 and the ionization will amount to about 10 percent of its value at the earth's surface. Similar considerations make it possible to compute the terrestrial gamma-ray corrections for balloon and airplane observations.

[17] I. S. Bowen, Phys. Rev. **41**, 24 (1932).

[18] R. A. Millikan, Phys. Rev. **39**, 397 (1932).

[19] This value, of course, corresponds to measurements made in a particular detecting instrument. Secondary and transition effects due to the walls of the instrument cause the observed value to differ somewhat from the corresponding ionization in free air; see A. W. Reitz, Zeits. f. Physik **69**, 259 (1931) and E. J. Workman, Phys. Rev. **43**, 859 (1933). Moreover, in free air near land areas there will be the order of 1 ion pair per cm$^3$ per sec. of additional ionization due to alpha-rays from Rn, Tn and their decay products in the air itself.

[20] A. Holmes, *Radioactivity and Earth Movements*, Trans. Geol. Soc. Glasgow **18**, III, 559 (1928–29; published 1931).

[21] Bohn, Ph.D. Thesis, California Institute, (1927); J. Frank. Inst. **210**, 461 (1930).

[22] McMillan, M.S. Thesis, California Institute (1928).

[23] Evans, Rev. Sci. Inst. **6**, 99 (1935).